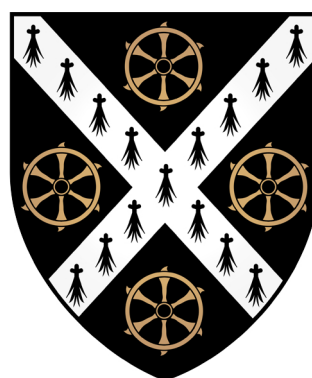


Hybrid Circuit QED with Spin Ensembles and Carbon-Nanotube-Based Superconducting Qubits



Matthias Mergenthaler
St. Catherine's College
University of Oxford

Supervisors:
Dr. Edward A. Laird
Dr. Peter J. Leek
Prof. G. Andrew D. Briggs

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To my parents.

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Abstract

Circuit quantum electrodynamics has received an increasing amount of attention over the last decade in light of its potential use in quantum information processing. The underlying principle of this architecture is the coherent coupling between microwave photons stored in superconducting resonators and superconducting qubits made from non-linear, non-dissipative electrical circuits. This architecture can be extended to other solid-state systems with transition frequencies in the microwave regime, such as nuclear and electron spins or Andreev bound states. These hybrid devices would allow the coherent exchange of information between such systems and superconducting qubits mediated by a microwave resonator. This could lead to advances in quantum information processing and new insights in the physics governing these interacting systems.

In this dissertation experimental work with different types of hybrid quantum circuits is presented. A first experiment investigates the coherent coupling of microwave photons with a spin ensemble in a correlated and uncorrelated state. The coupling strength and magnetic resonance is studied as a function of temperature and magnetic field direction. A pronounced temperature dependent anisotropy of the magnetic resonance is observed, which can be attributed to the onset of antiferromagnetic correlations in the spin ensemble when cooled below $T = 4$ K. A simple one-dimensional model gives a good description of this anisotropy.

A next experiment studies the transport characteristics of carbon nanotubes electrically accessed with normal and superconducting contacts. These devices were characterised using both DC and RF reflectometry techniques. The carbon nanotube devices with superconducting contacts exhibit transport characteristics of Josephson junctions with critical currents of up to $I_c \approx 18$ nA, as well as multiple Andreev reflections. These semiconductor-superconductor hybrid Josephson junctions are then used to realise a carbon-nanotube-based superconducting qubit with voltage tunable transition frequency using a local electrostatic gate. Strong coupling ($g \sim 100$ MHz) to a coplanar waveguide resonator is demonstrated via a resonator frequency shift dependent on applied gate voltage. Qubit parameters are extracted from spectroscopy and correspond well to the DC measurements of the carbon nanotube Josephson junctions. Qubit relaxation and coherence times in the range 10 – 100 ns are observed.

The hybrid devices investigated in this work present potential building blocks for more extensive hybrid architectures for quantum information processing.

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Part I

Basic Concepts

1

Introduction

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Classical information processing is an indispensable part of our daily lives today and without a doubt one of the greatest technological achievements of the last century. It enables modern communication over the Internet, an optimised exchange of information and more efficient processing of data. This "digital revolution", which affects almost all areas of life, was mainly possible due to the invention of the transistor in 1947 [1] and the following steady gain in performance of the microchips installed in all of our digital devices.

The underlying, most basic component of all microchips to date is the transistor. Microchips with ever-increasing transistor density have accelerated computing power exponentially over the past fifty years. This growth in computing power followed the so-called Moore's Law, which states that the number of transistors on a microchip doubles regularly [2]. However, this law possesses a natural limit, which is reached when, by the effort of steady downsizing, the size of the transistors reaches atomic dimensions. The idea of *quantum information processing* could

provide a radical new computational paradigm that takes computing to a new level beyond this natural limit of Moore's law.

1.1 Quantum Information Processing

In conventional computers, information is stored, manipulated and transferred in a binary fashion [3]. The idea of using the laws of quantum mechanics for information processing moves beyond this binary approach and was first mentioned by Richard Feynman in 1982 [4]. He concluded that a computer purely based on classical physics is unable to simulate quantum mechanical systems efficiently. Hence, he proposed to use a network of quantum mechanical systems to perform such simulations – this marks the birth of the idea of a quantum computer. Feynman's idea quickly gained momentum and in 1985 David Deutsch presented the more general idea of a *universal quantum computer*, similar to a universal Turing machine [5]. His work included a first example of a quantum algorithm, the Deutsch algorithm, which is fundamentally based on the principle of superposition and offers a speed-up compared to the classical analogue. This stimulated the development of several quantum algorithms, which all provide a computational speed-up with regard to their classical counterparts. Very prominent examples are the Deutsch-Josza algorithm [6], a generalisation of the Deutsch algorithm and Grover's algorithm [7], giving a quadratic speed-up in searching unstructured databases compared to the best known classical algorithm. However, the most famous algorithm was invented by Peter Shor in 1994 which gave a solution for factoring large numbers in polynomial time [8]. Once a universal quantum computer becomes available, Shor's algorithm poses as a potential threat to almost all of our modern security and encryption systems. However, integrating different notions of quantum mechanics into cryptographic protocols fortunately reveals new concepts for secure communication. These new insights into how fast and efficient a quantum computer could solve certain tasks led to an international race between academic researchers as well as private companies to build an efficient and scalable universal quantum computer.

The basic building block of any quantum computer is the *quantum bit* or *qubit*. The quantum mechanical two-level system that constitutes the qubit has a ground state, labelled as $|0\rangle$, and an excited state with higher energy, labelled $|1\rangle$. Formally any superposition state of the qubit $|\psi\rangle$ is made up of these two classically allowed states and given by $|\psi\rangle = a|0\rangle + b|1\rangle$ where a and b are complex numbers with $|a|^2 + |b|^2 = 1$. Composite systems of two or more qubits give rise to one of the most subtle properties of quantum mechanics. Some of these systems can be factored into product states and some cannot. For a system of two qubits that are in the state $|\psi_{21}\rangle = \frac{1}{\sqrt{2}}(|00\rangle + |11\rangle)$, there exist no two individual product states $|\psi_1\rangle$ and $|\psi_2\rangle$, such that $|\psi_{21}\rangle = |\psi_2\rangle \otimes |\psi_1\rangle$. Such a system is said to be *entangled* and all the information resides in correlations between the two qubits, which was mentioned by Einstein, Podolsky and Rosen [9].

In principle, any quasi two-level system governed by quantum mechanics can be called a qubit. However, for a qubit to be useful for quantum information processing it needs to be well isolated from its environment but at the same time precisely controllable. Hence in order to realise a quantum computer the qubit and the control and coupling architecture around it need to fulfil specific requirements – the so-called five DiVincenzo criteria [10]:

1. A scalable architecture with well defined qubits.
2. Distinct and reliable qubit state preparation.
3. Decoherence times much longer than gate operation times.
4. A universal set of quantum gates that perform accurate operations on the qubits.
5. Well defined readout capability for each individual qubit.

Due to the huge scientific interest in quantum information processing over the last two decades a wide variety of physical systems have been proposed and realised for the implementation of quantum computing architectures. Some of the more promising systems include trapped ions [11, 12], neutral atoms [13], electron or

nuclear spins [14–17] and superconducting (charge, phase or flux) qubits [18–23]. Despite each of these systems being in different stages of their development, none of them have a striking advantage over the others. In contrast, all of the different architectures offer particular advantages and disadvantages, such as longer coherence times, easier control of individual qubits or better scalability. Therefore, an idea explored by various groups around the world is to create and study hybrid quantum devices, which combine the advantages of two or more quantum mechanical systems for quantum information processing [24–27].

1.2 Hybrid Quantum Circuits

The quest for the optimal architecture of a universal quantum computer led to a long list of proposed and realised hybrid quantum devices. This work is focussed on hybrid quantum circuits which are particularly interesting for quantum information processing due to their straight forward interface with superconducting qubit architectures [26, 28]. The basic building block of all hybrid quantum circuit implementations is a superconducting microwave resonator. Early experiments combined these with spin ensembles such as nitrogen-vacancy centres, showing coupling to the photons [29–32], microwave photon storage and retrieval [33, 34] as well as coupling to a superconducting qubit mediated by the microwave resonator [35, 36]. These hybrid circuits essentially showcase the path towards a quantum memory based on highly coherent nuclear spins with superconducting qubits as the processing unit [37]. One caveat of these implementations is however that most spin ensembles exhibit only a small to no zero-field splitting, and hence an external magnetic field is necessary which is unfavourable for superconducting qubits.

Due to the continuous progress in nanofabrication techniques, a wide variety of nanoelectronic and mesoscopic devices which exhibit quantum behaviour can be engineered and fabricated. Strong confinement of charge carriers in these type of devices allows for the implementation of charge- or spin-based qubits in quantum dots [17] or impurity spins in silicon [38]. These could also be used as the processing unit in a quantum computing architecture and easily interfaced

with microwave resonators. Multiple implementations of such devices have already been presented [39–49]. These offer the benefit of all electric control compared to superconducting qubits, but charge qubits suffer from short coherence times and most implementations of spin qubits still rely on local magnetic fields. A more recent advance in hybrid quantum circuits using such nanoelectronic devices, is to build novel type of superconducting qubits with the main element, the Josephson junction replaced by semiconductor-superconductor hybrids [50–52]. These type of qubits also offer all-electric control and do not require magnetic fields for their operation. Another type of hybrid circuit is the combination of nanomechanical resonators and superconducting circuits. These can have an impact on quantum information processing through novel types of directional signal amplifiers [53, 54], on-chip circulators [55] or possible frequency conversion for long range optical signal transmission [56].

Hybrid quantum circuits envision a combination of all the best components in one large architecture for quantum computing, where each building block is utilised for the task it performs best at. More in depth summaries of the different types of hybrid quantum circuits can found in References [25, 26, 28, 57, 58].

1.3 Thesis Synopsis

The work presented here touches on three different types of hybrid quantum devices, namely a correlated spin ensemble coupled to a superconducting microwave resonator, carbon nanotube Josephson junctions and a superconducting qubit based on a carbon nanotube. In these devices the common building block is the superconducting material/circuit. **Chapter 2** will cover the underlying theory of circuit quantum electrodynamics and give a brief introduction to the individual building blocks. Additionally, **Chapter 3** contains a brief description of carbon nanotubes and single quantum dots, especially for their behaviour when contacted with superconducting leads. **Chapter 4** summarises the necessary sample fabrication procedures as well as the individual design parameters for two different types of microwave resonators and superconducting qubits. The experimental setup

used for all the measurements presented here is described in **Chapter 5**, alongside the experimental methods employed. **Chapter 6** presents a detailed study of electron spin resonance of a correlated spin ensemble employing the architecture of circuit quantum electrodynamics. The coupling between microwave photons and the ensemble excitations as well as the magnetic resonance are analysed as a function of temperature, external field strength and field direction. The results show a transition of the ensemble from an uncorrelated to the onset of a correlated state. The results in this chapter were previously published in Reference [59] and the content is based on this publication. In **Chapter 7** the transport characteristics of carbon nanotubes with normal and superconducting leads are investigated. An emphasis is put on the Josephson junction characteristics of the devices with superconducting leads, i.e. their critical current and resulting Josephson energy. The results are analysed in terms of feasibility to realise a superconducting qubit based on these carbon nanotube Josephson junctions. Following this, **Chapter 8** introduces a first implementation of such a carbon-nanotube-based superconducting qubit. The gate voltage dependent frequency tunability is investigated and exhibits vanishing qubit transitions up to qubit transition frequencies of ~ 4 GHz. Qubit spectroscopy at different qubit drive powers allows the extraction of the vital qubit and Josephson junction parameters. The results are very similar to parameters measured in Chapter 7. Additionally, the relaxation and coherence times of two qubit devices are measured at various qubit transition frequencies. Furthermore, potential signs of Rabi oscillations are observed. Finally, all of the results are summarised in **Chapter 9** and an outlook on potential future experiments is given.

2

Circuit QED

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In this thesis, the quantum coherent interaction between solid-state qubits, in the form of spins and superconducting qubits, and the electromagnetic field in microwave circuits is studied. These systems are described with the physics of *Circuit Quantum Electrodynamics* (circuit QED), the electrical circuit equivalent to *Cavity Quantum Electrodynamics* (cavity QED). Cavity QED was pioneered by *S. Haroche* and his collaborators in the 1980s [60], and recognised with the Nobel Prize in Physics in

2012. In this chapter the concept of light matter interaction is introduced based on the fundamental field of study cavity QED. Further, the basic concepts of circuit QED and the different building blocks used within this thesis are explained in detail.

2.1 Light-Matter Coupling

The interaction of light and matter has always been of fundamental interest in physics research. One of its most elementary concepts is the interaction between one photon and a single two-level system, i.e. an atom. In free space however, an atom is coupled to an infinite number of electromagnetic field modes [60, 61]. Inside a cavity the spectrum of the electromagnetic field is strongly modified and depending on the geometry only a single electromagnetic mode can propagate. While the interaction between an atom and the electromagnetic field in free space is typically weak, it is enhanced in a cavity due to the confinement of the field. Hence, a single atom inside such a cavity enables the study of the interaction of this atom with a single mode of the electromagnetic field and more importantly with a single photon. The area of research of these two coupled quantum systems is the aforementioned cavity QED.

The continuous spectrum of the electromagnetic field in free space causes the excited state population of an atom to decay irreversibly. However, according to Fermi's Golden Rule the atomic decay rate depends on the density of electromagnetic states, which can be considerably modified by confining the electromagnetic field to an optical cavity [62, 63]. The cavity is created by two mirrors separated by a distance d , which will only allow discrete modes to resonate. An atom placed inside this cavity will most likely interact strongly with only one of the allowed modes, see Figure 2.1. If the cavity is on resonance with the decay mode, the interaction enhances the radiative decay of the atom due to the Purcell effect [64]. On resonance, the distinct modes of the cavity and the fast radiative decay of the atom lead to a coherent exchange of a photon between the atom and the cavity [61].

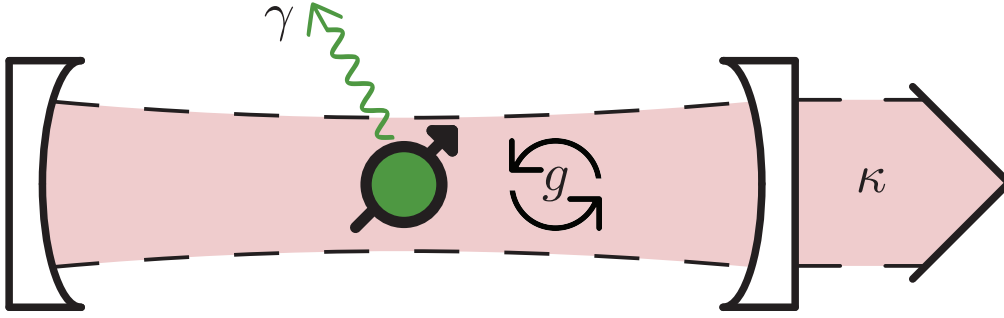


Figure 2.1: Schematic of a cavity QED system where a two-level system, e.g. an atom or single spin, is placed inside a cavity which supports a single electromagnetic mode. The electromagnetic field can leak out of the cavity with rate κ and the two-level system can relax from its excited state with rate γ . The two systems are coupled with the coupling rate g .

2.1.1 Regimes of Coupling

The three figures of merit in cavity QED are the atomic decay rate γ , the cavity decay rate κ and the coupling rate g (sometimes referred to as coupling strength), which quantifies the rate of energy exchange between the field and the atomic transition, see Figure 2.1. The strength of the interaction between the atom and the electromagnetic field can be quantified by the cooperativity, a dimensionless parameter given by

$$C = \frac{g^2}{\kappa\gamma}. \quad (2.1)$$

Three distinct parameter settings for the coupling can be identified from the cooperativity [65], cf. Table 2.1. In the weak coupling regime the coupling rate is smaller than the decay rate of the atom and the decay rate of the photons from the cavity, i.e. $g \ll \kappa, \gamma$ and hence $C < 1$. Since here the decay rates are much faster than the rate of energy exchange between the atom and the cavity, no coherent

weak coupling	high cooperativity	strong coupling
$g \ll \kappa, \gamma$ No coherent effects.	$C > 1$ Coherent effects such as Purcell effect.	$g \gg \kappa, \gamma$ Coherent exchange of excitations. → Vacuum Rabi mode splitting

Table 2.1: Table of the three possible coupling regimes. The range of values for the three figures of merit g , κ and γ in each coupling regime are given. Additionally, it is described which effects are observable.

effects can be observed in the weak coupling regime. In the intermediate regime of high cooperativity, $C > 1$, at least one of the two relaxation parameters is smaller than the coupling rate. In this regime it is possible to observe coherent effects such as the Purcell effect [66]. The last and most studied regime is the strong coupling regime. Here the coupling rate is larger than the two decay rates, i.e. $g \gg \kappa, \gamma$. This allows for a coherent exchange of excitations between the two subsystems and the vacuum Rabi mode splitting is observable [67]. Additionally, this regime is also very important for quantum information processing, as it allows operations between two atoms, i.e. qubits, to be performed while preserving their coherence. This is vital for performing two-qubit gates with high fidelity and producing entangled quantum states [68].

Not only atoms have been used to study cavity QED, but also solid state systems such as self-assembled quantum dots [69]. These quantum dots were placed in a cavity with dimensions in the nm regime, making it possible to observe the strong coupling regime [70, 71].

2.1.2 Circuit Quantum Electrodynamics

Analogous to cavity QED, circuit QED studies the interaction of matter and light in solid state circuits, see Figure 2.2 for a schematic of a circuit QED implementation. Here a single microwave photon is stored in an on-chip cavity and coherently coupled to a superconducting qubit, which can be thought of as an 'artificial' atom [72]. The realisation of strong coupling between a single microwave photon and a superconducting qubit [73] has led to significant advances in the study and control of such systems in the framework of circuit QED. As a result, it was possible to demonstrate quantum optics experiments on a chip [74], as well as realise many important experiments related to quantum computing [75–82].

The superconducting qubits used in circuit QED are non-linear superconducting oscillators, where the nonlinearity arises due to the integration of a Josephson Junction [22, 83, 84] as a non-linear inductor in an LC resonator. This creates a discrete anharmonic potential, where the first two levels define an effective two-level

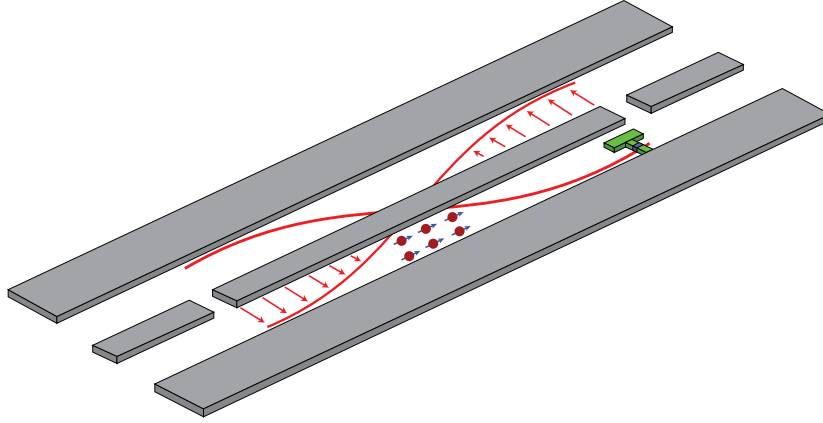


Figure 2.2: Schematic of a (hybrid) circuit QED implementation. The coplanar waveguide resonator is shown in grey. A superconducting qubit (green) is placed in the anti-node of the standing wave electric field of the fundamental mode of the resonator (red). For a hybrid circuit QED implementation spins (blue/red) are placed in the node of the electric field as here the magnetic field has an anti-node. More advanced hybrid circuit QED experiments exchange information between the qubit and the spins.

system, whose transition frequency can be designed to lie in the microwave regime (1 – 10 GHz). These qubits also exhibit a large dipole moment, which allows them to be strongly coupled to an electromagnetic field in the microwave regime. Therefore, on-chip electrical circuits can be used as a cavity [72] but it is also possible to probe superconducting qubits with three-dimensional cavities [85].

2.1.3 Hybrid Circuit Quantum Electrodynamics

Since the energy level spacing of most solid state qubits, i.e. spin qubits in various materials, corresponds to frequencies in the microwave regime, it has been proposed to interface these with the circuit QED architecture [26, 86]. This was the origin of the idea of *hybrid circuit quantum electrodynamics* (hybrid circuit QED). Here, the superconducting qubit is replaced with a different mesoscopic qubit and similarly coupled to microwave radiation. Superconducting qubits, spin qubits and other solid state qubits offer different advantages or disadvantages over the others, such as longer coherence times, easier control over individual qubits or better scalability. Therefore, it has also been proposed to create and study hybrid quantum devices which combine the advantages of two or more quantum mechanical systems for quantum information processing [24, 26], see Figure 2.2.

2.2 On-Chip Microwave Cavities

Most implementations of circuit QED use transmission line resonators as a cavity. The main requirement for them is a large enough quality factor, i.e. a small decay rate κ , such that the quantum properties of the cavity can be observed. Due to good grounding conditions, easy design parameters and good compatibility with photolithography, mainly coplanar waveguides are used for the resonators [72]. To learn more about the design and fabrication parameters of these type of resonators refer to Section 4.2.

Coplanar waveguide resonators of length l , with capacitance per unit length C_l and inductance per unit length L_l , can be modelled as the continuum limit of a chain of LC oscillators [87], see Figure 2.3 (b). Each individual LC oscillator consists of a capacitor C with an inductor L in parallel. If there is no dissipation present in these oscillators they can be described quantum-mechanically [88], which is necessary if they are implemented in circuit QED. Resonators with vanishing dissipation can be achieved by fabricating them from superconducting material and measuring them at low enough temperature $T < T_c$. The Lagrangian of a single LC oscillator, see Figure 2.3 (a), is given by $\mathcal{L} = q^2/(2C) - \phi^2/(2L)$, where ϕ is the flux through the inductor and q the charge on the capacitor. The Hamiltonian H can now be obtained by using the Legendre transformation $H = \dot{\phi}q - \mathcal{L}$ and using the definition $q = C\dot{\phi}$, yielding $H = q^2/(2C) + \phi^2/(2L)$. Analogous to the text book example of the quantisation of a particle in a harmonic potential and making use of the direct mappings $p \rightarrow q$, $x \rightarrow \phi$ and $\omega_r^2 \rightarrow 1/(LC)$ the quantised hamiltonian of a single LC oscillator is given by

$$\hat{H}_r = \hbar\omega_r \left(\hat{a}^\dagger \hat{a} + \frac{1}{2} \right) \quad (2.2)$$

where the annihilation operator $\hat{a} = 1/\sqrt{2\hbar Z}(\phi + iZq)$ and creation operator $\hat{a}^\dagger = 1/\sqrt{2\hbar Z}(\phi - iZq)$ are introduced to diagonalise the Hamiltonian, obeying the commutation relation $[\hat{a}, \hat{a}^\dagger] = 1$, with the characteristic impedance $Z = \sqrt{L/C}$ and the angular resonance frequency ω_r .

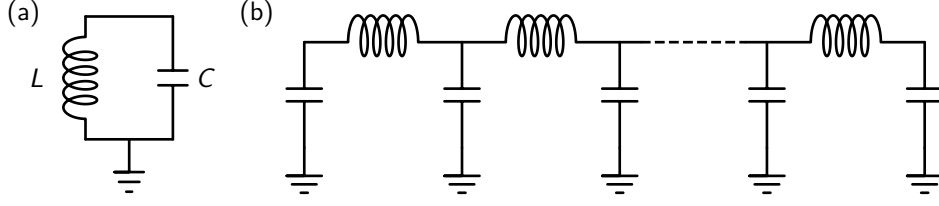


Figure 2.3: LC circuits. (a) The parallel LC oscillator with one resonance mode. (b) The coplanar waveguide resonator modelled as an infinite chain of LC oscillators.

Modelling the coplanar waveguide resonator as described above as the continuum limit of a chain of LC oscillators yields the effective Lagrangian $\mathcal{L} = \sum_{m=1}^{\infty} C_m \dot{\phi}_m^2 / 2 - \phi_m^2 / 2L_m$. This describes an infinite number of uncoupled LC oscillators with effective capacitances $C = C_m = C_l l / 2$, effective inductances $L = L_m = 2lL_l / (m^2 \pi^2)$, resonance frequencies $\omega_m = m\nu_{\text{ph}}\pi / l$ and phase velocity $\nu_{\text{ph}} = 1 / \sqrt{C_l L_l}$, see Figure 2.3 (b). Hence, the Hamiltonian of a coplanar waveguide resonator is then given as

$$\hat{H}_{\text{CPWR}} = \hbar \sum_m \omega_{r,m} \left(\hat{a}_m^\dagger \hat{a}_m + \frac{1}{2} \right) \quad (2.3)$$

where m is the mode number. In general, only the behaviour of a resonator in the vicinity of a particular frequency is of interest. In this case only one mode of the coplanar waveguide resonator has to be considered (most often the fundamental mode, $m=1$) and the dynamics of the other modes can be ignored. In such a case the Hamiltonian of the resonator can be reduced to that of a single LC oscillator and the quantum mechanical description portrayed in Equation 2.2 is sufficient.

2.3 The Jaynes-Cummings Hamiltonian

In circuit QED, and more generally in cavity QED, the interaction between an idealised two-level system, i.e. a qubit, and a single electromagnetic field mode inside a cavity is described by the famous Jaynes-Cummings model [89]. While there is no interaction present, the electromagnetic field can either be in the state $|n-1\rangle$ or $|n\rangle$, corresponding to the number of photons, n , in the cavity. Similarly the two-level system can either be in its ground state $|g\rangle$ or in its excited state $|e\rangle$,

both separated by $\hbar\omega_Q$. If both of these entities are interacting inside a cavity or resonator, see Figure 2.1 & 2.2, the total Hamiltonian is given by

$$\hat{H}_{JC} = \hat{H}_r + \hat{H}_Q + \hat{H}_{\text{int}} \quad (2.4)$$

where $\hat{H}_r$ describes the resonator field, cf. Section 2.2, and $\hat{H}_Q = \hbar\omega_Q\hat{\sigma}^+\hat{\sigma}^-$ the non-interacting two-level system. Similarly as for the resonator, $\sigma^+ = |e\rangle\langle g|$ and $\sigma^- = |g\rangle\langle e|$ are the raising and lowering operators for the two-level system.

The interaction Hamiltonian $\hat{H}_{\text{int}}$ is given, in the rotating wave approximation, by

$$\hat{H}_{\text{int}} = g\hbar(\hat{\sigma}^+\hat{a} + \hat{\sigma}^-\hat{a}^\dagger) \quad (2.5)$$

where as mentioned before g is the coupling strength. In the rotating wave approximation the total Hamiltonian given in Equation 2.4, is the so-called *Jaynes-Cummings Hamiltonian* [89] which describes the coupled system with the two hybridised ‘dressed-states’ $|g, n\rangle$ and $|e, n-1\rangle$.

However, the Jaynes-Cummings model has some limitations, since it treats the entity interacting with the resonator field as a two-level system. Whereas this would be true for electron spins, superconducting qubits and real atoms are complex quantum systems with multiple energy levels. Nevertheless, it is possible to use the Jaynes-Cummings model when other transitions are sufficiently detuned from and/or weakly coupled to the resonator in comparison to the transition of interest. Aside from that, the Jaynes-Cummings model is the description of a unitary closed quantum system, which does not include any losses due to the environment. In order to include loss mechanisms such as the spontaneous decay of the two-level system, quantified by the decay rate γ and the loss of photons from the resonator by transmission, scattering or absorption, described by the decay rate κ , a master equation approach has to be used [90].

2.3.1 The Tavis-Cummings Hamiltonian

An extension to the Jaynes-Cummings Hamiltonian is the case where N two-level systems are present in the cavity. The extended Hamiltonian is called *Tavis-Cummings Hamiltonian* [91] and for the interacting system is given by

$$\hat{H}_{\text{TC}} = \hat{H}_{\text{r}} + \frac{\hbar}{2} \sum_{i=1}^N \omega_{\text{Q}}^i \hat{\sigma}_i^+ \hat{\sigma}_i^- + \hbar \sum_{i=1}^N g_i (\hat{\sigma}_i^+ \hat{a} + \hat{\sigma}_i^- \hat{a}^\dagger) \quad (2.6)$$

where each two-level system has a unique transition frequency ω_{Q}^i and coupling strength to the resonator field g_i . In most cases an ensemble of identical two-level systems is coupled to the resonator and hence the assumption can be made that $\omega_{\text{Q}}^i = \omega_{\text{Q}}$ for all two-level systems. This reduces the Hamiltonian in Equation 2.6 to

$$\hat{H}_{\text{TC}} = \hat{H}_{\text{r}} + \frac{\hbar \omega_{\text{Q}}}{2} \sum_{i=1}^N \hat{\sigma}_i^+ \hat{\sigma}_i^- + \hbar \sum_{i=1}^N g_i (\hat{\sigma}_i^+ \hat{a} + \hat{\sigma}_i^- \hat{a}^\dagger). \quad (2.7)$$

The study of coupled ensembles in circuit QED showed a $\sqrt{N}$ enhancement to the coupling rate g [30, 31, 59, 92, 93]. This enhancement arises from the indistinguishability of the individual two-level systems with respect to a single photon in the resonator. Naturally, a single photon residing in the resonator will eventually be absorbed by the two-level systems. However, due to the indistinguishability of the two-level systems the photon is not absorbed by a single one, but by all of them simultaneously. This means once a photon is absorbed the two-level systems are in a superposition of each being excited while the others are in their ground state. The enhancement can be understood by introducing the collective operators

$$\hat{\Sigma}^+ = \frac{1}{\sqrt{N}} \sum_{i=1}^N \frac{g_i}{g} \hat{\sigma}_i^+, \quad \hat{\Sigma}^- = \frac{1}{\sqrt{N}} \sum_{i=1}^N \frac{g_i}{g} \hat{\sigma}_i^-, \quad (2.8)$$

where g is the average coupling strength across all two-level systems [94]. The collective operators obey the commutation relation $[\hat{\Sigma}^+, \hat{\Sigma}^-] = 1$, such that the collective excitation behaves like a harmonic oscillator, i.e. the ensemble of N two-level systems can contain many excitations. Expressing the interaction part of the Hamiltonian in Equation 2.7 in terms of these collective operators leads to

$$\hat{H}_{\text{TC}}^{\text{int}} = \hbar \sqrt{N} g (\hat{\Sigma}^+ \hat{a} + \hat{\Sigma}^- \hat{a}^\dagger), \quad (2.9)$$

which is almost identical to the Jaynes-Cummings Hamiltonian in Equation 2.5, but with a factor of $\sqrt{N}$ enhancement of the coupling strength g . It will be obvious from the following section why this enhancement is useful and additionally it will be used in the experiments presented in Chapter 6.

2.4 Spins and Magnetic Dipole Coupling

The Jaynes-Cummings model as well as the Tavis-Cummings model treat the system interacting with the resonator field as an ideal two-level system. Such a system could be resembled by a single electron spin. Since an electron spin exhibits a magnetic moment $\mu_s = -g_e\mu_B m_s$, where g_e is the Landé g-factor, μ_B the Bohr magneton and m_s the spin quantum number, it can be coupled to the magnetic field of the resonator mode. The single spin-photon coupling strength is proportional to the vacuum magnetic field B_{vac} of the resonator [95] and given by

$$g_s = \frac{g_e\mu_B B_{\text{vac}}}{4\hbar}, \quad (2.10)$$

where the factor 4 in the denominator comes from $m_s = 1/2$ as well as another factor 1/2 due to the spin only able to occupy half of the volume of the resonator. From the resonator geometry it can be estimated

$$B_{\text{vac}} \approx \frac{\mu_0 I_{\text{vac}}}{2(w + 2s)}, \quad (2.11)$$

where I_{vac} is the zero-point current in the resonator, and w and s the centre conductor width and spacing from the lateral ground planes, see Section 4.2.1. The zero-point current of a resonator with characteristic impedance Z_0 and resonance frequency f_0 is given by

$$I_{\text{vac}} = \pi f_0 \sqrt{\frac{\hbar}{Z_0}}. \quad (2.12)$$

For a resonator with $Z_0 = 50 \, \Omega$, $f_0 = 5.0 \, \text{GHz}$, $w = 10 \, \mu\text{m}$ and $s = 5 \, \mu\text{m}$ this yields $B_{\text{vac}} \approx 1.796 \times 10^{-9} \, \text{T}$ and therefore $g_s \approx 2\pi \cdot 12.57 \, \text{Hz}$. This is many orders of magnitude smaller than usual spin relaxation rates γ_s as well as common resonator decay rates κ . Hence, in order to reach strong coupling, the $\sqrt{N}$ enhancement of

the Tavis-Cummings model can be used. A spin ensemble of about $10^{12} - 10^{14}$ identical spins will most likely result in $g > \kappa, \gamma$.

2.5 Superconducting Qubits

A quantum bit or qubit represents the unit of quantum information, occupying a two-dimensional Hilbert space with basis $|0\rangle$ and $|1\rangle$ [96]. As opposed to a classical bit, which can only represent one of its two basis states 0 or 1, a qubit can be prepared in an arbitrary superposition of its basis $|\psi\rangle = a|0\rangle + b|1\rangle$, where $|a|^2 + |b|^2 = 1$. The idea of quantum information processing allows for schemes which promise exponential speedup of certain computational problems but also can give insight into fundamental physics [4, 8]. The notion of quantum computing has been a major driving force to advance research in multiple disciplines, including cavity and circuit QED, solid state and mesoscopic physics and nanotechnology. Within most of these fields an architecture implementing the idealised qubit concept represented by a two-level system has been developed. However, most of the quantum entities, apart from the spin-1/2 systems, used to implement such a qubit have more than two eigenstates. Nevertheless, in most cases the eigenstates are sufficiently distributed in a non-monotone (anharmonic) fashion, such that it is possible to choose two successive energy levels to unambiguously implement the two basis states of a physical qubit.

The foundation of superconducting qubits is the LC harmonic oscillator as these electrical circuits are easy to engineer using standard lithography techniques. The harmonic potential of the LC oscillator is modified by replacing the linear inductor with a non-linear inductive element, i.e. the Josephson junction. This yields an anharmonic potential where the first two energy levels can be used as the computational basis for the physical implementation of a qubit. In the following sections the physics of standard superconductor-insulator-superconductor (SIS) Josephson junctions and the implementation and evolution of the most common type of superconducting qubits is explained. At the end a more novel type of superconducting qubit known as a gatemon is discussed.

2.5.1 Josephson Junctions

The simplest form of a Josephson junction are two superconducting electrodes (S) separated/connected by a thin insulating barrier (I), see Figure 2.4 (a). If a thin enough barrier is used the macroscopic wave functions of both electrodes can overlap inside the insulator, creating a weakly coupled system. The overlap of the wave functions allows for Cooper pairs to coherently tunnel through the junction, yielding a superconducting tunnel current, i.e. supercurrent. This effect was first theoretically described by *B. Josephson* [83] in 1962 and hence such junctions were called *Josephson junctions*. The first Josephson equation describes the current phase relation in a Josephson junction by

$$I_s(t) = I_c \sin \varphi(t), \quad (2.13)$$

where $I_s(t)$ is the supercurrent through the junction, I_c the critical current of the junction that can maximally be carried before the junction becomes normal and $\varphi(t)$ the time-dependent phase difference between the two superconducting electrodes. The second Josephson equation describes the AC Josephson effect, relating φ to the voltage V across the junction by

$$V(t) = \frac{\Phi_0}{2\pi} \frac{\partial \varphi}{\partial t}, \quad (2.14)$$

where $\Phi_0 = h/2e$ is the magnetic flux quantum. Integrating Equation 2.14 and combining it with Equation 2.13 gives rise to an oscillating supercurrent with frequency V/Φ_0 given by

$$I_s(t) = I_c \sin \left(\frac{2\pi}{\Phi_0} Vt + \varphi_0 \right), \quad (2.15)$$

where φ_0 is an integration constant.

If the current I applied through the junction is smaller than the critical current, i.e. $I < I_c$, there is no associated voltage drop across the junction, which means there is no energy dissipated in the junction. Nevertheless, there is an energy associated with the tunnelling of Cooper pairs through the junction. By ramping an applied current through the junction up from $I_s = 0$ the phase difference

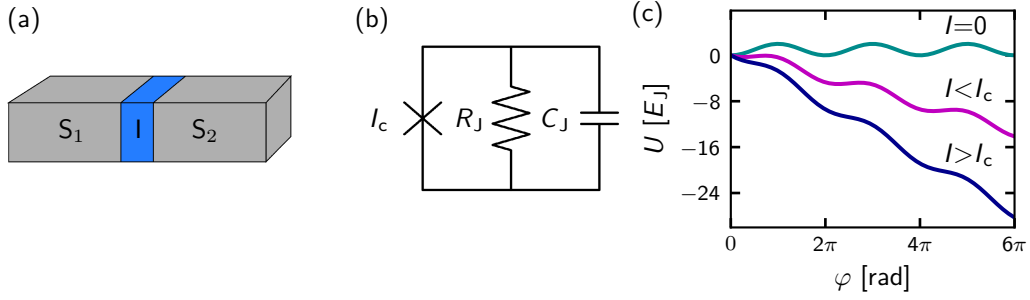


Figure 2.4: Josephson junction schematic and RCSJ model. (a) Schematic of a Josephson junction, where two superconductors (S_1 and S_2) are separated by an insulator (I). (b) Circuit diagram of the Josephson junction in the RCSJ model. (c) Tilted washboard potential. The potential tilts more with larger current bias I applied to the junction.

changes according to Equation 2.13, which in turn gives rise to a voltage across the junction due to Equation 2.14. The coupling energy is then given by integrating $\int I_s(t)V(t)dt$ over the ramp time, yielding

$$E(\varphi) = E_J(1 - \cos \varphi) \quad (2.16)$$

Following from this, the Josephson energy

$$E_J = \frac{\Phi_0 I_c}{2\pi} \quad (2.17)$$

is directly proportional to the critical current I_c .

The non-linearity of the inductance of a Josephson junction can now be determined by taking the time derivative of Equation 2.15 and comparing it with the expression for a voltage across a conventional inductance $V = L\partial I/\partial t$. This yields a phase dependent Josephson inductance given by

$$L_J = \frac{L_0}{\cos \varphi}, \quad (2.18)$$

where $L_0 = \Phi_0/(I_c 2\pi)$ is the specific Josephson inductance, which is only dependent on the critical current of the junction.

As can be seen, the critical current I_c and in turn the Josephson energy E_J are the important parameters of a Josephson junction. The *Ambegaokar-Baratoff relation* [97] gives a temperature dependence of I_c

$$I_c R_n = \frac{\pi \Delta(T)}{2e} \tanh \left(\frac{\Delta(T)}{2k_B T} \right), \quad (2.19)$$

where R_n is the normal state resistance of the junction, T the temperature and $\Delta(T)$ the superconducting gap of the material. For very low temperatures, i.e. $T \rightarrow 0$, this can be simplified to

$$I_c R_n = \frac{\pi \Delta(0)}{2e}. \quad (2.20)$$

Following BCS theory $\Delta(0)$ is related to the critical temperature T_c of the material by $\Delta(0) = 1.76 k_B T_c$ [98]. This allows the estimation of the vital junction parameter I_c through resistance measurements at room-temperature.

2.5.1.1 The RCSJ-Model

A Josephson junction can be in either of two states. A junction is in the *zero voltage state* as long as there is no voltage drop across the junction. This is the case when the DC bias current I applied to the junction is smaller than the critical current I_c of the junction. As soon as I exceeds I_c , the total current through the junction can no longer be supported by the supercurrent. At a finite temperature T , Cooper pairs can be thermally broken up into *quasiparticles* which can carry a normal current I_n through the junction. This current is a resistive current, i.e. $I_n = V/R_J$, where the voltage V drops across the junction with resistance $R_J = R_n$ for $I > I_c$. Hence the junction is in the *voltage state*.

In the voltage state the Josephson junction can effectively be modelled as a parallel circuit with resistance R_n and capacitance C_J [99], see Figure 2.4 (b). This description is known as the *resistively and capacitively shunted junction* (RCSJ) model. In this model the dynamics of the junction can be described by the differential equation

$$C_J \frac{\Phi_0}{2\pi} \frac{d^2 \varphi}{dt^2} + \frac{1}{R_J} \frac{\Phi_0}{2\pi} \frac{d\varphi}{dt} + \frac{d}{d\varphi} [E_J (1 - \cos \varphi - i\varphi)] = 0, \quad (2.21)$$

where $i = I/I_c$ is the normalised current. This description can be compared to the equation of motion of a particle with mass M moving along dimension x in a potential U with damping η

$$M \frac{d^2 x}{dt^2} + \eta \frac{dx}{dt} + \nabla U = 0. \quad (2.22)$$

From this it can be inferred that the potential U describing the phase difference of a Josephson junction is given by

$$U(\varphi) = E_J(1 - \cos \varphi - i\varphi), \quad (2.23)$$

which is also known as the *tilted washboard potential*. Further, the mass M of a *phase particle* moving in the potential U , its damping η and the quality factor Q_J of the Josephson junction are given by

$$M = C_J \frac{\Phi_0}{2\pi}, \quad (2.24)$$

$$\eta = \frac{1}{R_J} \frac{\Phi_0}{2\pi}, \quad (2.25)$$

$$Q_J = \sqrt{\frac{2\pi}{\Phi_0} I_c R_J^2 C_J} = \sqrt{\beta_C}, \quad (2.26)$$

where $\beta_C = (2\pi/\Phi_0)I_c R_J^2 C_J$ is the *Stewart McCumber parameter*, describing the damping of the Josephson junction. An underdamped Josephson junction exhibits a large β_C , i.e. large quality factor. An overdamped junction in turn exhibits a small β_C , i.e. small Q_J as the dissipation is high. The potential U for the phase difference is depicted in Figure 2.4 (c). The tilt of the potential increases with increasing bias current. For $i = 0$, the local potential barrier height is $U_0 \approx E_J$ and the oscillation frequency $\omega_0 = \sqrt{2\pi I_c / (\Phi_0 C_J)}$ is called the *Josephson plasma frequency*.

At $I > I_c$, i.e. a strong tilt of the potential, the phase particle can move freely down the potential. Reducing the bias current, the tilt of the potential is reduced until at $I < I_c$ local minima are obtained. For large damping the phase will immediately be trapped in one of these local minima. Hence, the phase does not evolve further and the junction is in the zero voltage state as soon as $I < I_c$. For small damping this situation is completely different. Here the phase particle has sufficiently high kinetic energy such that it can still easily move down the potential even for $I < I_c$. This means the applied current has to be almost zero until the junction reaches the zero voltage state. When increasing the current from zero, the behaviour of an over- and underdamped junction are the same and they stay in the zero voltage state until $I > I_c$ (neglecting thermally

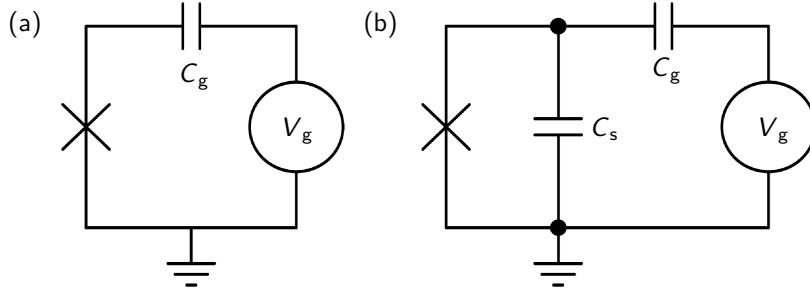


Figure 2.5: Circuit diagram of a Cooper pair box qubit (a) and a transmon qubit (b).

activated processes or tunnelling of the phase). This behaviour results in different current voltage characteristics for an over- and an underdamped Josephson junction. The overdamped Josephson junctions exhibit the same I-V curve if the current is increased or decreased. However, the underdamped junctions exhibit hysteresis in the I-V curve depending on whether the current is swept up or down.

2.5.2 The Cooper Pair Box

The Cooper pair box (CPB) is the first qubit prototype based on superconducting circuits [18, 100–102]. In a CPB two superconducting electrodes are separated by a Josephson junction, where one of them is connected to ground (reservoir) and the other one is floating (island), see Figure 2.5 (a). The island is additionally coupled via a capacitance C_g to a gate voltage source V_g . Once the CPB is superconducting, Cooper pairs ($2e$) in the reservoir can coherently tunnel through the Josephson junction, increasing the number n of excess Cooper pairs on the island. Obviously, the reverse tunnelling process is also allowed. The gate voltage V_g is a control parameter for this process and can induce charges $n_g = V_g C_g / (2e) + q_r / (2e)$, where $q_r / (2e)$ are residual charges due to the environment, e.g. charge noise. Hence the total number of charges on the island is given by $(n - n_g)$. Therefore, the CPB Hamiltonian can be written as

$$\hat{H}_{\text{CPB}} = 4E_C(\hat{n} - n_g)^2 - E_J \cos \hat{\phi}, \quad (2.27)$$

where $E_C = e^2 / (2C_\Sigma)$ is the charging energy required in order to charge the island with an additional electron, see Section 4.3.1 on how E_C can be influenced by the

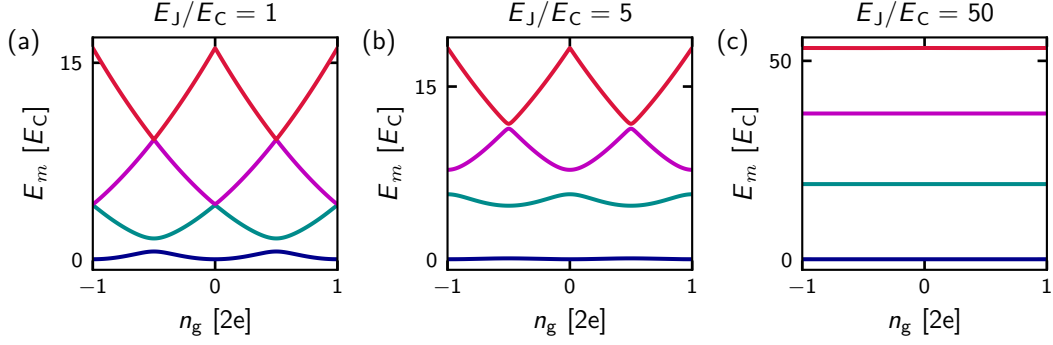


Figure 2.6: First four energy levels of a superconducting qubit based on the analytic solutions of the CPB Hamiltonian given by Equation 2.28, for different ratios of E_J/E_C as a function of the gate charge n_g . (a) Energy levels in the CPB limit for $E_J/E_C = 1$, where charge dispersion is very dominant. (b) The energy levels for $E_J/E_C = 5$. (c) Transmon regime of the CPB for $E_J/E_C = 50$. The energy levels are flat and hence resilient to charge noise.

circuit design. E_J is the Josephson energy given in Equation 2.17. The operators $\hat{n} = -\hat{q}/(2e)$ and $\hat{\varphi} = \hat{\phi}2e/\hbar$ describe the number of Cooper pairs transferred between the islands and the phase difference between the two superconducting electrodes, respectively. They are conjugate operators and satisfy the commutation relation $[\hat{\varphi}, \hat{n}] = -i$. It is possible to show that the Schrödinger Equation $\hat{H}_{\text{CPB}}\psi_m = E_m\psi_m$ can be solved analytically with the help of Mathieu's equations [103]. The solution yields the energy levels of the CPB given by

$$E_m(n_g) = E_C a_{2[n_g + k(m, n_g)]} \left(\frac{-E_J}{2E_C} \right), \quad (2.28)$$

where $a_\nu(q)$ denotes Mathieu's characteristic value and $k(m, n_g)$ is a function appropriately sorting the eigenvalues [104]. The energy levels suffer from charge dispersion, which describes the variation of the energy levels with respect to environmental offset and gate charge n_g [104], see Figure 2.6 (a) and (b). This determines the sensitivity of the CPB to charge noise, which has a detrimental effect on the coherence time of a qubit represented by a CPB. It is possible however to operate the CPB in a sweet spot where the qubit is to first order insensitive to charge noise [105].

2.5.3 The Transmon Qubit

As mentioned above the CPB suffers from charge dispersion which has a negative effect on the stability of the qubit. The transmission-line shunted plasma oscillation (or *transmon*) qubit, is a further development of the CPB which by design has vanishing charge dispersion. From the energy levels of the CPB it can be seen, that the ratio of the two characteristic energies E_J/E_C determines the energy level structure, cf. Equation 2.28 and Figure 2.6. Designing the qubit with $E_J/E_C \gg 1$, i.e. in the transmon regime, realises energy levels without charge dispersion, essentially reaching a permanent operation sweet spot for the qubit for all n_g and hence alleviating the need for the gate voltage V_g , see Figure 2.6 (c). In order to realise $E_J/E_C \gg 1$ with the same junction parameters, the charging energy E_C has to be reduced significantly. This can be fulfilled by capacitively shunting the Josephson junction with a large capacitance C_s (see Figure 2.5 (b)) which will increase C_Σ , see Section 4.3.1 for details. However, increasing the E_J/E_C ratio also decreases the anharmonicity of the energy levels [104]. For $E_J/E_C \gg 1$ the cosine term in the Hamiltonian given by Equation 2.27 can be expanded to fourth order and the quadratic term treated as a perturbation to the harmonic oscillator. This leads to the approximate eigenenergies

$$E_m \approx -E_J + \sqrt{8E_J E_C} \left(m + \frac{1}{2}\right) - \frac{E_C}{12} (6m^2 + 6m + 3). \quad (2.29)$$

From this it can be shown that the relative anharmonicity between the first two energy levels $\alpha = (E_{12} - E_{01})/E_{01}$ decreases more slowly with respect to E_J/E_C than the charge dispersion decreases with respect to E_J/E_C [104]. This allows for the implementation of an effective two-level system between the first two energy levels. The transition frequency of the transmon qubit is hence given by

$$f_{01} \approx \frac{\sqrt{8E_J E_C} - E_C}{h}. \quad (2.30)$$

In standard transmon qubits often a split junction design is used, see Figure 2.7 (a), where the single Josephson junction is replaced with a SQUID loop consisting out

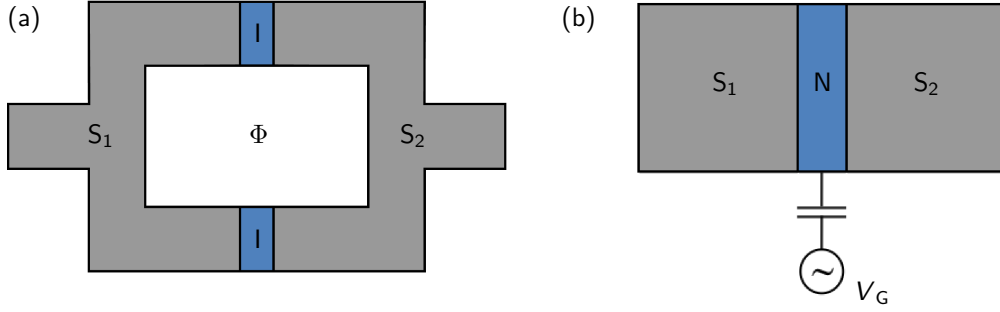


Figure 2.7: Split junction and gatemon schematic. (a) Split junction design, i.e. SQUID loop with two identical Josephson junctions, with external magnetic flux Φ threaded through the loop. (b) Gatemon schematic where two superconductors (S_1 and S_2) are separated by a normal conductor (N). The electrostatic potential, i.e. E_J can be tuned with gate voltage V_G .

of two identical Josephson junctions. The Josephson energy of such a SQUID loop depends on the external magnetic flux Φ threaded through the loop [106]

$$E_J(\Phi) = E_J |\cos \pi \Phi / \Phi_0|. \quad (2.31)$$

This effect allows for in-situ frequency tunable transmon qubits. However, split-junction transmon qubits suffer from decoherence due to flux noise.

2.5.4 The Gatemon Qubit

Gatemon qubits are a recent alteration of the transmon qubit, where the conventional SIS Josephson junction (most often made from Al/Al₂O₂/Al layers) is replaced by a semiconductor, normal conductor or a semi-metal with superconducting leads [50–52, 107]. These types of Josephson junctions are classified as SNS Josephson junctions and most often an electrostatic gate in close proximity to the normal conductor permits the tuning of their Josephson energy E_J by means of a gate voltage V_G ¹, see Figure 2.7 (b). This alleviates the susceptibility to flux noise of frequency tunable transmon qubits.

Recent experiments with SNS Josephson junctions made from carbon nanotubes [108], graphene [109] and InAs nanowires [110] revealed non-sinusoidal current-phase relations for this type of junction. Hence in order to describe a

¹For details on the gate tuning of E_J see Section 3.3.

gatemon qubit, the second term of the CPB Hamiltonian (Equation 2.27) needs to be modified, such that the particular junction behaviour is taken into account. Due to the few and highly transmissive conduction channels in SNS Josephson junction devices, charge transport across the junction is governed by Andreev reflections at the SN interface, see Section 3.3. Multiple Andreev reflections between the two interfaces result in discrete subgap states, i.e. Andreev bound states. Each of these states has a ground state energy $-\Delta\sqrt{1 - T_i \sin^2(\varphi/2)}$, where Δ is the induced superconducting gap and T_i the transmission of channel i [111–114]. Taking each conduction channel into account, the Josephson potential in a SNS junction due to Andreev bound states² is given by

$$U(\hat{\varphi}) = -\Delta \sum_i \sqrt{1 - T_i \sin^2(\hat{\varphi}/2)}, \quad (2.32)$$

where $\hat{\varphi}$ is the superconducting phase-difference operator as in Equation 2.27.

Following the analysis from *Kringshøj et al.* [114], the Hamiltonian of a gatemon qubit can now be written as a modified version of the CPB Hamiltonian given by Equation 2.27, where the gate offset charge n_g is omitted and the second term, i.e. the standard Josephson potential $-E_J \cos \hat{\varphi}$, is replaced by Equation 2.32 yielding

$$\hat{H} = 4E_C \hat{n}^2 - \Delta \sum_i \sqrt{1 - T_i \sin^2(\hat{\varphi}/2)}. \quad (2.33)$$

In order to examine the transition frequency f_{01} and anharmonicity α of a gatemon qubit the eigenenergies of Equation 2.33 have to be calculated. Finding the eigenenergies of this Hamiltonian analytically is very difficult. However, expanding $U(\hat{\varphi})$ given by Equation 2.32 to 4th order in $\hat{\varphi}$ yields

$$U(\hat{\varphi}) \approx \frac{\Delta}{4} \sum_i \left[\frac{T_i}{2} \hat{\varphi}^2 - \frac{T_i}{24} \left(1 - \frac{3}{4} T_i \right) \hat{\varphi}^4 \right] \quad (2.34)$$

$$= E_J \frac{\hat{\varphi}^2}{2} - E_J \left(1 - \frac{3 \sum T_i^2}{4 \sum T_i} \right) \frac{\hat{\varphi}^4}{24}, \quad (2.35)$$

where $E_J = \frac{\Delta}{4} \sum T_i$ has been used and the constant term has been omitted [104, 114–116]. It is obvious that the $\hat{\varphi}^2$ -term has the same form as the first order

²It should be noted that here only the lowest energy state is taken into account and hence all states of higher energy are disregarded.

expansion of the standard Josephson potential $-E_J \cos \hat{\varphi} \approx E_J \hat{\varphi}^2/2$. This yields the quantum harmonic oscillator Hamiltonian $\hat{H}_0 = 4E_C \hat{n}^2 + E_J \frac{\hat{\varphi}^2}{2}$ with eigenenergies $E_n = \hbar\omega(n+1/2)$ and eigenstates $|n\rangle$, where $\omega = \sqrt{8E_J E_C}/\hbar$. Now, treating $U'(\hat{\varphi}) = -E_J \left(1 - \frac{3\sum T_i^2}{4\sum T_i}\right) \frac{\hat{\varphi}^4}{24}$ as a perturbation to $\hat{H}_0$, allows us to calculate the corrections E'_n to the eigenenergies E_n by evaluating the matrix elements $E'_n = \langle n|U'(\hat{\varphi})|n\rangle$. The ground to first excited state transition frequency $f_{01} = (E_1 - E_0)/h$ of a gatemon qubit is hence given by

$$f_{01} = \sqrt{8E_J E_C} - E_C \left(1 - \frac{3\sum T_i^2}{4\sum T_i}\right), \quad (2.36)$$

where E_J and E_C are given in units of frequency. Additionally, the anharmonicity of the first three energy levels $\alpha = (E_2 - E_1) - (E_1 - E_0)$ of the gatemon qubit is then given by

$$\alpha = E_C \left(1 - \frac{3\sum T_i^2}{4\sum T_i}\right). \quad (2.37)$$

From this we can see that in the limit $T_i \rightarrow 0$ for all i , which is true for SIS Josephson junctions with infinite channels, we acquire the expressions $\alpha = E_C$ and $f_{01} = \sqrt{8E_J E_C} - E_C$ of a standard transmon qubit [104]. For the opposite limit $T_i = 1$ we get $\alpha = E_C/4$ and $f_{01} = \sqrt{8E_J E_C} - E_C/4$, effectively presenting a reduced non-linearity of a gatemon compared to a transmon qubit.

Assuming all channels N exhibit the same transmission $T_i = \mathcal{T}$, i.e. $\sum T_i = N\mathcal{T}$, and using $E_J = \frac{\Delta}{4} \sum T_i$, the expressions for the anharmonicity α and the qubit frequency f_{01} reduce to

$$f_{01} = \sqrt{8E_J E_C} - E_C \left(1 - \frac{3E_J}{\Delta N}\right) \quad (2.38)$$

$$\alpha = E_C \left(1 - \frac{3E_J}{\Delta N}\right). \quad (2.39)$$

These equations will be used for the qubit characterisation in the measurements presented in Chapter 8.

2.5.5 Photon-Qubit Coupling

The coupling between a superconducting qubit and a microwave resonator can be quantified by the voltage V of the resonator. This voltage results from the resonator's electric field which occurs between the centre conductor and the ground planes and in turn has an effect on the qubit's Hamiltonian. The electrostatic part of the Hamiltonian given in Equation 2.27 depends on the number of charges n_g induced by a gate voltage V_g . If the qubit is placed into a resonator this part has to be extended by the resonator voltage V . Since $V = q/C$, the voltage operator can be expressed as [117–119]

$$\hat{V} = \sqrt{\frac{\hbar\omega_r}{2C}} (\hat{a} + \hat{a}^\dagger), \quad (2.40)$$

where C is the capacitance of the resonator. The resonator voltage couples to the qubit via capacitance C_g and hence the number of Cooper pairs n_g induced by V_g has to be replaced by $n_g = (V_g + \hat{V})C_g/(2e)$. Inserting this into Equation 2.27 and expanding the square leads to an additional term in the CPB Hamiltonian

$$\hat{H}_{\text{coup}} = 2\hbar g(\hat{a} + \hat{a}^\dagger)\hat{n}, \quad (2.41)$$

where a constant energy offset has been omitted. The coupling strength g in Equation 2.41 is further defined as

$$g = \beta \frac{e}{\hbar} \sqrt{\frac{\hbar\omega_r}{2C}}, \quad (2.42)$$

with $\beta = C_g/C_\Sigma$ being the voltage division across the Josephson junction, see Section 4.3.1. Making the assumption that there are only two qubit states, e.g. valid at $n_g = 0.5$, $\hat{n}$ can be replaced with the Pauli spin operator $\sigma_x/2$. Additionally applying the rotating wave approximation, which neglects fast oscillating terms, the interaction Hamiltonian of the Jaynes-Cummings model is recovered

$$\hat{H}_{\text{coup}} = \hbar g(\hat{\sigma}^+ \hat{a} + \hat{\sigma}^- \hat{a}^\dagger). \quad (2.43)$$

More generally for multilevel systems, such as transmon or gatemon qubits the coupling strength between the energy levels i and j also depends on the transition matrix element, hence Equation 2.42 is modified to

$$g = 2\beta eV_0 \langle i | \hat{n} | j \rangle / \hbar, \quad (2.44)$$

where $V_0 = \sqrt{\hbar\omega_r/(2C)}$ is the root mean square (rms) voltage of the resonator due to (quantum-mechanical) vacuum fluctuations. This can be derived from the relation $\frac{1}{2}\hbar\omega_r = \frac{1}{2}CV_0^2$ and using the standard definition for the rms voltage of a sine wave. For $E_J/E_C \gg 1$ the the matrix elements can be derived with a perturbative approach [104, 118]

$$|\langle j+1 | \hat{n} | j \rangle| \approx \sqrt{\frac{j+1}{2}} \left(\frac{E_J}{8E_C} \right)^{1/4}, \quad (2.45)$$

where all non-nearest neighbour transitions, i.e. $|\langle j+k | \hat{n} | j \rangle|$ with $|k| > 1$, are negligible.

2.5.6 Dispersive Regime and Qubit Readout

Up to now, the Jaynes-Cummings model was considered in the case where the detuning Δ between the qubit transition frequency and the resonator frequency was small or negligible, i.e. the qubit and resonator could exchange energy coherently due to the vacuum Rabi oscillations. In implementations of circuit QED for quantum information processing this is not ideal as it leads to a faster excited state decay of the qubit due to the Purcell effect. Therefore, almost all of the implementations of circuit QED operate in the so called *dispersive regime*, where $\Delta = |\omega_r - \omega_q| \gg g$. Here, virtual photons mediate the dispersive interactions between the qubit and the resonator which lead to frequency shifts $\chi = g^2/\Delta$ [117–119]. In this limit the Jaynes-Cummings Hamiltonian can be written as [72]

$$\hat{H}_D \approx \hbar(\omega_r + \chi\hat{\sigma}_z)\hat{a}^\dagger\hat{a} + \frac{\hbar}{2}(\omega_Q + \chi)\hat{\sigma}_z, \quad (2.46)$$

where the frequency shift in the qubit Hamiltonian is the Lamb shift.

The first term of the Hamiltonian given by Equation 2.46 shows that there is a shift of the resonator frequency of $\pm\chi$ depending on the qubit state $|e\rangle$ or $|g\rangle$, since

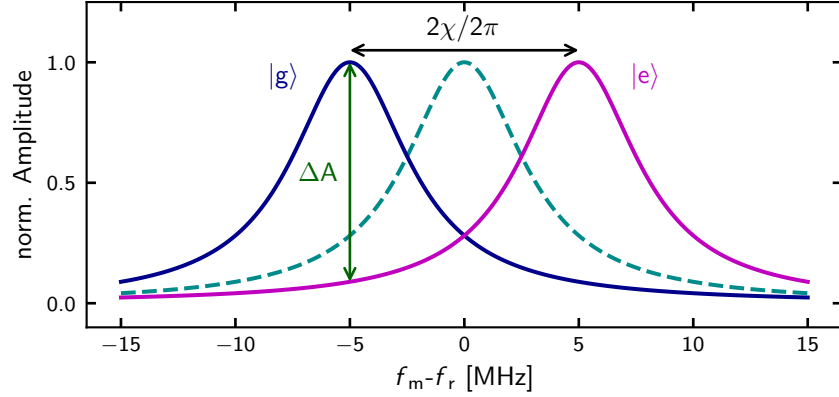


Figure 2.8: Qubit state dependent resonator shift of the transmission amplitude. Solid lines represent the resonator spectrum for the qubit being in the ground state $|g\rangle$ (blue) or excited state $|e\rangle$ (purple). The dashed line indicates the unshifted bare resonator transmission spectrum.

$\hat{\sigma}_z |g\rangle = |g\rangle$ and $\hat{\sigma}_z |e\rangle = -|e\rangle$. This shift of the resonator frequency is illustrated in Figure 2.8. When the qubit is in state $|e\rangle$ the resonator frequency is offset by 2χ compared to the resonance frequency when the qubit is in state $|g\rangle$. Therefore, depending on the qubit state $|g\rangle$ (blue) or $|e\rangle$ (purple) a measurement of the resonator transmission yields a different amplitude at the measurement frequency f_m , see Figure 2.8. The measurement frequency f_m can be chosen to optimise the amplitude contrast ΔA between both states. Hence, this effect can be used to determine the qubit state from the resonator frequency or transmission amplitude at a fixed measurement frequency f_m . This essentially is a quantum non-demolition (QND) measurement of the qubit state, where the qubit state is projected but not destroyed [120]. Measurements using this technique are presented in Chapter 8.

3

Carbon Nanotubes and Quantum Transport

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Carbon nanotubes and their quantum transport characteristics with regard to normal or superconducting leads are an essential building block of the experiments presented in Chapter 7 and Chapter 8. Here in this chapter, the basic properties of carbon nanotubes will be discussed as well as the concept of single quantum dots. Additionally, the transport mechanisms through SNS Josephson junctions, i.e. carbon nanotubes with superconducting leads, will be reviewed.

3.1 Carbon Nanotubes

Carbon, being the chemical basis of all known life, is the only element which has stable allotropes in all four structural dimensions, ranging from zero to three dimensions. One of these allotropes is the three-dimensional version of a rolled-up graphene sheet, the *carbon nanotube*. Carbon nanotubes are long extremely thin cylinders purely made out of carbon atoms arranged in a hexagonal pattern, exhibiting remarkable physical properties. The first carbon nanotubes were discovered by *Sumio Iijima* [121] using a high resolution transmission electron microscope. After their discovery, carbon nanotubes sparked a lot of scientific interest and most of their properties have been studied and reviewed in a lot of detail [122–126]. Depending on their length, diameter and chirality, carbon nanotubes exhibit a broad range of electronic, thermal and structural properties.

Carbon nanotubes are formed by a rolled-up graphene sheet, which consists of a mono-layer of carbon atoms arranged in a hexagonal lattice structure. The nanotubes can exhibit different diameters, where single-walled nanotubes (SWNT) have diameters of $\sim 1 - 3$ nm and multi-walled nanotubes (MWNT) diameters $\gtrsim 3$ nm. The hexagonal lattice structure of SWNT is characterised by the pair of chiral indices (m, n) representing the chiral vector $\mathbf{C} = m\mathbf{a} + n\mathbf{b}$, where $\mathbf{a}$ and $\mathbf{b}$ are graphene basis vectors, see Figure 3.1 (a). The chirality of SWNTs can be divided in three structural classes defined by (m, n) , specifically zig-zag ($m = 0$), armchair ($m = n$) and chiral (arbitrary (m, n)), see Figure 3.1 (b). The properties of MWNTs are similar to SWNTs, however the inner carbon nanotubes are protected from chemical interactions by the outermost wall. MWNTs can be formed either by a larger nanotube containing smaller ones inside or a single graphene sheet rolled around itself multiple times, similar to a rolled-up sheet of paper [126]. The walls are held together by the van der Waals force and are separated by ~ 3.3 Å, similar to the distance between graphene layers in graphite [124, 126]. The bonds in between the carbon atoms are sp^2 -hybridised lending the carbon nanotubes a high tensile strength as well as elasticity. However, defects and rearrangements of the carbon bonds weaken the carbon nanotube.

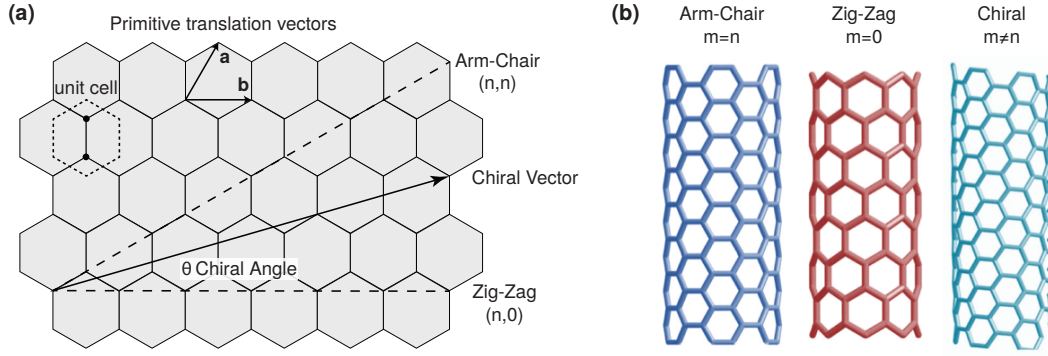


Figure 3.1: (a) Hexagonal lattice structure of graphene, showing the chiral vector, chiral angle, unit cell and the primitive translation vectors, i.e. graphene basis vectors. (b) The three different types of carbon nanotubes defined by the chiral vector.

3.1.1 Electronic Band Structure

The band structure of carbon nanotubes can be derived from its origin of the graphene band structure. A single carbon atom has six electrons which occupy the configuration $1s^2 2s^2 2p^2$ [127]. The first atomic shell $1s^2$ is fully occupied, including the twofold spin degeneracy. The outer shell includes one s and three p orbitals (p_x, p_y, p_z) and hence, due to spin, eight states out of which four are occupied. Since the $2s - 2p$ energy splitting is small compared to typical bonding energies, all four outer orbitals can hybridise and form covalent bonds. In graphene the p_z orbital is parallel to the normal of the lattice vectors and has odd parity. The other orbitals of the outer shell, i.e. $2s, 2p_x, 2p_y$ have even parity, which allows them to form a sp^2 hybridisation. Across neighbouring carbon atoms in the graphene lattice, the sp^2 hybridisation results in a low energy σ band (bonding) and high energy σ^* (antibonding) band, see Figure 3.2 (a). The p_z orbitals of neighbouring atoms also hybridise in bonding and antibonding bands, labelled π and π^* respectively, see Figure 3.2 (a). In regular graphene only the bonding orbitals are filled, with three electrons occupying σ and one electron occupying π . Since the electron occupation of σ remains unchanged in graphene, the electrical transport behaviour is only determined by the π and π^* band.

The band structure of graphene can now be constructed by considering the hexagonal lattice structure representing the Brillouin zone. In k -space the corners

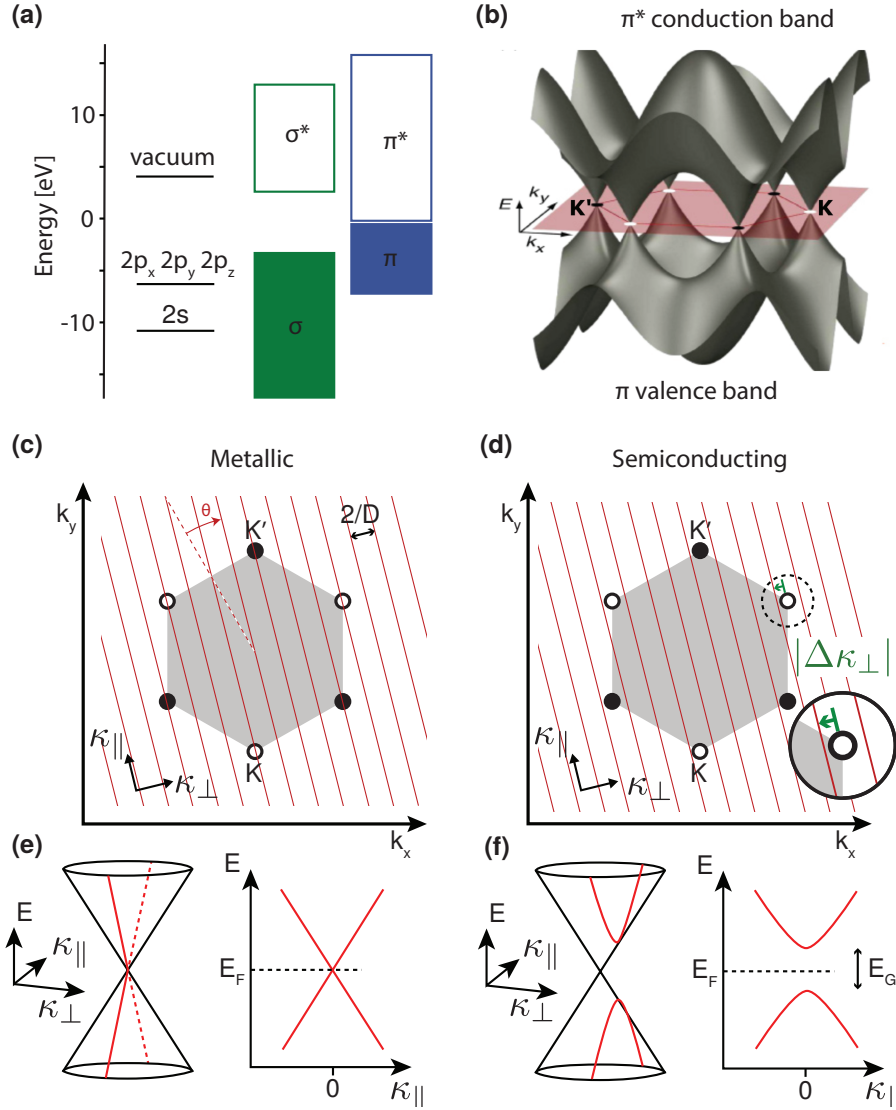


Figure 3.2: Band structure of graphene and carbon nanotubes. (a) Graphene energy bands in the free and hybridised case. Hybridisation leads to two different types of bonding and antibonding bands. (b) Band structure of graphene in the tight-binding model. The π and π^* bands intersect at the Dirac points (K and K') on the Fermi level. (c) Graphene unit cell in k -space. The quantisation lines intersect with the Dirac points, running at an angle $\pi/3 + \theta$ from the k_x axis, where θ is the chiral angle. (d) Graphene unit cell with quantisation lines offset from the Dirac points by $|\Delta\kappa_{\perp}|$. (e), (f) Dispersion relations close to the Dirac point for the two different cases in (c) and (d) respectively. (f) The offset of the quantisation lines in (d) give rise to a bandgap E_G in the carbon nanotube.

of this honeycomb shape are alternately labelled K and K' , also known as *Dirac points*. Each set of three corner points K (K') can be connected with reciprocal lattice vectors and hence according to Bloch's theorem all K (K') correspond to

equivalent electron states (K and K' are not equivalent) [127]. Even though there is no band gap present in graphene, the π and π^* orbitals only coincide at the Dirac points, where the density of states is zero, see Figure 3.2 (b). All available electrons exactly fill the π band which results in the Fermi level E_F crossing at exactly the intersection between the π and the π^* band. Therefore, graphene is neither a true metal nor a semiconductors, but a semimetal.

Rolling up a sheet of graphene into a carbon nanotube with diameter much larger than the interatomic spacing, leaves the band structure of graphene approximately unperturbed except for introducing a periodic boundary condition [127]. This is commonly called the *zone-folding approximation*. The introduced boundary condition can be expressed as $\mathbf{k} \cdot \mathbf{C} = 2\pi p$, where $\mathbf{k}$ is the translation vector in k-space, $\mathbf{C}$ the chiral vector and p and integer. Further, the diameter of SWNTs is proportional to the chiral vector and given by $D = |\mathbf{C}|/\pi$ [122]. Hence, the component of $\mathbf{k}$ perpendicular to the nanotube axis and hence parallel to $\mathbf{C}$ is given by $k_{\perp} = 2p/D$ [127]. From this it is obvious to see that the electron wavenumber perpendicular to the nanotube axis is only allowed to obtain quantised values, which can be represented as discrete lines in k-space spaced at a distance $2/D$, see Figure 3.2 (c). Since usually nanotubes are much longer than their diameter the component of $\mathbf{k}$ parallel to the nanotube axis can be considered to be continuous. Taking a cross-section of the graphene band structure along the quantisation lines yields the one-dimensional dispersion relation $E(\kappa_{\parallel})$ of a carbon nanotube. Since only the branch closest to the Fermi energy contributes to the transport properties, only the quantisation line closest to the Dirac point has to be considered and all other can be neglected. From this two possible configurations become apparent: The quantisation line crosses through the Dirac points resulting in the one-dimensional dispersion relation being linear near $\kappa_{\parallel} = 0$ and crossing through E_F , see Figure 3.2 (e). This yields zero bandgap and hence a metallic carbon nanotube. Conversely, if the quantisation line bypasses the Dirac points at a distance $|\Delta\kappa_{\perp}|$ (see Figure 3.2 (d)), the cross-section yields a carbon nanotube dispersion relation of two hyperbolae with a bandgap of $E_g = 2\hbar v_F |\Delta\kappa_{\perp}|$ separating

them [127], see Figure 3.2 (f). Hence, the nanotube is semiconducting. The Fermi velocity $v_F = 1/\hbar \cdot dE/dk \approx 8 \times 10^5$ m/s can be determined by the slope of the dispersion relation in the linear regime.

Using the zone-folding approximation, the bandgap can be determined with a simple rule: If $n - m$ is a multiple of 3, then the carbon nanotube is metallic [122, 128]. Otherwise, the carbon nanotube is semiconducting and exhibits a bandgap $E_g = 4\hbar v_F/3D \approx 700$ meV/D [nm] [127]. Therefore, two-thirds of carbon nanotubes are semiconducting and one-third is metallic. This also means all armchair nanotubes are metallic, whereas zigzag and chiral carbon nanotubes can either be metallic or semiconducting. However, it has been observed experimentally that even metallic carbon nanotubes exhibit a small bandgap, which is either strain-induced or due to the curvature of the nanotube [129–133]. Further details of this narrow bandgap in metallic carbon nanotubes can be found in Reference [127].

3.1.2 Contacting Carbon Nanotubes with Metal Leads

In order to make use of the exceptional mechanical and electrical properties of carbon nanotubes in nanoelectrical circuits they have to be accessed via metal leads. Further, quantisation along the nanotube axis requires the electron wave function to be confined or reflected at the barriers defining a single nanotube circuit. These barriers can be defined by local gates or by the barrier naturally occurring at the nanotube metal interface. In principle ohmic contacts between the metal leads and the carbon nanotube are desired, but the small contact area makes this challenging.

The barrier at the interface between the carbon nanotube and the metal leads can be described with the theory of Schottky barriers [134]. For the observation of quantum effects, such as Coulomb blockade or Andreev states in carbon nanotube circuits, ohmic contacts, i.e. low Schottky barriers are required. However, for applications in Schottky diodes high barriers are desirable [135]. Contacting a semiconducting nanotube with metal results in band bending at the interface and the formation of Schottky barriers. The band alignment at the interface determines the barrier height. Making the assumption that E_F is in the middle of

the carbon nanotube gap, the height of the Schottky barrier for electrons (e) and holes (h) is given by

$$\Phi_{\text{SB}}^h = \phi_{\text{CNT}} + E_g/2 - \phi_m \quad (3.1)$$

$$\Phi_{\text{SB}}^e = \phi_m + E_g/2 - \phi_{\text{CNT}}, \quad (3.2)$$

where E_g is the semiconducting nanotube gap and ϕ_m and ϕ_{CNT} are metal and nanotube work-functions, respectively. If the Fermi level of the metal falls within the semiconducting gap of the carbon nanotube, a Schottky barrier exists for both electrons and holes. However, if the Fermi level is aligned with the hole band of the carbon nanotube, i.e. $\phi_m > \phi_{\text{CNT}}$, ohmic contacts for holes are achieved. Conversely, for the case that $\phi_m < \phi_{\text{CNT}}$ ohmic contacts for electrons are achieved. More generally, in the latter case, a metal with a low work-function such as Al gives a good n-contact. In the opposite case, for a metal with a large work-function such as Pd or Rh results in better p-contacts [136, 137]. If the interface exhibits an ohmic contact for electrons (holes) there is a large barrier for holes (electrons). The width of the Schottky barrier can be tuned using an applied gate voltage which tunes the energy levels. *Chen et al.* [138] studied the influence of different contact metals on the conductance of semiconducting nanotubes and found that p-type Pd contacts result in the highest conductance. This is in line with the corresponding work-functions $\phi_{\text{Pd}} = 5.1$ eV and $\phi_{\text{CNT}} \sim 4.9$ eV. In general, not only the work-function is important for the metal to nanotube contact but also the surface chemistry at the interface. This property can be described as *wettability* and is exceptionally good for Pd compared to other metals such as Pt, Ti, etc. [139]. Hence, Pd contacts are used to contact the carbon nanotubes in the experiments described in Chapter 7 and Chapter 8, in order to achieve reproducible ohmic, high transparency contacts which are favourable to observe Andreev physics, see Section 3.3.

3.2 Quantum Dots

A quantum dot can be formed by confining electrons in all three spatial dimensions to a small island, which is weakly coupled to the environment via tunnel barriers. These

tunnel barriers allow the study of charge transport through a quantum dot [140, 141]. Performing this type of transport measurement showed that quantum dots exhibit a similar shell structure as known in atoms if single electrons are discretely added to them [142]. Therefore, quantum dots are sometimes also called artificial atoms. Their interaction with light has also been studied. In this setting, photon assisted tunnelling - due to the resonant absorption of a microwave photon by an electron inside a quantum dot - has been observed in transport measurements [143, 144]. Single quantum dots and their respective transport properties can be realised in many different systems, ranging from semiconductor nanostructures such as GaAs [145] to bottom- or top-gated carbon nanotubes [146].

In the following section, the basic transport properties of carbon nanotube single quantum dots will be reviewed. More detailed and general summaries of quantum transport can be found in books [145, 147] and review articles [140, 141, 148].

3.2.1 Carbon Nanotube Single Quantum Dots

Contacting a carbon nanotube with metallic leads introduces potential barriers for electron transport. The carbon nanotube segment defined by the two metallic contacts becomes isolated if the barrier resistance is larger than the resistance quantum $R_b \geq h/e^2$ and a quantum dot is formed. The electron wave vector parallel to the nanotube axis has to fulfil the standing wave boundary condition

$$\kappa_{\parallel} = \frac{n\pi}{L}, \quad (3.3)$$

where $n \in \mathbb{N}$ and L is the nanotube segment length. Combining this with the quantisation of the transverse component described in Section 3.1.1 yields discrete energy levels in the carbon nanotube quantum dot. Hence, single electron charging effects can be observed in this regime. To date, a multitude of experiments have been performed studying charge transport through carbon nanotubes [127, 146]. In most charge transport experiments the electrical current I or the differential conductance $\partial I / \partial V$ through the device is measured. The conductance G is proportional to the

transmission probability T_i of an electron passing through channel i of the carbon nanotube. Hence the conductance is expressed as

$$G = \frac{e^2}{h} \sum_i T_i. \quad (3.4)$$

In the case of ballistic transport and perfect ohmic contacts $T_i \rightarrow 1$. Since carbon nanotubes exhibit four transport channels due to the spin and valley degeneracy their maximum conductance is $4e^2/h$, which corresponds to a resistance of 6.45 k Ω . This can be used to pre-select carbon nanotube devices at room temperature. In carbon nanotube quantum dots with a single gate, the conductance can be tuned by applying a voltage to this gate. If the gate is well decoupled from the leads T_i stays constant.

In order to understand the charge transport properties of a single quantum dot, it is the easiest to take a closer look at its electric circuit representation, which is depicted in Figure 3.3 (a). The single quantum dot can be thought of being an electric island which is coupled by tunnel barriers to two reservoirs, namely the source (S) and drain (D) contact. The two barriers, allowing electrons to be exchanged between the dot and the reservoirs, are modelled as a capacitor (C_S and C_D) in parallel with a resistor. The quantised, discrete energy levels inside the quantum dot are defined by the electrostatic potential on the dot. This can be tuned by gate voltage V_G applied to a gate, which is coupled via capacitance C_G to the dot [140, 141, 145, 147, 149].

Measuring the transport current (or differential conductance) through the dot as a function of the gate voltage V_G at a small source-drain bias V_{SD} , gives rise to a discrete, evenly spaced set of current peaks, see Figure 3.3 (b). These peaks in the transport current are caused by Coulomb interactions between the electrons inside the dot, which lead to allowed (resonances) and disallowed regions of conductance. The latter effect is called Coulomb blockade. The mentioned characteristics of the transport current can only be observed, if the energy needed to add one electron to the dot is much larger than the thermal energy of the electrons

$$E_C = e^2/C_\Sigma \gg k_B T_e, \quad (3.5)$$

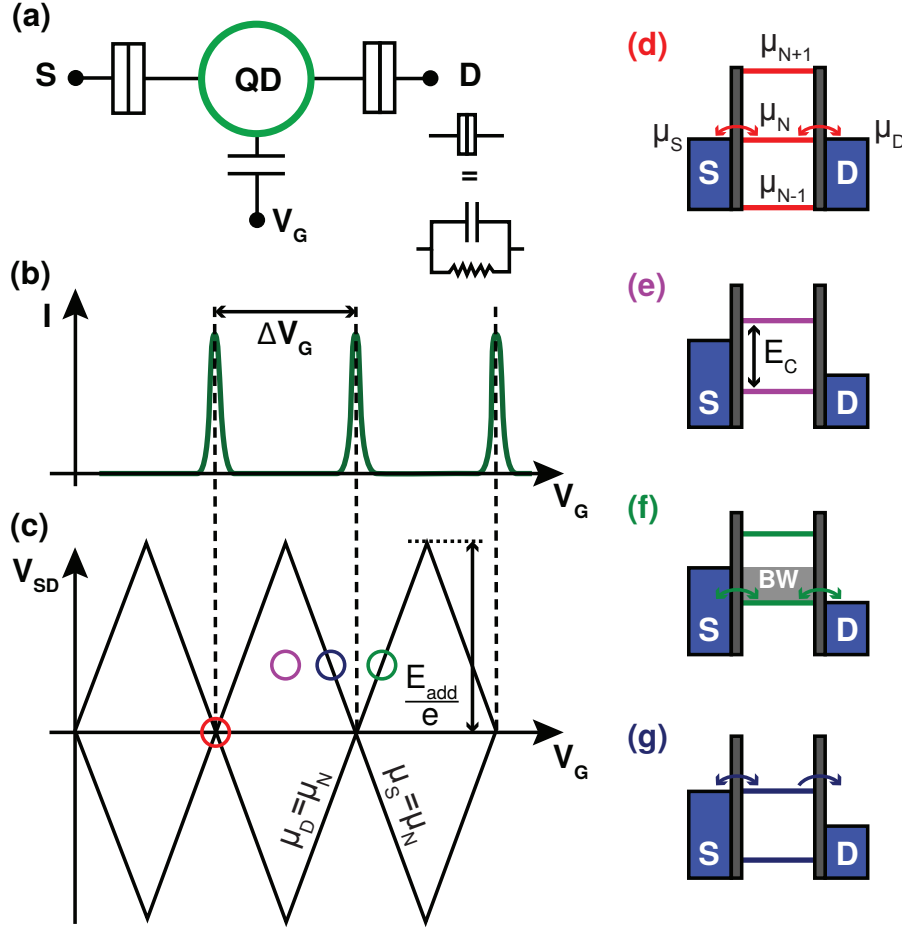


Figure 3.3: (a) A single quantum dot represented as an electrical circuit. Source (S) and drain (D) contact allow the measurement of charge transport current through the quantum dot which is mediated by the tunnel barriers. These are represented with a parallel combination of a capacitor and a resistor. The gate is capacitively coupled to the dot. (b) Charge transport current as a function of the gate voltage V_G , showing the distinct conductance resonances. (c) Schematic of a typical charge stability diagram of a single quantum dot exhibiting the distinct Coulomb diamonds. The addition energy E_{add} is labelled and μ_S , μ_N and μ_D correspond to the electrochemical potentials of the source contact, the quantum dot and the drain contact respectively. (d-g) Energy level diagrams of the quantum dot for different configurations on the stability diagram. Tunnel processes of electrons are indicated by arrows. (d) Configuration of conductance resonance at zero source-drain bias. (e) Coulomb blockade configuration at finite bias. (f,g) Transport configuration at finite bias. The difference between μ_S and μ_D is labelled as the bias window (BW).

where $C_\Sigma = C_S + C_D + C_G$ is the self-capacitance of the dot. This energy is the so-called charging energy E_C , which is defined as the electrostatic energy difference between a dot occupied with N or $N - 1$ electrons [140, 141, 145, 147, 149]. Metallic and semiconducting carbon nanotubes exhibit a dot level spacing

given by $\Delta = \hbar v_F \pi / L$ [122, 150].

Until now only the situation for a fixed source-drain bias was considered, i.e. $V_{SD} = 0$ V. Measuring the transport current as a function of the gate voltage and the source-drain bias shows the Coulomb resonances evolving as straight lines away from $V_{SD} = 0$ V. This leads to the so called Coulomb diamonds inside which the tunnelling through the dot is blocked [140, 141, 145, 147, 149], see Figure 3.3 (c). In terms of source-drain bias, the height of these Coulomb diamonds is a direct measure for the addition energy $E_{\text{add}} \approx E_C + \Delta$ which is the linear sum of the charging energy E_C and the single particle excitation energy Δ , see Figure 3.3 (c). The gate voltage difference ΔV_G between two resonances in conductance can be used to compute the corresponding lever arm [140, 141, 145, 147, 149]

$$\alpha_G = \frac{|V_{SD}|}{|\Delta V_G|}. \quad (3.6)$$

The energy levels of the dot (μ_N) with respect to the Fermi level of the source- and drain-contacts are sketched in Figure 3.3 (d-g) for different positions in the Coulomb diamonds. In situation (d) one electron is allowed to tunnel from one of the contacts into the dot and out again, since the source and drain Fermi levels (μ_S, μ_D) are aligned with one of the dot's energy levels. At position (e) however, the dot is in the Coulomb blockade since no energy level inside the dot is within the difference between the source and drain Fermi level. The latter is the so-called bias window (BW), which is depicted in Figure 3.3 (f). The configuration in (f) and (g), where the Fermi level of either the source (μ_S) or the drain (μ_D) contact coincides with an energy level of the quantum dot (μ_N), form the right (positive slope β_S) and left (negative slope β_D) boundary of one of the Coulomb diamonds respectively [137, 140, 141, 145, 147, 149]. Further, when $\mu_N = \mu_D$ the slope of the diamond edge is given by

$$\beta_D = \frac{V_{SD}}{V_G} = -\frac{C_G}{C_S}. \quad (3.7)$$

And equally when $\mu_N = \mu_S$ the other slope of the diamond edge is given by

$$\beta_S = \frac{C_G}{C_G + C_D}. \quad (3.8)$$

Details to the derivation of the formulae for the two slopes of the diamond edges can be found in References [137, 145]. Additionally, the width of a Coulomb diamond in gate voltage is equal to e/C_G . The slopes and the width of the Coulomb diamond can easily be calculated from measurements and allow the estimation of all three of the coupling capacitances.

So far only electron transport through the ground states of the carbon nanotube quantum dot have been considered. However, if the tunnelling rates of the leads exceed the relaxation rate of an excited state of the quantum dot, transport through this state becomes possible. For transport through the n -th excited state its electrochemical potential $\mu_N^{(n,0)}$ must fall within the bias window. In the charge stability diagram the measured current through the quantum dot as a function of V_{SD} and V_G is converted to the differential conductance $\partial I/\partial V$. Here the transition through the excited states shows up as lines parallel to the edges of the Coulomb diamond [145]. Further, when V_{SD} is larger than the energy of the lowest excited state, inelastic co-tunnelling becomes possible, i.e. an electron from the ground state leaves the quantum dot into the lead and another electron enters the dot excited state from the other lead with higher Fermi energy. This process shows up as step-like increase in the differential conductance parallel to $V_{SD} = 0$ V, which can be weakly gate-dependent [145].

3.3 SNS Josephson Junctions

In Section 2.5.1 conventional aluminium-aluminium oxide, i.e. SIS, Josephson junctions and their behaviour have been introduced. Here, a different type of Josephson junction is presented, namely SNS Josephson junctions where the insulator is replaced with a normal conductor. In the experiments presented in Chapter 7 and Chapter 8 the normal conductor is represented by a carbon nanotube. The introduction of a normal conductor with a few transport channels has a pronounced effect on the transport characteristics of the Josephson junction formed by the hybrid superconducting circuit, which will be described in detail in the following sections.

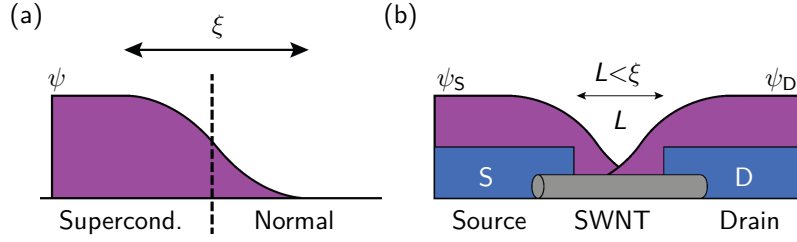


Figure 3.4: Simplified picture of the superconducting proximity effect. (a) The superconducting wave function decays into the normal conductor on a length scale ξ . Hence, a finite density of Cooper pairs is found in the normal conductor. (b) Two superconductors are coupled by a carbon nanotube weak link. Cooper pairs can tunnel into the carbon nanotube and are transferred to the other superconductor if the contact spacing $L < \xi$, where ξ is the coherence length of Cooper pairs in the carbon nanotube.

3.3.1 Superconducting Proximity Effect

Upon connecting a normal conductor to a superconductor, the electrons in the normal conductor exhibit correlations from the superconductor. This induces a superconducting gap in the density of states of the normal conductor and hence the effect is called proximity induced superconductivity (i.e. proximity effect). The superconducting wave function decays into the normal conductor on the length scale of the coherence length at the superconductor/normal conductor interface [151–153], see Figure 3.4 (a). The proximity effect can be observed in various SN-systems where the normal conductor N can be graphene [154], carbon nanotubes [155], normal metals [156], ferromagnetic materials [157, 158], and semiconductors such as nanowires and two-dimensional electron gases [159, 160], see Figure 3.4 (b). In all of these systems the strength of the proximity effect depends on the electronic properties of the normal material and the quality of the contacts to the superconducting leads. An explanation of the proximity effect can be given using Andreev reflections and Andreev bound states, which will be described in the next sections.

3.3.2 Andreev Reflections

At the normal-superconductor interface the main transport mechanism is that of Andreev reflections, which were first discovered by *A. F. Andreev* [161] and *de Gennes and Saint-James* [152, 153]. To understand the process of Andreev reflections a single normal metal/superconductor interface is considered. If the energy E of

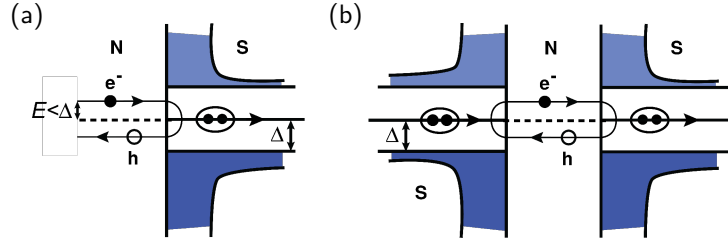


Figure 3.5: Andreev reflection and Andreev bound states. (a) Schematic of an Andreev Reflection at a normal/superconductor interface. An electron with energy $E < \Delta$ is impinging on a superconductor with superconducting gap Δ . The electron is reflected as a hole in the normal metal and a Cooper pair is added to the superconducting condensate. (b) Schematic of an Andreev Bound State in a SNS junction with leads of superconducting gap Δ . At energies below the superconducting gap, the Andreev Reflections process leads to constructive and destructive interference of the circulating electrons. Constructive interference leads to confined states called Andreev Bound States.

an electron in the normal region traveling towards the interface is larger than the superconducting gap Δ , $E > \Delta$, quasiparticle transport is possible. However, if the energy of the electron $E < \Delta$ single particle tunnelling is suppressed exponentially due to the gap in the density of states of the superconductor. This suggests that the electron has to be reflected at the interface. Nevertheless, an electron with momentum $\mathbf{k}$ can enter the superconductor by pairing up with a second electron from the normal metal of momentum $-\mathbf{k}$ and form a Cooper pair in the superconducting condensate. The second electron leaves a hole in the normal metal with momentum $\approx \mathbf{k}$, which is reflected and moving away from the superconductor, see Figure 3.5 (a). This process conserves energy and momentum in the normal metal. In this Andreev Reflection a Cooper pair is coherently transferred into the superconductor from the normal metal, leaving the normal metal with a charge deficit of $2e$.

3.3.3 Andreev Bound States

In a junction of a normal metal sandwiched between two superconductors (SNS junction) Andreev reflections happen continuously on both interfaces, as the reverse process of an electron-hole reflection can also occur. An electron impinging on the interface to the superconductor will be reflected as a hole. The reflected hole will impinge on the surface of the other superconductor and is conversely reflected as an electron. This results in electrons and holes circulating coherently

within the normal metal of the SNS junction, see Figure 3.5 (b). Similarly to Fabry-Pérot interference in quantum dots, electrons and holes bouncing back and forth in between the superconducting contacts can interfere constructively when a multiple of 2π is acquired as the total phase of their wave function during one cycle. This results in discrete states, called *Andreev bound states*. Upon the presence of an Andreev bound state (i.e. constructive interference), Cooper pairs are transferred from one superconductor to the other which results in a supercurrent passing through the junction. However, for destructive interference no current passes through the junction. There are two contributions to the total acquired phase, the phase acquired during Andreev Reflection and the phase picked up through the electron motion, which is negligible in the short junction approximation valid for short segments of ballistic conduction channels. In this regime it can be shown that two doubly degenerate Andreev bound states appear for each transport channel [147], with the energy given by

$$E_i(\varphi) = \pm \Delta \sqrt{1 - T_i \sin^2 \frac{\varphi}{2}}, \quad (3.9)$$

where T_i is the transmission probability of channel i and $\varphi = \varphi_1 - \varphi_2$ is the superconducting phase difference across the SNS junction. Each Andreev bound state comes in pairs of left and right circulating electron-hole trajectories with opposite energy. Josephson junctions are usually in their ground state, which is why the negative part of Equation 3.3.3 was used in Section 2.5.4. In order to see that the Andreev bound states carry the supercurrent through the junction, the phase dependent energy $E(\varphi) = \sum_i E_i(\varphi)$ of all channels i has to be considered. The supercurrent through the junction is now proportional to $\partial E / \partial \varphi$ [147] and can be expressed as

$$I(\varphi) = -\frac{2e}{\hbar} \sum_i \frac{\partial E_i}{\partial \varphi} = \frac{e\Delta}{2\hbar} \sum_i \frac{T_i \sin \varphi}{\sqrt{1 - T_i \sin^2 \varphi/2}}. \quad (3.10)$$

3.3.3.1 Critical Current

Considering a SNS junction with perfect transmission for all channels i , i.e. $T_i \rightarrow 1$, Equation 3.10 can be reduced to $I(\varphi) = I_c \sin \varphi/2$, with the critical current given by

$$I_c = \frac{e\Delta}{\hbar} \sum_i T_i = \frac{2\pi\Delta}{e} G, \quad (3.11)$$

where G is the conductance of the normal conductor segment given in Equation 3.4. This shows that the critical current in a SNS junction can be modulated by modulating the conductance of the normal conductor segment, which can be achieved, e.g. in carbon nanotubes with an external gate voltage.

3.3.4 Multiple Andreev Reflections

Until now, the transport through SNS junctions has not been considered under the influence of an external bias voltage V_{SD} . For simplicity, perfectly transmitting contacts and ballistic transport through the normal conductor will be assumed. The stability diagram of an SNS junction exhibits subgap conduction features, which appear as peaks in the differential conductance at finite bias voltages $V_{SD} = 2\Delta/ne$ with $n \in \mathbb{N}$. These features were first observed by *Taylor and Burstein* [162] and can be explained as a result of multiple Andreev reflection processes [163–165], see Figure 3.6.

Quasiparticle transport occurs for $V_{SD} \geq 2\Delta$ (see Figure 3.6 (a)), which corresponds to a step in the transport current and hence a peak in the differential conductance of a charge stability diagram. If the bias voltage is decreased to $V_{SD} = \Delta/e$ an electron from the left lead is Andreev reflected at the interface of the right lead, see Figure 3.6 (b). The resulting reflected hole will occupy the empty state in the left lead if $V_{SD} \geq \Delta/e$. This effectively transforms a quasiparticle from the left lead into a Cooper pair in the right lead. This process is called the first order Andreev reflection and results in a conductance peak at Δ/e in the stability diagram. The n -th order Andreev reflection is defined by the number of n Andreev reflections at the two interfaces of the leads, which each results in a conductance peak at $2\Delta/(n+1)e$. In Figure 3.6 (c) a second order Andreev reflection process

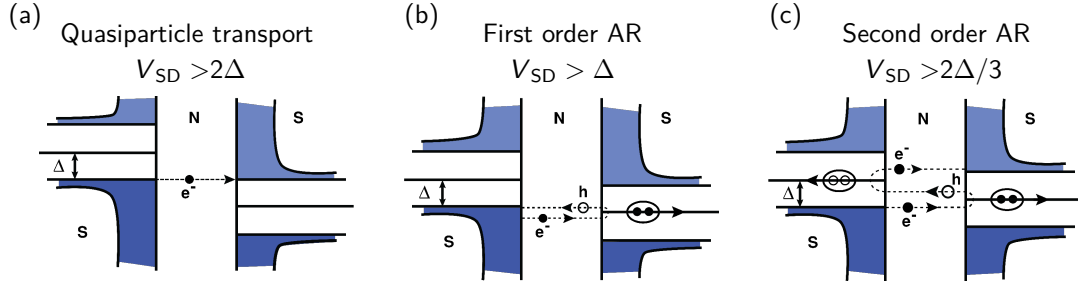


Figure 3.6: Multiple Andreev reflections. (a) Quasiparticle transport occurs when $V_{SD} > 2\Delta$. (b) First order Andreev reflection if $\Delta \leq V_{SD} \leq 2\Delta$. The electron undergoes one Andreev reflection, effectively transferring a Cooper pair. (c) Second order Andreev reflection if $2\Delta/3 \leq V_{SD} \leq \Delta$. A Cooper pair is transferred due to two successive Andreev reflections. Additionally a quasiparticle is transferred.

is depicted for $V_{SD} \geq 2\Delta/3$, where a current is created through two successive Andreev reflections. Hence, in this situation a Cooper pair and a quasiparticle are transferred through the junction giving rise to a conduction peak at $2\Delta/3e$. The contribution to transport of the n -th order Andreev reflection is proportional to $(T_i)^n$, with T_i being the transmission probability of channel i [137]. Therefore it is obvious that for non-ideal contacts, higher order Andreev reflections will have a lower conductance amplitude and hence be less visible.

Evidence of the different Andreev processes described above has been observed in carbon nanotube devices with superconducting leads and the measurements are presented in Chapter 7.

Part II

Experimental Principles

4

Fabrication and Design

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In order to perform experiments with micro- and nano-scale electronic devices, their structure has to be designed and fabricated. Specific design choices require different types of fabrication procedures. Hence with regard to well functioning devices, it is essential to understand the influence of certain design parameters on the device behaviour and choosing the correct fabrication procedure. For that reason in this chapter the essential micro- and nano-fabrication procedures used for

fabricating the devices mentioned in this thesis are summarised. In addition, design parameters for coplanar waveguide resonators as well as superconducting qubits are introduced. Finally, crucial parameters of $\lambda/2$ and $\lambda/4$ microwave resonators are characterised on real devices.

4.1 Fabrication Processes

Fabrication processes are a cornerstone of experimental circuit QED experiments. The reproducibility of device parameters and fabrication results are of highest importance and rely on a carefully conducted fabrication. In this section the different fabrication procedures and steps are explained in detail. For a step-by-step fabrication recipe of each of the devices presented in this thesis refer to Appendix A. Unless otherwise mentioned, all of the described fabrication steps were carried out in the cleanroom facilities in the DWB, the Clarendon Laboratory or at Begbroke, which are all facilities of the University of Oxford.

4.1.1 Chip Preparation

Initially a Si/SiO₂ or quartz wafer is diced into smaller 10x10 mm chips. The small scale is necessary for the carbon nanotube growth as the boat in which the chips are placed into the growth furnace can only hold chips of this size. Additionally, the standard chip holder cassette of the electron beam lithography machine is made to hold 10x10 mm chips accurately in position.

Chip Cleaning

For every cleanroom process it is essential to start with a clean substrate. Especially in our case, residue of any kind can greatly influence the fabrication outcome, the essential device parameters and their performance. Hence, the individual chips are initially cleaned in hot acetone (50 °C) in an ultrasonic bath and followed by an acetone and isopropanol (IPA) rinse. Subsequently, the chips are blow dried using a nitrogen (N₂) gun. This procedure gives a good initial clean of the substrate but does not completely remove organic residue, i.e. resist and similar chemicals. For the

quartz chips, as the wafer has previously been sputtered with a NbTiN thin film, a second cleaning step is performed in an oxygen plasma asher (*Quorum Technologies, PT7160 Plasma Barrel Reactor*). The Si/SiO₂ chips however are put in a piranha etch (1:3, hydrogen-peroxide:sulfuric-acid) for 10 minutes. Subsequently, the Si chips are rinsed with DI water, acetone and IPA and afterwards blow dried with N₂. The chips are now clean and ready for their next fabrication step which is either a lithography step, which requires resist deposition, or the carbon nanotube growth.

Resist Deposition

The lithography step requires a resist layer which is sensitive to either UV-light or electron beam exposure in order to expose a pattern onto the chip. Depending on the type of the lithography and the process after the pattern exposure, different types of resists are used, refer to specific device recipes in Appendix A. However, the deposition of a single layer or a double layer of any type of resist follows the same procedure which is explained in the following paragraph.

Before spinning the resist onto the chip it is baked on a hot plate at 120 °C in order to get rid of water molecules attached to the surface of the substrate to improve adhesion of the resist. Subsequently, the chip is mounted onto the chuck of a spin coater using a vacuum pump. The required resist is then dropped onto the chip until it is fully covered using a micro-pipette. The chip is then spun with a few 1000 rotations per minute (rpm) for a time of 30 – 60 s (resist dependent) to achieve a uniform resist film. An initial low rpm ramp for 10 – 15 s is used to get rid of excess resist. The main spinning speed, i.e. rpm, determines the final thickness of the resist (cf. Appendix A and individual resist data-sheet). After spinning, the chip is baked on a hot plate at 110-180 °C for 1-10 minutes. For a double layer of resist the chip is again mounted on the chuck of the spin-coater and the procedure repeated.

4.1.2 Photolithography

Photolithography exploits UV-radiation in order to write a pattern in photo-resist sensitive to UV-light. A semi-automatic mask-aligner from EVG (*EVG-620*) is

used to align the photomask containing the pattern to the chip. Once aligned, the chip is brought into contact with the mask and subsequently exposed with UV-light. Since light is used in this process, the minimum feature size is diffraction limited and dependent on the contact mode between the mask and the chip. With a vacuum-hard contact¹, features down to 2 μm can be achieved. For the devices used in this thesis two different type of photolithography processes have been used, i.e. positive and image-reversal exposure processes. These will be described in detail in the next two paragraphs.

Positive Exposure

If a positive photo-resist is used for the lithography, the pattern which is exposed to UV-light will be directly transferred to the chip, see Figure 4.1 (a)-(c). Positive photo-resist is mainly used for etching processes as these exhibit a vanishing undercut, i.e. positive wall-profile after development. The undercut and the resist wall-profile can be controlled with the exposure dose, which can be set in $\mu\text{J}/\text{cm}^2$ on the EVG machine. If the positive resist is used for etching, a resist should be used which exhibits a smaller selectivity to the etchant than the material (substrate or metal film) to be etched. In our case positive photo-resist was used for patterning coplanar waveguide resonators on NbTiN thin films on quartz via a reactive-ion-etch (RIE). For this process S1813 photo-resist was used for the patterning, exact recipe see Appendix A.

Image-Reversal Exposure

For processes involving the evaporation of thin metal films and their subsequent lift-off, a negative resist wall-profile, i.e. undercut, is necessary in order to achieve clean metal edges after lift-off. For this process, an image-reversal photo-resist is used and the mask carries the pattern in opposite polarity to a positive resist mask, see Figure 4.1. The initial exposure follows the same procedure as for a positive photo-resist and its behaviour is the same. A subsequent image-reversal bake crosslinks

¹The chip is pressed with a certain force against the mask while a vacuum is created between the mask and the wafer chuck.

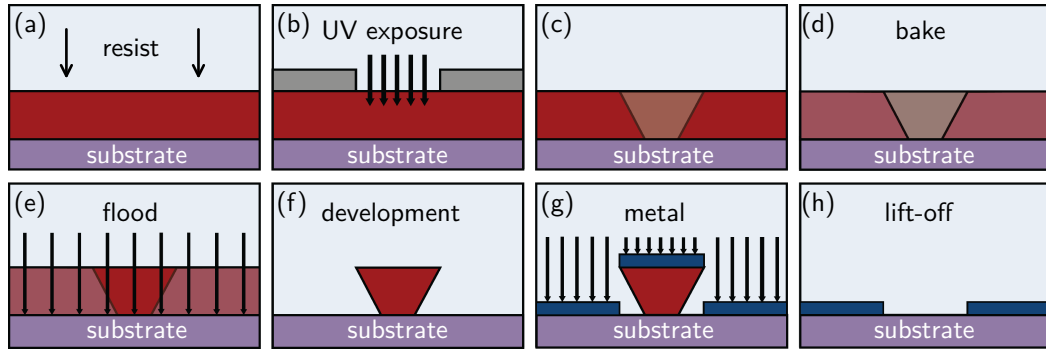


Figure 4.1: Photolithography with image-reversal resist. (a) Resist is spun on the substrate. (b) After baking, a mask is brought into contact and the pattern exposed with UV-light. (c) Exposed resist is soluble in developer and hence behaves like a positive photo-resist with typical positive wall-profile. Processes with positive resist would carry on with development and subsequent etch/metal deposition, followed by resist stripping. (d) Image reversal bake, which crosslinks exposed resist and keeps unexposed resist photo-active. (e) Flood exposure of the whole substrate without a mask. (f) Development of the flood exposed resist in corresponding developer, leaving the pattern exposed in (b) with a clear undercut. (g) Metal evaporation with electron beam evaporator. (h) Resist lift-off in 50 °C hot acetone.

the exposed resist area while the unexposed resist stays photo-active. A flood-exposure without a mask is used to make the previously un-exposed resist soluble in the developer. This turns the positive resist wall-profile of the first exposure into a negative resist wall-profile with the inverted pattern, cf. Figure 4.1 (c)-(f). The undercut is determined by the dose of the first exposure. Here we used an image-reversal process in order to pattern coplanar waveguide resonators on Si/SiO₂ chips with subsequent Al evaporation and lift-off. The photo-resist used was AZ 5214E, exact recipe see Appendix A.

4.1.3 Electron Beam Lithography

In a positive electron beam (ebeam) lithography process, patterns are written in a resist based on PMMA (polymethyl methacrylate) via the photo scission of the polymer backbone using the irradiation of a beam of high energy electrons, see Figure 4.2 for process flow. In the following development process the exposed resist of lower molecular weight is removed chemically. The PMMA resist is available in different molecular weights (495K and 950K) which corresponds to a higher resolution in the writing process. The thickness of the resist can be controlled via

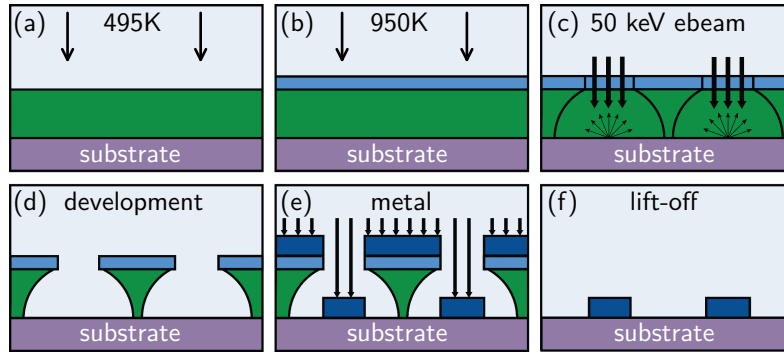


Figure 4.2: Electron beam lithography process. (a) A PMMA 495K resist layer is spun on the substrate. (b) After baking a thin, higher resolution PMMA 950K resist is spun onto the first layer and baked subsequently. (c) Electron beam lithography of the desired pattern with 50 keV electrons. (d) Development of the exposed pattern with MIBK in IPA (1:3). (e) Metal deposition with an electron beam evaporator. (f) Resist lift-off in 50 °C hot acetone.

its concentration in the solvent anisole and the spinning speed. The dose of the ebeam process, set in $\mu\text{C}/\text{cm}^2$, determines the feature sharpness as well as the undercut, which is achieved through backscattered electrons and the proximity effect, see Figure 4.2 (c). An ebeam process based on PMMA allows for a resolution of a few 10s nm. For the device fabrication in this thesis an ebeam writer from JEOL (*JEOL 5500FS*) with an acceleration voltage of 50 keV is used. The patterns can be drawn in any CAD software tool that supports the gds format, which is used by the JEOL machine. The JEOL ebeam offers three different apertures for various feature precision as well as two-point chip and three-point device alignment. By using these alignment procedures, the ebeam adjusts for linear and rotational offsets of the chip to the design coordinates. After manual calibration and focussing of the ebeam to the specific aperture, the writing process is automated as the ebeam scans the pattern across the chip.

4.1.4 Development

After the exposure the resist is developed in the corresponding developer which provides a very high selectivity between the exposed and the unexposed area. Resist dependent, after 40 s to a few minutes of the chip being agitated in the developer the exposed resist is removed and the pattern is visible on the chip, cf. Figure 4.1 (f)

and Figure 4.2 (d). The development process is stopped in either DI water or IPA. Subsequently the N₂ gun is used to blow dry the chip. The development process can leave some resist residue in the exposed areas, i.e. the undercut regions. In order to remove this residue either a gentle UV ozone clean (*ProCleaner Plus*, *BioForce Nanosciences*) or a stronger oxygen plasma ashing is performed. For the resist specific development recipe refer to Appendix A.

4.1.5 Evaporation

Most of the devices presented in this thesis rely on metal being deposited on the substrate which was previously patterned with either photo- or ebeam-lithography. Metals, such as Au, Al, Ti and Pd can be evaporated by heating them up far past their melting point using an accelerated electron beam. In order to evaporate thin metal films of good quality, a constant low evaporation rate of 0.25 – 0.5 nm/s and a good vacuum with pressures below 10^{-6} – 10^{-7} mbar are necessary. Here we used a home-built electron beam evaporator, that monitors the evaporation rate via a crystal counter and the main chamber is pumped by a cryo-pump. Pre evaporation pressures $< 10^{-7}$ have been achieved and evaporation pressures ranged from $< 10^{-6}$ to $< 10^{-5}$ depending on the material being evaporated.

4.1.6 Lift-Off

After the evaporation the remaining resist needs to be stripped from the chip. As most of the chips also contained carbon nanotubes, the least invasive lift-off technique based on 50 °C hot acetone was used for all devices, see Figure 4.1 (h) and Figure 4.2 (f). The chip was left in the hot acetone bath for 1-12 hours without agitation, which for larger features was enough to almost completely lift the resist/metal stack off the chip. Afterwards, a pipette was used to agitate the acetone across the film in order to aid the lift-off of finer features. Before the chip was dried, it was placed in a shallow beaker filled with fresh IPA and inspected under an optical microscope. Once it was made sure that all residual metal and resist came off completely, the chip was rinsed in IPA and blow dried with the N₂

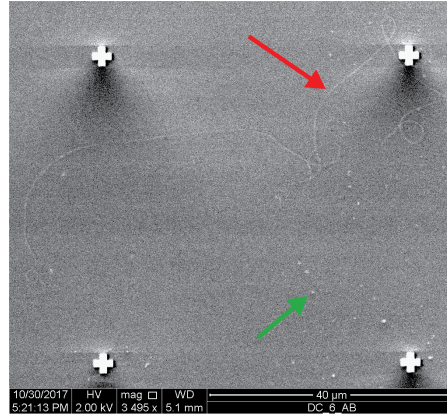


Figure 4.3: Scanning electron microscope image of the Si substrate showing a carbon nanotube (red arrow) growing from the FeMo catalyst (green arrow) and chip alignment marks (white crosses).

gun. The chips are now ready for further inspection at room-temperature, imaging, wire-bonding and measurements at cryogenic temperatures.

4.1.7 Reactive-Ion-Etching

For the etching of NbTiN films dry reactive-ion-etching (RIE) with SF_6 ions in an *Oxford Instruments Plasmalab 80* was used. In a reactive ion etcher gas is ionised by applying a strong RF electromagnetic field to the wafer-stage. The ions are accelerated towards the chip and collide with the material to be etched. The etch happens due to kinetic energy transfer but also due to the chemical reaction of the ions with the surface of the materials. Due to its directionality, RIE can deliver very anisotropic etch profiles. The ions can be chosen such that the RIE process offers a higher selectivity for the material to be etched than for the etch mask, i.e. photo-resist. In order to make the resist resilient against the RIE a hard-bake after the development is necessary. This also reflows the resist which leads to a more vertical resist wall-profile. After the RIE the resist is very difficult to be removed with only acetone, hence stronger stripping solvents such as DMSO and NMP-based resist strippers were used in order to remove the remaining resist.

4.1.8 Carbon Nanotube Growth

The carbon nanotubes used for the devices presented in this thesis were kindly grown by Andreas Baumgartner from the Quantum- and Nanoelectronics group at the University of Basel. Single-wall carbon nanotubes are grown on the substrate via chemical vapour deposition (CVD) from an iron-based nanoparticle catalyst. The catalyst is made from iron molybdenum (FeMo) nanoparticles dissolved in an IPA solution. The solution is spin-coated onto the substrate and the grade of the IPA dilution allows the tuning of the carbon nanotube growth density. In order to avoid clustering of the nanoparticles and favour growth of small diameter carbon nanotubes, the catalyst is sonicated previous to coating the substrate. The chips are then placed into a CVD furnace and heated to the desired growth temperature $T = 950\text{ }^{\circ}\text{C}$ in an argon (Ar) atmosphere. Once the growth temperature is reached, the nanotubes are grown from a methane and hydrogen (H_2) atmosphere. The methane is dissociated by the catalyst and serves as a carbon source for the nanotube growth. The hydrogen reduces the excess carbon in order to avoid bundle and amorphous carbon growth. After ~ 10 minutes the chips are cooled to room-temperature in an Ar/ H_2 atmosphere. This recipe favours the growth of single-wall carbon nanotubes, details can be found in the PhD-Theses by *J. Furer* [166] and by *J. B. Gramich* [167].

4.2 Coplanar Waveguide Resonator Design and Characterisation

Coplanar waveguide resonators (CPWR) are an integral part of two-dimensional on-chip implementations of circuit QED as these are used for control and readout of the coupled system, i.e. superconducting qubits or spin ensembles. The resonator frequency f_0 , its internal Q_{int} and external quality factor Q_{ext} as well as its characteristic impedance Z_0 are important device parameters. In this section we will discuss the design considerations for $\lambda/2$ and $\lambda/4$ resonators. In principle a standard single layer photolithography process can be used to fabricate these

resonators and details can be found in the previous section. For a more in depth analysis of coplanar waveguide resonators refer to work from *Goppl et al.* [168].

4.2.1 Design

Coplanar waveguides (CPW) are the two-dimensional equivalent of a coaxial cable, where the centre conductor and the ground are in the same horizontal plane. These structures allow for localised fields with a small mode volume which is beneficial for the coupling to other systems placed in close vicinity.

CPWs can be fabricated from a thin metallic (thickness d) centre conductor of width $w = a$, which is surrounded by two ground-planes separated by a distance b , all on top of a dielectric material with dielectric constant ϵ_d and loss tangent $\tan \delta$, see Figure 4.4 (left). The characteristic impedance Z_0 of CPWs is solely defined by the ratio a/b , which allows a scaling of the CPW dimensions from microns (intense and localised fields) to millimetres (interfacing with printed circuit boards), while keeping Z_0 constant. The characteristic impedance of CPWs is given by

$$Z_0 = \sqrt{\frac{L_l}{C_l}}. \quad (4.1)$$

Here L_l is the inductance and C_l the capacitance per unit length of the resonator. These can be calculated using conformal mapping techniques such that

$$L_l = \frac{\mu_0 K(k'_0)}{4K(k_0)} \quad (4.2)$$

$$C_l = \frac{4\epsilon_0\epsilon_{\text{eff}}K(k_0)}{K(k'_0)} \quad (4.3)$$

where $\epsilon_{\text{eff}} \sim (1 + \epsilon_d)/2$, K denotes the complete elliptic integral of the first kind with the arguments $k_0 = a/b$ and $k'_0 = \sqrt{1 - k_0^2}$ [117–119, 169]. Since all of the measurement equipment exhibits an impedance of $50 \, \Omega$, it is desirable to choose the same impedance for the CPW. For a $d = 100 \, \text{nm}$ thick aluminium layer on a Si/SiO₂ substrate, $a = 10 \, \mu\text{m}$ and $b = 20 \, \mu\text{m}$ are chosen for the CPWs for good $50 \, \Omega$ impedance matching and its compliance with standard photolithography processes.

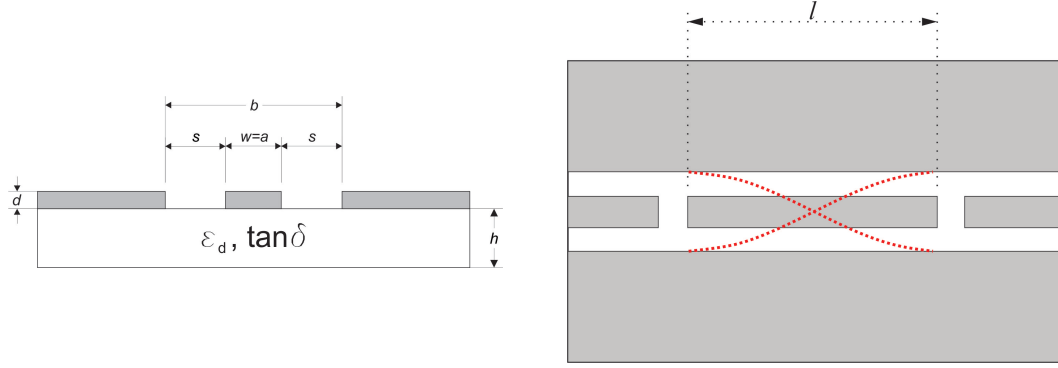


Figure 4.4: Geometry of a coplanar waveguide (left) and a coplanar waveguide resonator (right). The grey shaded region is the metalised part, i.e. superconductor, of thickness d and the white region is the dielectric substrate with a dielectric constant ϵ_d and a loss tangent $\tan \delta$.

4.2.1.1 Half-Wave Resonators

Introducing two gaps in the centre conductor of the CPW separated by a distance l , it is possible to form a cavity, where the gaps serve as “mirrors” such that a standing wave can form inside the resonator. The fundamental resonance frequency of such a CPW resonator is given by

$$f_0 = \frac{c}{\sqrt{\epsilon_{eff}}} \frac{1}{2l}, \quad (4.4)$$

where c is the speed of light and l is the length of the centre conductor between the two gaps, see Figure 4.4 (right). The resonator is capacitively coupled to the input/output transmission line and can be measured in transmission. The coupling can be increased by reducing the gap in the centre conductor or by increasing the area of the resonator facing the feedlines. This way the external coupling can be engineered specifically for the required experiment [168].

In general the quality factor (Q-factor) of a resonator is defined by

$$Q = \frac{f_r}{\delta f} = \frac{\text{Energy stored}}{\text{Power lost}}. \quad (4.5)$$

A measurement of the resonator spectrum yields the loaded quality factor Q_L which is a combination of Q_{int} and Q_{ext}

$$\frac{1}{Q_L} = \frac{1}{Q_{\text{int}}} + \frac{1}{Q_{\text{ext}}}. \quad (4.6)$$

The internal Q-factor, Q_{int} , of CPWRs is primarily reduced by dissipative photon losses, via dielectric, radiative and resistive interactions, which contribute to Q_{int} in the following fashion

$$\frac{1}{Q_{\text{int}}} = \frac{1}{Q_{\text{cond}}} + \frac{1}{Q_{\text{diel}}} + \frac{1}{Q_{\text{rad}}}, \quad (4.7)$$

where $Q_{\text{cond}} = (\omega L)/R_1$, $Q_{\text{diel}} = 1/(\tan \delta)$ and $Q_{\text{rad}} \approx 3.5 (l/a)^2$ [170]. If the metal of the CPWR is replaced by a superconductor and cooled below its critical temperature the resistive losses decrease to a minimum and only the dielectric losses dominate, as due to the chosen dimension (see Section 4.2.1) the radiative losses are quite small. The external Q-factor, Q_{ext} is solely defined by the coupling capacitances C_κ to the external measurement lines [119, 168].

4.2.1.2 Quarter-Wave Resonators

CPWs capacitively coupled to a transmission line at one end and grounded on the other end yield a $\lambda/4$ resonator. The electric field distribution inside such a CPWR has a node at the grounded end and an antinode facing the transmission line. The fundamental resonance frequency f_0 of the resonator can fit a quarter wavelength ($l = \lambda/4$) into the resonator. Hence, the resonance frequency is given by

$$f_0 = \frac{c}{\sqrt{\epsilon_{\text{eff}}} 4l}. \quad (4.8)$$

The external Q-factor, i.e. the coupling strength to the transmission line, can be engineered by changing the distance between the resonator and the transmission line and the length over which the two are coupled to each other. This can be modelled as a simple coupling capacitance C_κ which for the fundamental mode results in

$$Q_{\text{ext}} = \frac{\pi}{4} \frac{1}{\omega_0^2 C_\kappa^2 Z_0 Z_{\text{port}}}, \quad (4.9)$$

where $\omega_0/2\pi = f_0$, Z_0 the characteristic impedance of the resonator and Z_{port} the characteristic impedance of the transmission line [117, 171]. For devices presented in Chapter 8 *Ansyz HFSS* and *Ansyz Maxwell* simulations have been used to estimate Q_{ext} and resonators with two different coupling capacitances were fabricated.

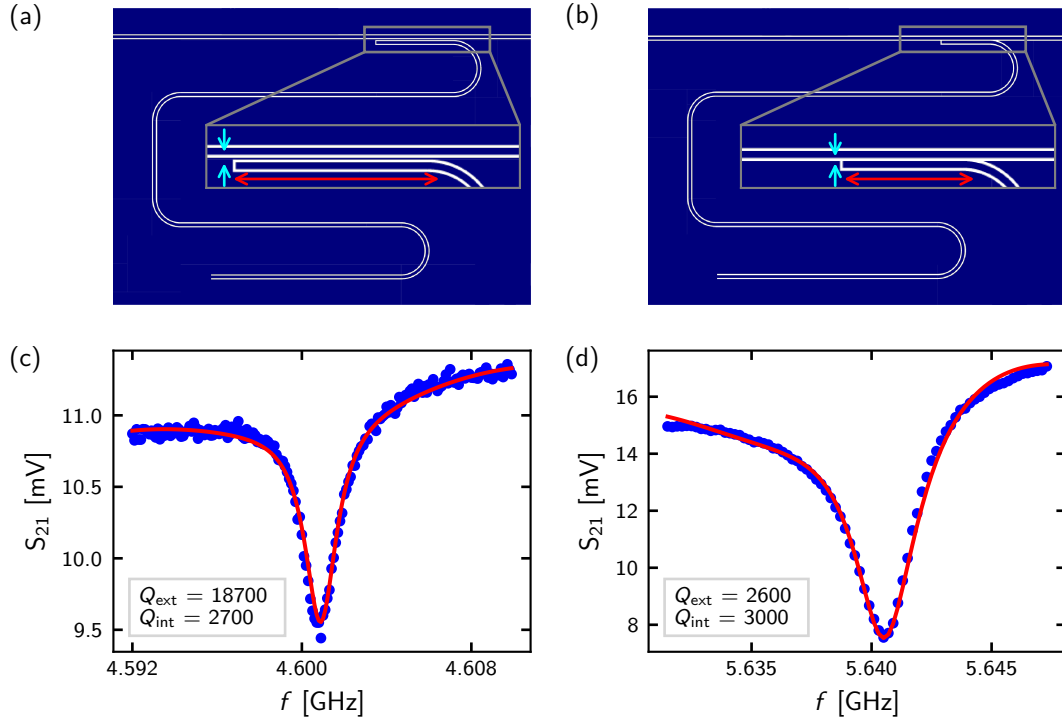


Figure 4.5: Quarter-wave resonators with different Q_{ext} . (a) & (b) Rendered CAD drawing of a $\lambda/4$ resonator coupled to a transmission line. Inset: Enlarged view of the individual resonator coupling to the transmission line. Cyan arrows indicate the distance between the resonator and the transmission line, (a) $22 \mu\text{m}$ and (b) $15 \mu\text{m}$. Red arrows indicate the coupling length between the resonator and the transmission line, (a) $300 \mu\text{m}$ and (b) $185 \mu\text{m}$. (c) Transmission spectrum of a single $\lambda/4$ resonator with external coupling shown in (a). Transmission is fitted with Eq. 4.14 resulting in $Q_{\text{ext}} = 18700$. (d) Transmission spectrum of a single $\lambda/4$ resonator with external coupling shown in (b). Transmission is fitted with Eq. 4.14 resulting in $Q_{\text{ext}} = 2600$.

Figure 4.5 (a)-(b) show rendered CAD drawings of two individual $\lambda/4$ resonators with different coupling capacitances. The difference in Q_{ext} can be extracted by analysing the transmission spectrum of the resonators and fitting it with Equation 4.14, see Figure 4.5 (c)-(d). The coupling shown in Figure 4.5 (a) inset yields $Q_{\text{ext}} = 18700$ and similarly the coupling in Figure 4.5 (b) yields $Q_{\text{ext}} = 2600$.

4.2.2 Resonator Spectrum

Here the measured resonator spectrum of $\lambda/2$ and $\lambda/4$ resonators is analysed and key resonator parameters extracted.

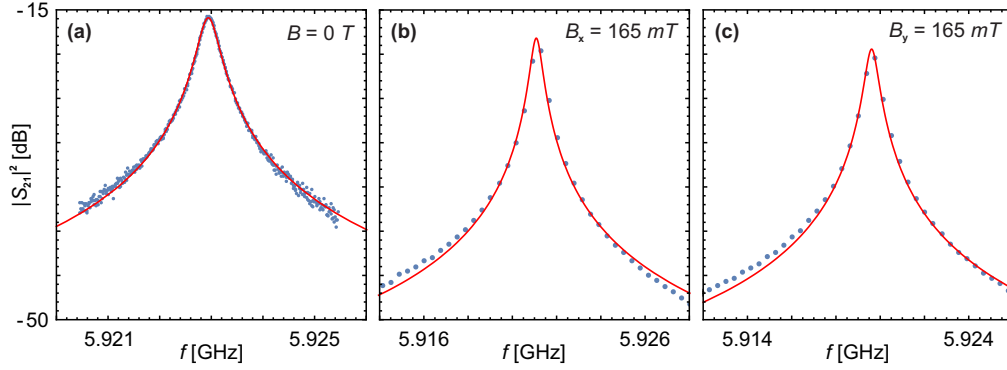


Figure 4.6: Points: Microwave transmission of the $\lambda/2$ resonator at different magnetic field strengths and orientations. The magnetic field orientations x and y correspond to the two major in-plane axes of the resonator chip. Lines: Fits to Eq. (4.10), with fit parameters listed in Tab. 4.1.

4.2.2.1 Half-Wave Resonators

Measuring the power transmission spectrum $|S_{21}|^2$ of a $\lambda/2$ resonator around its resonance frequency f_0 yields a transmission peak of the resonator. The transmission peak follows a Lorentzian line shape

$$|S_{21}(\omega)|^2 = A_0 \frac{\kappa^2}{(\omega - \omega_0)^2 + \kappa^2}, \quad (4.10)$$

where A_0 is the transmitted peak power, $\omega_0/2\pi = f_0$ the resonance frequency and $2\kappa \equiv \omega_0/Q_L$ the linewidth of the resonator. Figure 4.6 shows transmission spectra of a $\lambda/2$ resonator made from NbTiN on quartz (see Chapter 6 for design details) at zero and finite magnetic fields. In order to extract the resonance frequency f_0 and the loaded quality factor Q_L of the resonator the measured transmission $|S_{21}(\omega)|^2$ was fitted with Equation 4.10. The resonator parameters yielded by the fits are presented in Table 4.1.

	B^{ext}	f_0	$\kappa/2\pi$	Q_L
-	0 T	5.923 GHz	195 kHz	15200
x	165 mT	5.921 GHz	251 kHz	11800
y	165 mT	5.919 GHz	283 kHz	10500

Table 4.1: Parameters extracted from Fig. 4.6

4.2.2.2 Quarter-Wave Resonators

To measure the spectrum of $\lambda/4$ resonators they need to be capacitively coupled to a transmission line, cf. Section 4.2.1.2. A single transmission line allows multiple $\lambda/4$ resonators of different frequencies to be characterised. The microwave transmission spectrum (S_{21}) of the transmission line then exhibits an absorption dip at the resonance frequency of each individual resonator, see Figure 4.7 (a). Near resonance the spectrum of a single $\lambda/4$ resonator is described by the complex $S_{21}(\omega)$ parameter given by

$$S_{21}(\omega) = \frac{S_{21}^{\min} + 2iQ_L \left(\delta x + \frac{\delta\omega}{\omega_0} \right)}{1 + 2iQ_L \delta x}, \quad (4.11)$$

where $\omega_0/2\pi = f_0$, ω the applied frequency, $S_{21}^{\min} = Q_{\text{ext}}/(Q_{\text{ext}} + Q_{\text{int}})$, $1/Q_L = 1/Q_{\text{ext}} + 1/Q_{\text{int}}$ and $\delta x = (\omega - \omega_0)/\omega_0$ [149, 172]. The parameter $\delta\omega$ includes an asymmetry in the line shape, which is attributed to reflections in the transmission line [172]. For $\delta\omega = 0$ a symmetric line shape is recovered. However, the model presented in Equation 4.11 does not include any distortion of the resonance due to the measurement setup, i.e. reflections in the microwave lines resulting from impedance mismatches, etc.

The effects of the measurement setup can be included into Equation 4.11 in the following fashion [149]. The semi-rigid coax cables used in the setup cause attenuation of the microwave signal which can be approximated by a constant part a and a linearly frequency dependent part b which have a direct effect on the complex amplitude

$$a + b \cdot \delta x. \quad (4.12)$$

Due to the optical length of the cables a frequency dependent phase v is acquired through the cables. Additionally, the initial phase of the signal generators is not known, which adds a constant phase offset Θ . The phase can hence be corrected by

$$e^{i(v\delta x + \Theta)}. \quad (4.13)$$

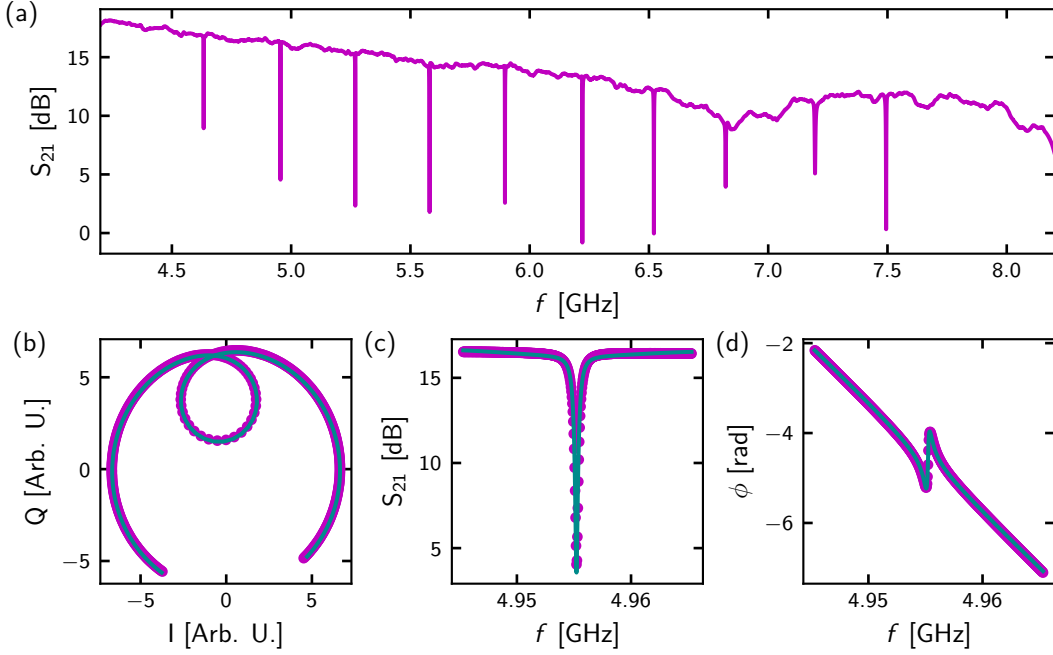


Figure 4.7: Transmission spectrum of $\lambda/4$ resonators. (a) Multiplexed S_{21} measurement of 10 $\lambda/4$ resonators capacitively coupled to a transmission line, for chip design see Figure 8.1 (b). Each resonator shows up as a clear resonance dip in the spectrum. The background is due to linear attenuation and unpredictable resonances in the measurement setup. (b)-(d) Different representations of transmission data (purple points) of a single $\lambda/4$ resonator capacitively coupled to a transmission line. Cyan lines are best-fit curves according to Eq. 4.14. (b) I-Q quadrature representation, (c) transmission amplitude and (d) transmission phase data.

The last correction term is due to frequency independent power transfer from the source to the acquisition port without crossing the sample, which leads to a constant offset $\mathcal{I}_c + i\mathcal{Q}_c$ in the complex plane.

The resonance of a $\lambda/4$ resonator coupled to a transmission line embedded in a complex measurement setup can now be described by the full model

$$S_{21,\text{meas}} = (a + b \cdot \delta x) S_{21}(\omega) e^{i(v\delta x + \Theta)} + (\mathcal{I}_c + i\mathcal{Q}_c). \quad (4.14)$$

This equation is used to fit the resonator response in the complex IQ plane, which in turn gives accurate results for the characteristic values of the resonator, i.e. f_0 , Q_{int} and Q_{ext} . In Figure 4.7 (b) the IQ quadrature data of a single resonator is shown and fit by Equation 4.14, yielding $f_0 = 4.955$ GHz, $Q_{\text{int}} = 30200$ and $Q_{\text{ext}} = 9700$. As can be seen from Figure 4.7 (c)-(d), the fit to the quadrature data also produces good fits to the magnitude and phase response of the resonator.

4.3 Qubit Design

Superconducting qubits are basically anharmonic LC oscillators that consist of only two circuit elements, a capacitor and a non-linear inductor, i.e. Josephson junction. These two circuit elements have a charging energy E_C and a Josephson energy E_J associated with them which make up the Hamiltonian of the qubit, cf. Section 2.5. In the following section the design aspects for these two energies are considered in order to faithfully design and predict qubit parameters prior to their fabrication and measurement. Since the capacitance of the Josephson junction C_J is small compared to the shunt capacitance of the qubit, E_C and E_J can be considered independently.

4.3.1 Charging Energy and Capacitance Network

The charging energy E_C of the qubit as well as the coupling strength g between the resonator and the qubit depend on the capacitance network formed by the resonator, the qubit and the ground-plane. An electrostatic solver, *Anslys Maxwell* is used in order to simulate a 3-dimensional model of the qubit design, see Figure 4.8 (a). In this model a small box around the qubit consisting of the dielectrics Si/SiO₂, vacuum and the superconducting parts (Al) of the circuit is simulated. Since the qubit is shorted to ground, the circuit model effectively consists of only 3 electrodes, i.e. the ground plane (1), the qubit island (2) and the resonator centre conductor (3). The software package of the electrostatic solver uses finite elements in order to find a solution for the full capacitance matrix C_{ij} of the 3 electrodes $i, j \in \{1, 2, 3\}$. Typical values are $C_{12} = 30.2$ fF and $C_{23} = 7.9$ fF. The capacitance between the resonator and the ground-plane $C_{13} \gg C_{12}, C_{12}$. It is possible to show that the reduced capacitance network shown in Figure 4.8 (c) is equivalent to the full network shown in Figure 4.8 (b) by introducing an effective shunt

$$C_s = C_{12} \quad (4.15)$$

and gate capacitance

$$C_g = C_{23}. \quad (4.16)$$

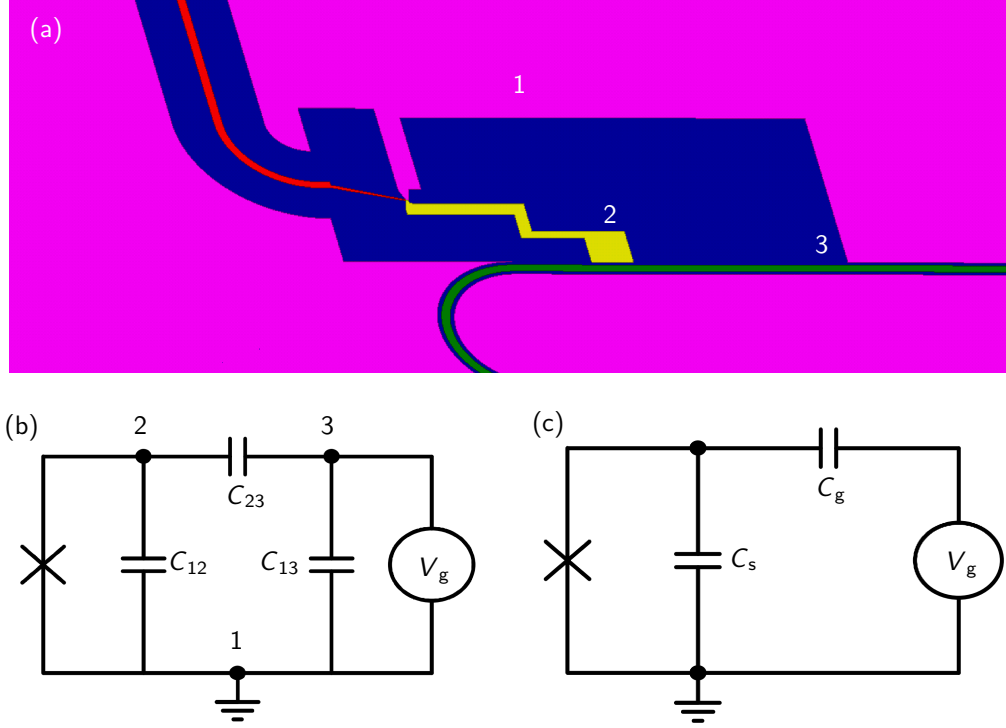


Figure 4.8: Simulation for qubit design. (a) Image of an exemplary qubit design in *ANSYS Maxwell* used for electrostatic simulations. The three components, i.e. ground (pink), island (yellow) and resonator (green) are assigned to the indices $i, j = 1, 2$ and 3. (b) Equivalent circuit representation of (a) including all mutual capacitances. (c) Reduced equivalent circuit diagram of the qubit.

Hence, the charging energy of the qubit is given by $E_C = e^2/C_\Sigma$ where $C_\Sigma = C_g + C_s = C_{12} + C_{23}$. The voltage division

$$\beta = \frac{V_\Sigma}{V_g} = \frac{C_g}{C_\Sigma} = \frac{C_{23}}{C_{12} + C_{23}} \quad (4.17)$$

describes the fraction of the voltage that drops across the Josephson junction if a voltage V_g is applied between the resonator and ground. For full details of the capacitance network analysis of a superconducting qubit refer to *Koch et al.* [104].

The coupling of the qubit's ground to first excited state transition to the resonator is quantified by the vacuum Rabi frequency which is given by

$$g = 2\beta e V_0 \sqrt{\frac{1}{2} \left(\frac{E_J}{8E_C} \right)^{1/4} \frac{1}{\hbar}} \quad (4.18)$$

where $V_0 = \sqrt{\hbar\omega_r/(2C)}$ is the rms zero point voltage of the resonator and C the effective resonator capacitance, for details see Chapter 2.

4.3.2 Josephson Energy

The Josephson energy of the carbon nanotube Josephson junctions is in situ tunable by changing the electrostatic potential inside the carbon nanotube with a voltage applied to a nearby gate. The Josephson energy is given by

$$E_J = \frac{\hbar I_c}{2e}, \quad (4.19)$$

where I_c is the critical current of the junction and e the single electron charge. The critical current of the junction is tunable by tuning the conductance of the carbon nanotube channel with an applied gate voltage, cf. Section 3.3.3.1. The maximally achievable critical current in carbon nanotube Josephson junction depends on the induced superconducting gap Δ and the contact transparency $\mathcal{T}$ to the carbon nanotube. Both of these parameters are heavily dependent on a robust and reproducible fabrication of such devices. Numerous junctions and different superconducting bi-layers have been fabricated and tested to optimise the fabrication procedure [137].

5

Measurement Setup

Contents

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In this chapter the measurement setup and measurement techniques used throughout the experiments of this thesis are described. For testing purposes some of the samples are measured at room-temperature. However, all experiments exhibiting quantum behaviour need to be cooled to low temperatures and hence require cooling systems as well as more elaborate measurement circuitry, both of which are described in the following sections.

5.1 Room-Temperature Setup

The DC samples for the experiments presented in Chapter 7 as well as the qubit samples for experiments in Chapter 8 were tested and pre-selected at room-temperature. For this the resistance of each of the devices was measured using a DC probe station from *Semiprobe* (Model: Lab Assistant) in combination with a *Keithley 2450 Sourcemeter*. With the probe station, fine probe needles can be carefully lowered onto the bondpads of the leads of the devices and their resistance measured via the sourcemeter. This allowed the selection of appropriate devices for the more elaborate low-temperature measurements.

5.2 Cryogenic Setup

All significant experiments presented in this thesis are performed at temperatures around 10 – 20 mK. This low temperature is necessary in order to operate the microwave resonators in their ground state ($k_B T < \hbar\omega$) and to reach an electron temperature (T_e) as low as possible, minimising effects of thermal broadening in the quantum dot devices.

5.2.1 Dilution Refrigerator

In order to perform experiments at very low temperatures the measurements were carried out in a cryogen free dilution refrigerator *Triton 200* from *Oxford Instruments*, with a specified base temperature of 10 – 20 mK¹. Throughout the work of this thesis two different Triton 200 systems were used, one standard version (depicted in Figure 5.1 (a), used for experiments in Chapter 8) and the second one a bottom-loader equipped with a (1, 1, 6) T vector magnet (used for experiments in Chapter 6 and Chapter 7). Low temperature is achieved exactly the same way in both systems. A pulse-tube cooler cools the first two temperature stages and a mixture of He₃ and He₄ is used to cool the last three stages. Each stage consists of a circular, gold coated copper plate which is utilised to thermalise all measurement

¹The calibration of the ruthenium oxide temperature sensor is only valid down to ~ 20 mK, but lower temperatures were initially measured with nuclear-orientation thermometry.

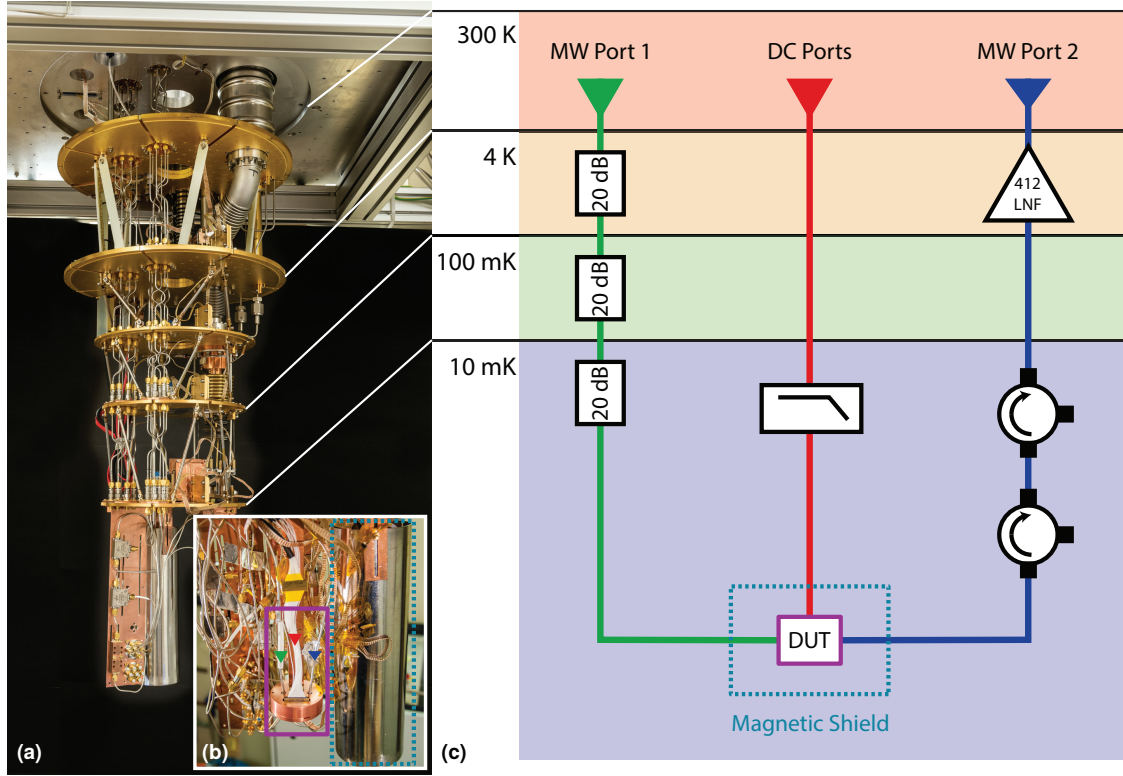


Figure 5.1: Cryogenic measurement setup. (a) Dilution refrigerator *Triton 200* from *Oxford Instruments* equipped with multiple microwave measurement circuits as presented in (c), each accessible via microwave switches from *Radiall*. The different significant temperature stages are indicated. (b) Sample holder (purple) mounted on the mixing chamber stage of the dilution refrigerator. The different ports for microwave (green and blue) and DC (red) measurements are indicated as in (c). The sample is additionally placed into a magnetic shield (cyan dashed). (c) Schematic of the microwave and DC measurement circuits. The microwave input line (green) is attenuated with three 20 dB attenuators at different temperature stages. The device under test (DUT) is mounted at the lowest temperature stage. The microwave output line (blue) has two circulators straight after the sample and a HEMT amplifier at the 4 K stage. The DC lines (red) are thermalised at each stage and a lowpass filter on the mixing chamber.

lines at each temperature. The measured sample is thermally anchored at the lowest temperature stage, i.e. mixing chamber (MC), see Figure 5.1 (b).

5.2.2 Microwave Measurement Lines

The microwave measurements were performed at GHz frequencies, which requires coaxial cables to carry the signal from room-temperature to the mixing chamber and back. However, it is crucial to extinguish any source of noise from higher temperatures as this would have a detrimental effect to the quantum behaviour

of the measured samples. At GHz frequencies $1/f$ noise is usually a minor issue. However, Johnson noise due to the thermal motion of the charge carriers in the conductor is dominant. The noise power spectral density can be assumed to be linear in temperature and conductor resistance [118]. Since the signal powers needed in the measurements is very low, the most effective way to suppress thermal noise is to simply attenuate the signal together with the noise. Therefore, the input line of the microwave measurement setup in the dilution refrigerator is attenuated by 20 dB at the 4 K, 100 mK and 10 mK temperature stage, see Figure 5.1 (c). The coaxial cables used in the microwave input line are made from stainless steel (inner and outer conductor) and hence contribute an additional ~ 10 dB of attenuation.

The signals retrieved from the sample are very small and can reach the single photon regime. In order to detect these signals in the presence of thermal noise and amplifier noise, usually larger than the signal itself, exceptional care has to be taken to minimise losses along the output line. The sample is protected from thermal noise of higher temperature stages and from the amplifier by two circulators from *Raditek inc.*, where the third port is terminated with a well thermalised $50\ \Omega$ load, see Figure 5.1 (c). This makes a single circulator act like a one-way microwave valve. Each circulator exhibits a typical isolation of ~ 20 dB and transmits the signal with low insertion loss < 1 dB. In order to reduce cable losses between the sample and the first amplifier, the coaxial cables are made from superconducting NbTi which exhibit little to no attenuation compared to stainless steel. A good amplifier is absolutely crucial in order to detect the ultra low signals. For this purpose a low noise amplifier (LNA) from *Low Noise Factory* (LNF) was used, see Figure 5.1 (c). This amplifier, additionally to its low noise temperature between $2 - 4$ K, offers a high dynamic range, large bandwidth from $4 - 12$ GHz and a gain of up to 40 dB.

5.2.3 DC Measurement Lines

Just as for the microwave measurement lines it is also important to reduce the sources of noise on the DC measurement lines, depicted in Figure 5.1 (c). The DC lines are made from copper loom where each loom contains 24 DC wires. The loom is

wrapped around copper posts at each temperature stage in order to assure thermal connection to the system and minimise thermal radiation to reach the mixing chamber, i.e. the sample. At the mixing chamber, the DC lines are additionally filtered and thermalised with an RC copper powder filter, in order to reduce the electron temperature T_e . While the phonon temperature reaches $T \leq 20$ mK in the cryogenic systems, the electron temperature was previously measured with Coulomb blockade thermometry² to only reach $T_e \sim 100$ mK.

5.3 DC Measurements

The DC measurements presented in Chapter 7 were carried out using battery powered low noise electronics from *QT Delft*. The electronics consist of high voltage sources (up to ± 90 V), 16 digital to analog converters which can be used as low noise voltage sources in a range of ± 2 V, current sources as well as current and voltage amplifiers. The electronics were controlled with a computer which was connected via an optical link in order to exclude any 50 Hz noise from the mains sockets. The DC signals (voltages or currents) could be set in a python software. The currents or voltages measured are quite small, hence one of the current or voltage amplifiers were used. The current amplifier translates the current into a voltage, which could later be decoded in the software to the measured current. The signals were acquired using an analog to digital converter on a digitiser board from *Alazartech* (ATS 9440).

The experiments in Chapter 8 only required constant DC voltages to be applied to the device and no detection of any transport current or voltage. For this purpose a *Keithley Sourcemeeter 2450* and a battery powered isolated voltage source from *Stanford Research Systems* (SIM 928 in mainframe SIM900) were used. In both cases exceptional care was taken in order to achieve proper grounding, such that ground loops are avoided and all of the equipment is referenced to the same ground, reducing noise and drift of the voltages.

²In Coulomb blockade thermometry the width of a Coulomb peak is measured and using the lever arm converted to an upper bound of the electron temperature.

5.4 Microwave Measurements

All the microwave measurements performed in this thesis are transmission type measurements, i.e. a microwave tone of certain frequency and power is injected at port #1 of the fridge and the device/resonator response is measured at port #2 of the fridge, see Figure 5.1 (c). Most commonly these type of measurements are performed using a vector network analyser (VNA), which can directly measure the scattering parameter S_{21} of a transmission measurement. All simple transmission measurements presented in this thesis were performed with a VNA from *Rohde & Schwartz* (ZVA 50). However, the capabilities of a standard VNA are stretched to their limit if spectroscopy measurements with two microwave tones or time-resolved measurements are performed. These types of measurements require a more involved microwave signal synthesis and signal acquisition setup, which will be described in the following sections.

5.4.1 Signal Synthesis

The microwave signal synthesis setup consists of three main components a rubidium frequency standard (clock) from *Stanford Research Systems*, three microwave signal generators from *Rohde & Schwartz* (Model: SGS 100A) and an arbitrary waveform generator (AWG) from *Tektronics* (Model: AWG5014C), see Figure 5.2. The clock is used to send a 10 MHz reference signal to the AWG, the microwave signal generator used as the local oscillator (LO) and the data acquisition card (ATS). The other two microwave signal generators (#1 and #2) use the reference of the LO for phase locking between all three of them. The signal generators can be used in continuous wave mode but also include an IQ mixer for upconversion which is necessary for time-resolved measurements. The IQ modulation necessary for the upconversion is supplied by the AWG. Any type of pulse scheme can be loaded onto the AWG, which then passes the pulses to the respective microwave signal generator. Additionally, the AWG sends a rising edge trigger to the data acquisition card in order to mark the start of the measurement. For two tone spectroscopy, i.e. qubit spectroscopy (see Section 8.3.1), the microwave tones from two signal

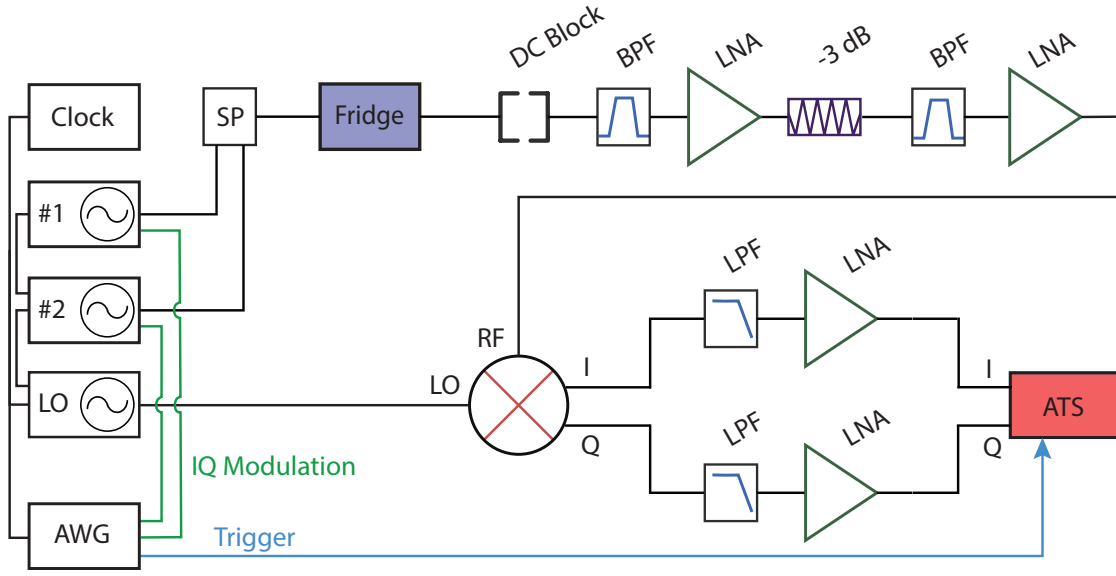


Figure 5.2: Schematic of the room-temperature microwave measurement setup. The setup consists of two parts, the signal generation and the signal acquisition. The signal generation consists of a clock three phase locked microwave generators and an AWG. The signals from generator #1 and #2 are combined in a splitter (SP) and fed into the fridge. The signal from the local oscillator (LO) generator is passed to the acquisition circuit. The signal leaving the fridge is passed through a DC block and a chain of amplifiers and filters before it is down-converted to an intermediate frequency and the two quadratures of the signal further amplified. The signal is digitised and recorded with an analog to digital converter card from *AlazarTech* (ATS).

generators (#1 - resonator drive tone, #2 - qubit drive tone) are combined in a splitter (SP) and fed into microwave port 1 of the fridge, see Figure 5.2.

5.4.2 Data Acquisition

The microwave signals from the sample have already been amplified at the 4 K stage of the fridge. Nevertheless, the signals are further filtered and amplified at room temperature. The LNA in the fridge is protected from any DC voltage with a DC block on the output of the fridge, see Figure 5.2. The signal is then filtered with a bandpass filter from *Keenlion* in order to reduce the background noise level within the bandwidth of the amplification chain. Subsequently, the signal is amplified with a wideband low noise amplifier from *Minicircuits* and thereafter even further filtered and amplified. A 3 dB attenuator is placed in between the two amplifiers in order to prevent standing waves between the active components. At this point

the signal has a convenient amplitude for acquisition. In order to analyse GHz signals with a computer the signal has to be downconverted. For this a standard heterodyne detection scheme is used. The fully amplified and filtered signal (RF) is mixed with the phase coherent and phase locked signal of the LO using an IQ mixer from *Marki Microwave*. The LO signal is offset by $\nu_{\text{IF}} = 125$ MHz from the measurement frequency, allowing convenient sampling of the I and Q quadratures at the intermediate frequency (IF) ν_{IF} . At this stage each quadrature is filtered with a lowpass filter from *Minicircuits* with a cut off frequency just above ν_{IF} , which greatly enhances the signal to noise ratio of the measurement. Subsequently the quadratures are again amplified, such that the signal has an amplitude in the millivolts range. Finally, both of the quadratures are acquired with an analog to digital converter on a data acquisition card from *AlazarTech* (ATS 9360) at a sampling rate of 1.8 GS/s. From the I and Q quadratures, the amplitude A and phase ϕ of the signal can be extracted via $Ae^{i\phi} = I + iQ$. Both the extracted amplitude and phase give valuable information about the devices measured using microwave spectroscopy.

5.5 RF Reflectometry Setup

Radio frequency (RF) reflectometry measurements were carried out in Section 7.4.3, which require a specific measurement setup inside the dilution refrigerator as well as at room temperature, see Figure 5.3. The input signals in these RF measurements were generated with a vector microwave source from *Agilent* (8267D) and fed into port 1. The input line from port 1 is attenuated by 32 dB and coupled towards the sample board using a directional coupler. The signal reflected from the sample board is routed through the directional coupler into the output line where it is amplified at the 4 K stage with a low noise high electron mobility transistor from the *Weinreb* group at Caltech and measured at port 2. The output signal at port 2 is measured using a homodyne downconversion circuit, converting the RF signal into a DC Voltage. For the downconversion the signal from the microwave source is split such that half of the signal is injected into the reflectometry circuit and the other half into the local oscillator port of the mixer. Further, the amplified reflected

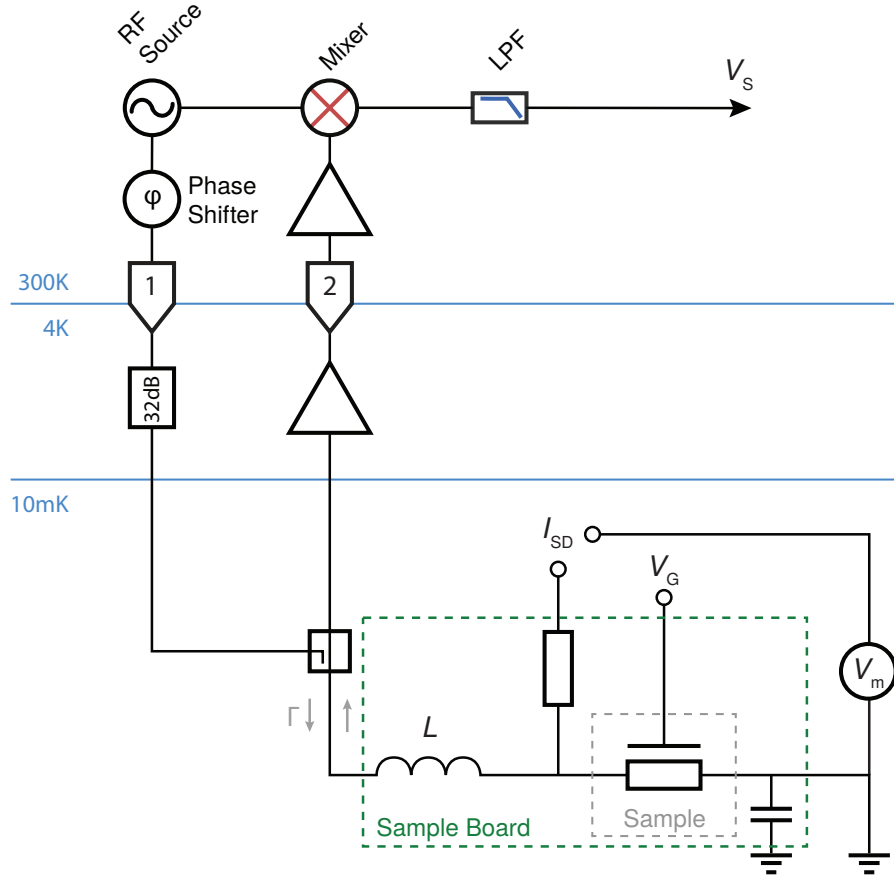


Figure 5.3: Radio frequency measurement circuit in the dilution refrigerator, including a simplified schematic of the sample board and the demodulation circuit at room temperature. The attenuated RF input line on port 1 is coupled to the sample board with a directional coupler. The signal reflected from the sample is amplified and routed to the demodulation circuit on port 2. The demodulation circuit consists of a phase shifter, a mixer and a low pass filter and is used to downconvert the reflected signal to a DC voltage V_s which is recorded with an analogue to digital converter. The sample board consists of an inductor L with the sample in series. The sample can be tuned with gate voltage V_G and is biased with a current I_{SD} . The voltage across the sample V_m can be measured in parallel to the RF signal, since the sample is effectively grounded for RF signals through a capacitor.

signal is fed into the RF port of the mixer. Since the mixer produces a signal at twice the RF frequency as well as at DC, the signal from the mixer is filtered with a low pass filter such that only the DC signal V_s remains for the homodyne detection. The DC signal is then acquired using an analog to digital converter on a digitiser board from *Alazartech* (ATS 9440). Since the DC signal depends on the amplitude and the phase shift of the reflected RF signal, both quadratures of the RF signal can be measured if the phase of the input signal is adjusted with

the phase shifter included in the downconversion circuit.

In Figure 5.3 a simple schematic of the sample board, including a sample exhibiting transistor type behaviour, is presented. This simple sample board circuit includes an inductor L and the sample, which usually has variable conductive, capacitive and inductive properties depending on the applied gate voltage. In the reflectometry measurements the reflected signal from the sample board is measured, which is given by the complex reflection coefficient Γ multiplied with the signal injected into port 1. The system comprising of the sample board and the sample is a resonant system with an impedance Z , which is a function of frequency and the sample properties, such that

$$\Gamma = \frac{Z - Z_0}{Z + Z_0}, \quad (5.1)$$

where $Z_0 = 50 \, \Omega$ is the impedance of the coaxial cables carrying the RF signals inside and outside of the dilution refrigerator. If the RF circuit is connected to one of the leads of the sample, shifts in both phase and amplitude of the reflected signal correspond to changes in capacitance, inductance or conductance of the sample. The measured RF signal can be related to the sample behaviour by comparison with simultaneous DC measurements.

Further details on the specific measurement setup and measurement technique can be found in the DPhil thesis of *F. Schupp* [173].

Part III

Main Results

6

Strong Coupling of Microwave Photons to Antiferromagnetic Fluctuations in an Organic Magnet

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Spin ensembles embedded in superconducting microwave circuits were one of the early implementations of hybrid quantum circuits. The ensembles are coupled to microwave resonators via their magnetic dipole moment and making use of the $\sqrt{N}$ enhancement discovered in the Tavis-Cummings Hamiltonian, see Section 2.3.1. This hybrid circuit QED architecture [26, 30, 32, 94, 174–176] has potential use in quantum memories [34, 35] as well as for microwave-to-optical conversion [177]. The first demonstrations used paramagnetic ensembles, but correlated states such as ferrimagnets lead to stronger coupling because of their high spin density as opposed to paramagnetic ensembles [31, 178]. However, this comes at the price

of on-chip magnetic fields, to which superconducting qubits (used as processors), microwave resonators (used for read out and coupling mediation) and SQUID arrays (used for resonator tuning) are sensitive. Antiferromagnetic spin ensembles avoid this obstacle by combining high spin densities with no net magnetisation. Perpendicular spin axis alignments of antiferromagnetic domains can also be used as a classical memory, which is robust against high magnetic fields, invisible to magnetic sensors, and can be packed with high density. Antiferromagnetic memory devices can be manipulated and read out via electrical currents [179, 180]. Furthermore, antiferromagnetic heterostructures would combine spintronic and magnonic functionalities [181]. Harnessing these possibilities makes it necessary to understand the range of interactions that occur in antiferromagnetic systems. As model systems, organic magnets can be engineered chemically to create well-defined magnetic interactions [182, 183], which could be probed via circuit QED to test models of magnetism in different dimensions [184]. Characteristic interaction strengths in organic magnets are such that these materials typically approach or undergo a phase transition only at mK temperatures [185–188], making them difficult to study with conventional electron spin resonance (ESR). Strong coupling to antiferromagnetic correlations would allow these materials to be studied at low temperatures, low microwave frequencies, and low magnetic fields. In this Chapter a first implementation of strong coupling between microwave photons and antiferromagnetic correlations in an organic magnet is presented. The need of an external magnetic field imposes restrictions on the superconducting material to be used for the coplanar waveguide resonators, see Section 4.2.2.1 for a characterisation of coplanar waveguide resonators in magnetic fields. The experiments presented in this chapter have been published in Reference [59] and the content has been adapted from there.

6.1 Hybrid Superconducting Circuit with a Spin Ensemble

In this chapter, strong coupling between microwave modes of a superconducting resonator and a crystallised organic radical, di(phenyl)-(2,4,6-trinitrophenyl)-iminoazanium (DPPH) is demonstrated. Spin resonance measurements of this material reveal antiferromagnetic correlations at a temperature $T \sim 4$ K and below, although no magnetic ordering is observed down to a temperature of 16 mK for the sample presented here (see Appendix B for susceptibility and muon spectroscopy measurements of the sample carried out by collaborators). Measurements showing coupling both to spin excitations (in the paramagnetic phase at high temperature $T \gtrsim 4$ K), and to excitations displaying antiferromagnetic correlations at lower temperature are presented [189, 190]. The angle dependence of the magnetic resonance is studied in order to investigate the anisotropy of the exchange interactions, evident from a separation in magnetic field of parallel and perpendicular resonances as the crystal is cooled. Further, the ensemble coupling as a function of temperature is analysed, which shows paramagnetic behaviour above $T \sim 500$ mK but becomes temperature-independent below $T \sim 50$ mK. The spin modes deviate from paramagnetic behaviour due to antiferromagnetic (AFM) fluctuations being present, despite being above the AFM phase transition temperature.

6.2 Device and Measurement Setup

The superconducting resonator was fabricated from a 110 nm NbTiN film, which was sputtered onto a quartz substrate, and patterned using optical lithography and reactive ion etching. The measured resonator, see Figure 6.1, has a centre conductor width of $w = 50 \mu\text{m}$ and a separation of $s = 5.3 \mu\text{m}$ from the lateral ground planes for 50Ω impedance matching on the quartz substrate, which exhibits a lower dielectric constant ϵ compared to silicon. DPPH molecules are stable radicals with a single unpaired electron on the nitrogen atom in the centre of the molecule, see Appendix B. Due to the spin of the free electron, DPPH powder or crystals

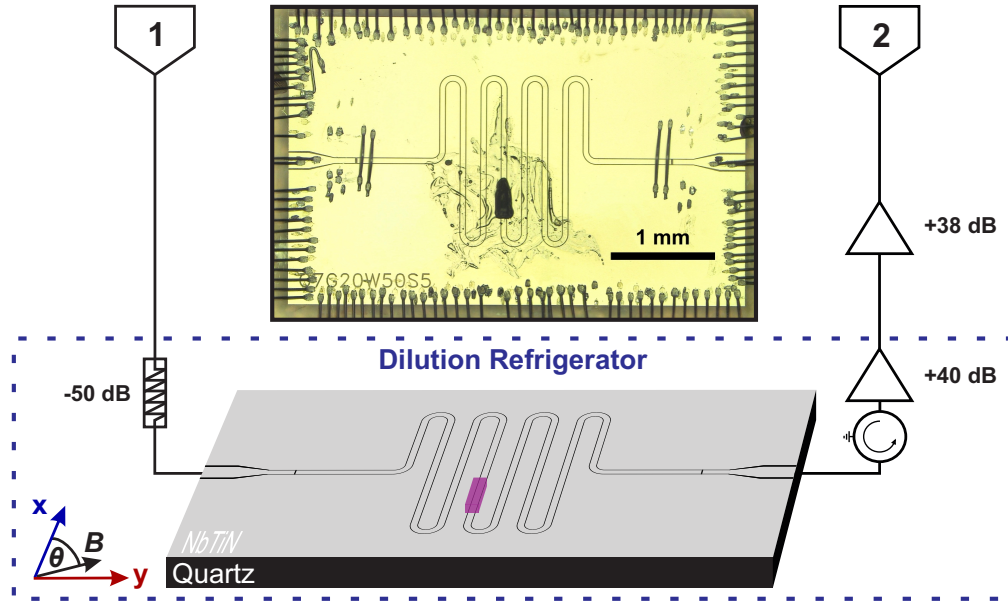


Figure 6.1: Experimental schematic. The coplanar resonator (inset photograph) is mounted in a dilution refrigerator and measured via two-port microwave transmission. A DPPH crystal (purple in schematic, black in photograph) is attached with vacuum grease near the magnetic field antinode of the resonator’s fundamental mode. Axes of the in-plane static magnetic field are indicated.

exhibit paramagnetic properties. However, intra molecular effects can also lead to magnetic ordering [191]. The DPPH crystals used for the following experiments were kindly synthesised by Jennifer Le Roy from the Materials Department here at the University of Oxford. The single crystals were grown via a saturated solution of DPPH in toluene, sitting in a hexane bath at 5 °C over two weeks. Using this method DPPH crystallises in a triclinic P-1 space group with a unit cell consisting of four DPPH, one hexane, and one toluene molecule, see Appendix B for details of the crystal growth and the crystal structure. From two identically prepared growth batches the largest crystals were measured. Measurements from both crystals (Crystal I and Crystal II) exhibit similar behaviour and are presented here. Each measured crystal was attached near the magnetic field anti-node of the resonator’s fundamental mode, with the long axis aligned along the CPWR, defining the x axis of the measurements, see Figure 6.1. The measurements were performed in a dilution refrigerator with base temperature $T \approx 15$ mK, fitted with a vector magnet such that an in-plane magnetic field $\mathbf{B} \equiv (B_x, B_y, 0)$ could be applied.

The hybrid device was measured via microwave transmission spectroscopy using a network analyser. At base temperature and in zero magnetic field, the resonator (with a crystal attached) exhibits a fundamental mode at frequency $\omega_0/2\pi = f_0 = 5.92$ GHz and a loaded quality factor of $Q_L = 1.51 \times 10^4$. An external magnetic field of magnitude $B \equiv |\mathbf{B}| = 165$ mT applied along x (along y) reduces this to $Q_L = 1.17 \times 10^4$ ($Q_L = 1.04 \times 10^4$), cf. Section 4.2.2.1.

6.3 Measurements

The following sections describe the experiments investigating the coupling between microwave photons and the spin ensemble in a correlated and uncorrelated state. Both the magnetic resonance as well as the coupling strength are studied as a function of temperature and magnetic field direction.

6.3.1 Electron Spin Resonance

The coupling to the crystal is investigated by measuring the resonator transmission $|S_{21}|^2$ at two different temperatures as a function of frequency f and magnetic field, see Figure 6.2. The bare resonator frequency is evident as a transmission peak that is nearly independent of magnetic field. By sweeping the external magnetic field, the spin resonance frequency f_{SR} is tuned through degeneracy with the resonator frequency $\omega_r/2\pi = f_r$, giving rise to an anticrossing when $f_{\text{SR}} \approx f_r$.

Because of the large number N of molecular spins, it is appropriate to parameterise the coupling to the resonator by an effective ensemble coupling g_{eff} [29, 30, 94], which is proportional to $\sqrt{N}$. In order to extract g_{eff} from the data, the system is modelled as two coupled oscillators, yielding the hybridised resonance frequency [192]

$$\omega_{\pm} = \omega_r + \frac{\Delta}{2} \pm \frac{1}{2}\sqrt{\Delta^2 + 4g_{\text{eff}}^2}, \quad (6.1)$$

where $\omega_{\pm}/2\pi = f_{\pm}$, $\Delta = g\mu_B (B_{x,y} - B_{\text{MR}})/\hbar$ is the frequency detuning and B_{MR} is the field at which magnetic resonance (MR) occurs. The transmission peak locations in Figure 6.2 are fitted to Equation 6.1 assuming a fixed Landé factor $g_e = 2.0037$ [193], resulting in the fit parameters g_{eff} and B_{MR} for Crystal I,

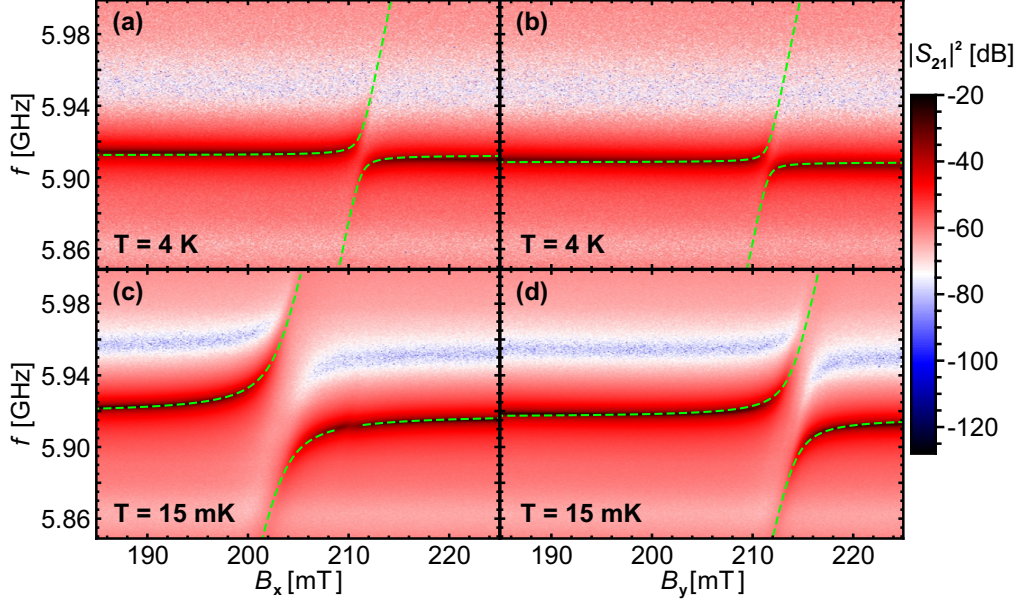


Figure 6.2: Transmission as a function of external magnetic field $B_{x,y}$ and resonator probe frequency f , measured at two different temperatures for Crystal I. Transmission maxima occur at resonance frequencies of the combined system, with anticrossings indicating hybridisation between crystal magnetic resonances and the cavity modes. Superimposed on each panel are fits to the resonance frequencies (dashed lines) using Equation 6.1.

shown in Table 6.1 for the two field directions and temperatures. The same spectroscopy measurements were repeated with Crystal II from a different growth batch, which used a newly opened bottle of DPPH powder precursor, see Appendix B for the microwave transmission spectroscopy data. Again using Equation 6.1 to fit the resonance peak locations yields similar values compared to the ones extracted from Crystal I, see Table 6.1. However, the magnetic resonance field is slightly lower than for the device with Crystal I, consistent with the lower cavity frequency, which is attributed to the fact that Crystal II was slightly larger and presumably acted as a dielectric load.

The spin dephasing rate $\gamma(T)$ can be deduced by fitting a standard input-output model [29, 31, 93, 192, 194]

$$|S_{21}(\omega)|^2 = \left| \frac{\kappa_c}{i(\omega - \omega_r) - \kappa + \frac{g_{\text{eff}}^2}{i(\omega - \omega_{\text{MR}}) - \gamma}} \right|^2, \quad (6.2)$$

where κ_c is the coupling rate of the resonator to the external microwave circuit and $2\kappa/2\pi \equiv f_0/Q_L$ is the total relaxation rate of the resonator. Equation 6.2 was used

Crystal	T (K)	Axis	B_{MR} (mT)	$g_{\text{eff}}/2\pi$ (MHz)	$\gamma/2\pi$ (MHz)	C
I	4	x	211.19 ± 0.05	12.1 ± 0.4	15.0 ± 0.3	18
I	4	y	211.53 ± 0.05	9.6 ± 0.4	15.2 ± 0.6	10
I	0.015	x	203.12 ± 0.02	38.7 ± 0.1	29.6 ± 0.3	200
I	0.015	y	213.75 ± 0.05	26.9 ± 0.3	25.5 ± 0.4	102
II	4	x	208.36 ± 0.02	17.01 ± 0.1	11.5 ± 0.2	24
II	4	y	209.32 ± 0.02	12.75 ± 0.1	11.3 ± 0.2	14
II	0.015	x	200.60 ± 0.01	38.04 ± 0.1	29.6 ± 0.3	80
II	0.015	y	210.86 ± 0.06	29.59 ± 0.3	25.6 ± 0.4	34

Table 6.1: Resonance parameters of Crystal I and Crystal II extracted from microwave transmission spectroscopy measurements, depicted in Figure 6.2 for Crystal I (see Appendix B for Crystal II), for different temperatures and magnetic field orientations.

to fit $|S_{21}(\omega)|^2$ at the resonance fields B_{MR} , taking κ_c and γ as fit parameters and holding constant the parameters g_{eff} , ω_r and κ deduced above. Extracted values of γ for both crystals are shown in Table 6.1.

A dimensionless parameter giving a measure of the coupling efficiency is the cooperativity $C \equiv g_{\text{eff}}^2/\kappa\gamma$, cf. Section 2.1.1. This parameter is extracted for each temperature and field axis for both crystals, see Table 6.1. Already at $T = 4$ K, either system is in the regime of high cooperativity ($C > 1$), implying coherent transfer of excitations from the microwave field to the ensemble, while at $T = 15$ mK the strong coupling condition $g_{\text{eff}} \gg \kappa, \gamma$ is reached, where the ensemble coupling is faster than the decay of both the spin ensemble and the resonator. At low temperature the cooperativity for Crystal II is significantly lower compared to Crystal I even though it exhibits similar values for g_{eff} and γ . This is due to the larger dimensions of the crystal, which leads to more dielectric losses in the resonator reflected in a smaller quality factor and hence larger κ . At higher temperature the cooperativity for Crystal II is slightly higher than for Crystal I, this is due to the lower relaxation rate of the spin ensemble measured at this temperature. The resonator decay rate due to dielectric losses at this temperature has a smaller contribution as in both cases the resonance is broadened due to temperature and hence the resonator exhibits similar values for κ in both devices.

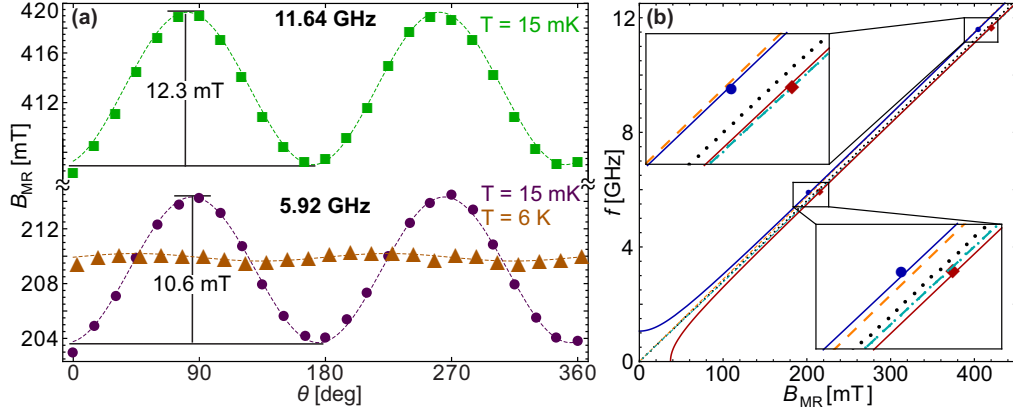


Figure 6.3: Angular dependence and antiferromagnetic resonance for Crystal I. (a) Resonance magnetic field as a function of field angle θ . Measuring at $T = 15$ mK, the resonance field varies sinusoidally with θ , with amplitude $\Delta B_0 = 10.6$ mT for the fundamental mode ($f_0 = 5.92$ GHz, circles) and $\Delta B_1 = 12.3$ mT for the first harmonic mode ($f_1 = 11.64$ GHz, squares). At high temperature, the fundamental mode shows nearly isotropic resonance (triangles). (b) Plot of the MR frequency as a function of resonance magnetic field. Data points are the resonance magnetic fields along x (circles) and along y (diamonds), taken from the maximum and minimum data points of panel (a) for data at the fundamental or first harmonic mode. The black dotted line is the PM dispersion relation with Landé factor $g_e = 2.0037$. The dashed orange and dot-dashed cyan lines are fits using a PM dispersion relation, with separate g -factors along the two axes taken as fit parameters. From the insets it is apparent that these fits do not describe the data well. Red and blue solid curves are a fit to the AFM dispersion relation in Equation 6.4, which agrees well with the data.

6.3.2 Angular Dependence and Antiferromagnetic Resonance

In this section it is shown that the two measured crystals exhibits antiferromagnetic correlations at low temperature. Whereas at high temperature, compare Figure 6.2 (a)-(b) for Crystal I, the magnetic resonance field B_{MR} is nearly independent of external field direction, at $T = 15$ mK there is a pronounced anisotropy, see Figure 6.2 (c)-(d) for Crystal I (cf. Figure B.1 for Crystal II). This behaviour is explored further for Crystal I in Figure 6.3 (a), which compares the dependence of B_{MR} on field angle θ at $T = 6$ K and $T = 15$ mK. Measuring near the fundamental mode of the resonator f_0 , the angle dependence is well fit by

$$B_{MR} = B_{MR}^{\text{offset}} + \Delta B_i \sin^2(\theta + \Delta\theta) \quad (6.3)$$

with offsets B_{MR}^{offset} and $\Delta\theta$ together with anisotropy ΔB_i as fit parameters, where $i \in \{0, 1\}$ labels the cavity mode. At low temperature, we find $\Delta B_0 = 10.6$ mT,

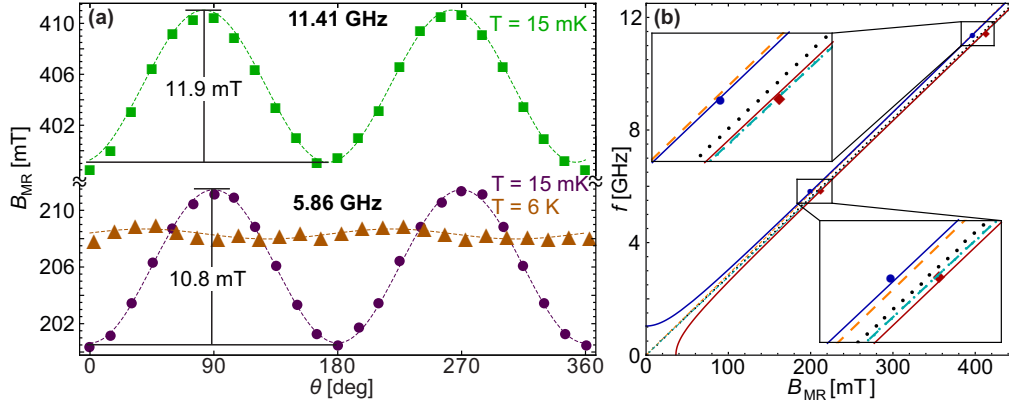


Figure 6.4: Angular dependence and antiferromagnetic resonance for Crystal II. (a) Resonance magnetic field B_{MR} as function of field angle θ at different cavity modes and temperatures with fits to Equation 6.3. (b) Plot of the MR frequency as a function of resonance magnetic field. Data points are the resonance magnetic fields along x (circles) and along y (diamonds), taken from the maximum and minimum data points of panel (a) for data at the fundamental or first harmonic mode. The black dotted line is the PM dispersion relation with Landé factor $g_e = 2.0037$. The dashed orange and dot-dashed cyan lines are fits using a PM dispersion relation, with separate g -factors along the two axes taken as fit parameters. From the insets it is apparent that these fits do not describe the data well. Red and blue solid curves are a fit to the AFM dispersion relation in Equation 6.4, which agrees well with the data.

whereas at 6 K there is almost no angle dependence. The same measurement was repeated for Crystal II, see Figure 6.4 (a), which exhibits similar behaviour. At low temperature the angular dependence of B_{MR} displays an amplitude of $\Delta B_0 = 10.8$ mT and at 6 K is also independent of the magnetic field angle.

At high temperature, i.e. $T = 6$ K, this is consistent with a paramagnet with nearly isotropic g -factor. The anisotropy at low temperature could arise from field screening by the superconductor, from temperature-dependent g -factor anisotropy, from trapped flux in the magnet coils, or from a transition to magnetic correlations in the crystals. Field screening from the superconducting thin film is excluded by measurements with different crystal orientations of Crystal II, see Figure 6.5. To confirm that the anisotropy is associated with the crystal rather than the resonator the low- T fundamental-mode measurement of Figure 6.4 (a) was repeated after rotating Crystal II by 90° in the xy plane. Compared to Figure 6.4 (a) the data shows the expected shift by 90° , see Figure 6.5 (a), confirming that the anisotropy arises from the crystal structure rather than e.g. screening by the resonator.

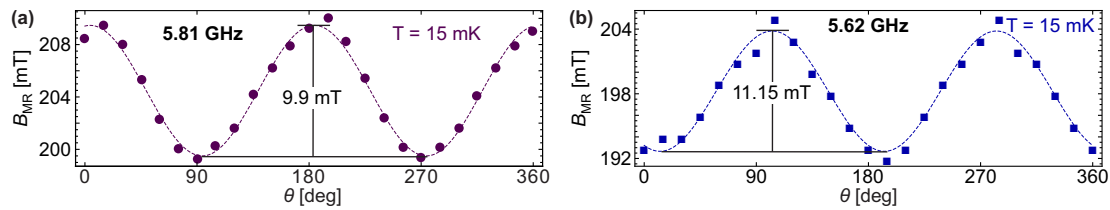


Figure 6.5: Angular dependence for rotated Crystal II. (a) Resonance magnetic field B_{MR} as function of field angle θ , after the crystal has been rotated by 90° in the xy plane. Data is measured near the fundamental mode and at $T = 15$ mK. Compared to Figure 6.4 (a) the trace is shifted by 90° , as expected due to the crystal rotation. (b) Equivalent data after rotation by $\sim 90^\circ$ in the yz plane from configuration of Figure 6.4.

Additionally, the crystal was measured in a configuration rotated by approximately 90° in yz , in order to attempt to map the anisotropy axis in three dimensions. The angle dependence in this configuration is shown in Figure 6.5 (b). Although the anticrossing was harder to identify for some field angles in this configuration, the estimated resonance positions show the same sinusoidal dependence as for Figure 6.4 (a) and indicate uniaxial behaviour with anisotropy axis along x in Figure 6.1. To further exclude g -factor anisotropy, the angular measurement was repeated at the first harmonic of the resonator $f_1 = 11.64$ GHz, upper trace in Figure 6.3 (b) for Crystal I [$f_1 = 11.41$ GHz, upper trace in Figure 6.4 (b) for Crystal II]. Whereas g -factor anisotropy would lead to $\Delta B_1 = 2\Delta B_0$, in fact it is found that $\Delta B_1 = 12.3$ mT $\approx \Delta B_0$ (Crystal I) and $\Delta B_1 = 11.9$ mT $\approx \Delta B_0$ (Crystal II). Trapped flux in the coils is also excluded by the temperature dependence, since the coils are thermally isolated from the sample. Therefore an onset of AFM correlations between 15 mK and 4 K is deduced for both crystals.

In order to confirm antiferromagnetic behaviour, the magnetic resonance dispersion relation for the two principal axes is plotted in Figure 6.3 (b) for Crystal I and in Figure 6.4 (b) for Crystal II. Although in both cases each branch contains only two data points, they clearly do not satisfy a paramagnetic (PM) dispersion relation $f = g\mu_B B_{MR}/h$ (dotted line in the figures), even allowing for g -tensor anisotropy (dashed/dot-dashed lines in the figures). However, for Crystal I as well as Crystal II the data-points are well fit by an AFM dispersion relation [195] derived from a

two-sublattice model with a molecular-field approximation at zero temperature [196]:

$$f = \frac{g\mu_B}{h} \sqrt{B_{\text{MR}}^2 \pm K}, \quad (6.4)$$

where the $+$ ($-$) branches describes field alignment parallel (perpendicular) to the anisotropy axis. Here the fit parameters are K , which parameterises the exchange and anisotropy field of the crystal and separate g -factors g_x and g_y for the two field directions [195, 197]. For Crystal I, fitting all four data points simultaneously, the best fit parameters are $K = 0.0014 \text{ mT}^2$, $g_x = 2.04$, and $g_y = 1.99$, similar to a previously reported value $g = 2.0037$ in the PM phase [193]. The same fit to the data from Crystal II yields best fit parameters $K = 0.0013 \text{ mT}^2$, $g_x = 2.04$, and $g_y = 2.00$ very similar to the ones from Crystal I. Hence at low temperature, the magnetic resonance excitations are no longer single spin flips, but antiferromagnetic fluctuations.

6.3.3 Temperature Dependence

The temperature evolution of the effective spin polarisation can be studied via the effective ensemble coupling strength g_{eff} , see Figure 6.6 (a) for Crystal I. Above 50 mK, g_{eff} decreases with increasing temperature, as expected from thermal depolarisation of the spin ensemble. For a paramagnet, the effective coupling is [94]

$$g_{\text{eff}}(T) = g_s \sqrt{N_{\text{P}}(T)} = g_s \sqrt{N \tanh(hf/2k_{\text{B}}T)}, \quad (6.5)$$

where g_s is the root-mean-square coupling per individual spin and $N_{\text{P}}(T)$ is the net number of polarised spins out of N coupled radicals. Above $T = 0.5 \text{ K}$, shaded region of Figure 6.6 (a), Equation 6.5 gives a good fit to the data of Crystal I. Calculating $g_s/2\pi = 5 \text{ Hz}$ from the geometry of the resonator (following Section 2.4) and taking the number of coupled radicals as a fit parameter gives $N_x = 1.5 \times 10^{14}$ for $\mathbf{B}$ along x of Crystal I. This is in fair agreement with $N = 1.7 \times 10^{14}$ estimated from the geometry of Crystal I, see Appendix B. The data for $\mathbf{B}$ along y gives a smaller value $N_y = 7.1 \times 10^{13}$, as it is expected due to the smaller perpendicular overlap with the alternating resonator field. For Crystal I high cooperativity ($C > 1$)

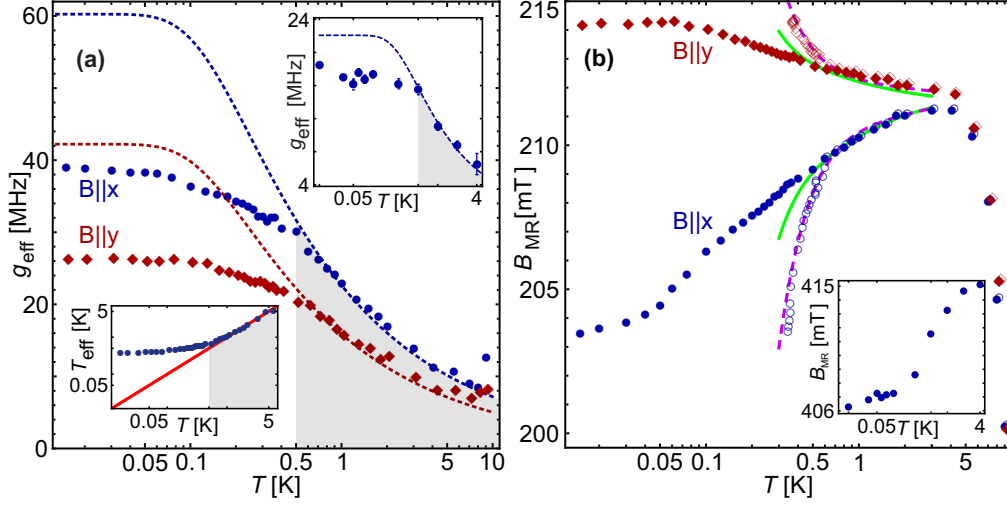


Figure 6.6: Temperature dependence of Crystal I. (a) Temperature evolution of g_{eff} for $\mathbf{B}$ applied along x and y . Above $T \sim 500$ mK, the data agree with a PM model [dashed lines, fit to Equation 6.5 over the shaded temperature range]. Inset right: Similar data and fits at the resonator’s first harmonic. Inset left: Effective spin-temperature calculated with Equation 6.5 (points). (b) Filled symbols: Resonance magnetic field along x and y as a function of temperature. As temperature decreases, the resonance magnetic field moves away from its paramagnetic value (assuming $g_e = 2.0037$). At intermediate temperatures, both branches are fit by a spin chain model (solid curves; see text). Unfilled symbols: B_{MR} along x and y as a function of effective temperature according to Equation 6.5. The data for $T \leq 3$ K is fit by a spin chain model (dashed curve; see text). Inset: Similar data at the first harmonic mode.

is already reached far above base temperature, for example at $T = 0.5$ K, where $C_x = 66$ and $C_y = 28$. The agreement with the two-level model given in Equation 6.5 confirms that the magnetic resonance spectroscopy probes a transition from the spin ground state (rather than between two excited states).

Below 0.5 K, g_{eff} is found to be smaller than the fits would predict. This may reflect screening of each spin by its neighbours as the antiferromagnetic phase is approached, although the $\sqrt{N_{\text{P}}}$ enhancement of g_{eff} is still expected to apply [178]. It may also reflect a failure of the spin ensemble to thermalise. By comparing the measured $g_{\text{eff}}(T)$ with the value predicted by Equation 6.5, an effective spin temperature T_{eff} for Crystal I can be extracted, see Figure 6.6 (a) inset left. At the lowest temperature, the effective number of coupled spins is $N_{\text{P}} = (g_{\text{eff}}/g_s)^2 \approx 5.9 \times 10^{13}$ for $\mathbf{B}$ along x . Similar behaviour is observed at the resonator’s first harmonic mode, see Figure 6.6 (a) inset right, with smaller overall

coupling because the crystal is not located at a field antinode.

For Crystal II the temperature dependence of g_{eff} exhibits similar behaviour, see Appendix B. Equivalent fits to the data of Crystal II yield $N_x = 1.85 \times 10^{14}$ for $\mathbf{B}$ along x and $N_y = 1.2 \times 10^{14}$ for $\mathbf{B}$ along y , which is as expected from the slightly larger dimensions of Crystal II compared to Crystal I. The measured g_{eff} is again found to be smaller than suggested by the model in Equation 6.5. As before this is attributed to the spin ensemble failing to thermalise and similar as for Crystal I an effective spin temperature T_{eff} is extracted, see Appendix B. The effective number of coupled spins at the lowest temperature is found to be $N_{\text{P}} \approx 5.7 \times 10^{13}$ for $\mathbf{B}$ along x for Crystal II. The slightly lower number of effectively coupled spins is most likely reflected in the crystal being offset from the centre of resonator or potentially not being mounted as close to the resonator as Crystal I.

Studying the temperature dependence of the magnetic resonance can give experimental insight into the spin correlations, where analytical solutions for models of interacting spins in three dimensions do not exist. The shift of the magnetic resonance frequency away from the high-temperature (paramagnetic) value is a measure of short-range correlations. The magnetic resonance field of Crystal I as a function of cryostat temperature for parallel and perpendicular field alignment is presented in Figure 6.6 (b) by the filled symbols (for similar data of Crystal II see Appendix B). Both datasets exhibit a kink at $T \sim 50$ mK which could suggest a phase transition, and indeed such a transition to an AFM state at $T \sim 0.3$ K has been previously observed in DPPH [189, 190]. However, for the DPPH crystals presented here, separate investigations using ac susceptibility and muon spectroscopy show that there is no phase transition down to $T = 16$ mK, see Appendix B. The transition temperature in DPPH is known to vary widely depending on the crystallising solvent [198], and the incorporated toluene and hexane in the crystals used here presumably inhibit ordering at accessible temperatures, cf. Appendix B. Therefore, the low-temperature kink in Figure 6.6 (b) (filled symbols) (and similarly for Crystal II in Appendix B) is attributed to the failure of the spins to thermalise inside the resonator, which is a fair assumption due to mounting the crystal with

vacuum grease. This interpretation is supported by plotting the same data for Crystal I as a function of the effective spin temperature T_{eff} [extracted as in Figure 6.6 (a) left inset], which shows that the kink disappears, see Figure 6.6 (b) unfilled symbols. The same effect is observed for Crystal II, see Appendix B. At high temperature ($T \gtrsim 5$ K), the resonances shift to lower field because of the (independently measured) decrease in resonator frequency due to kinetic inductance.

The temperature dependence of the resonance frequencies is simulated by calculating the short range spin-spin correlations between DPPH molecules. The spin Hamiltonian is:

$$\mathcal{H} = \mathcal{H}^0 + \mathcal{H}', \quad (6.6)$$

where $\mathcal{H}^0 = -2 \sum_{i,j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - g\mu_B \sum_i \mathbf{B} \cdot \mathbf{S}_i$ incorporates isotropic exchange and Zeeman energy, and $\mathcal{H}'$ represents the anisotropic exchange between molecules, e.g. dipole-dipole interactions. Here $\mathbf{S}_i = \{S_i^x, S_i^y, S_i^z\}$ is the spin of the i^{th} molecule, and $J_{ij} < 0$ is the isotropic exchange. Equation 6.6 assumes an isotropic g -tensor, which is not required by symmetry but is justified experimentally by the isotropy of the magnetic resonance field well above the phase transition, see Figure 6.3 (a). The bulk permeability of the material is neglected. In the absence of anisotropy, i.e. $\mathcal{H}' = 0$, Equation 6.6 leads to a temperature independent ESR resonance frequency with $f = g\mu_B B$, which is identical to the ESR resonance for noninteracting spins, despite the isotropic interaction [199]. Any shift of this resonance frequency indicates an effect of $\mathcal{H}'$. Assuming $\mathcal{H}^0 \gg \mathcal{H}'$, the frequency shift is [199–201]:

$$h\delta f = - \frac{\langle [[\mathcal{H}', S^+], S^-] \rangle}{2\langle S^z \rangle} \quad (6.7)$$

where $\langle \dots \rangle$ indicates the temperature-dependent expectation value, $\mathbf{S} \equiv \sum_i \mathbf{S}_i$ is the total spin operator, and $S^\pm \equiv S^x \pm iS^y$.

To gain insight into the role of antiferromagnetic fluctuations, a simple model of a one-dimensional uniaxial anisotropic antiferromagnet was used [200]. This is also suggested by the crystal packing, where solvent molecules may act as blocks between chains, see Appendix B. Therefore, $\mathcal{H}^0 = -2J \sum_i \mathbf{S}_i \cdot \mathbf{S}_{i+1} - g\mu_B \sum_i \mathbf{B} \cdot \mathbf{S}_i$

and $\mathcal{H}' = 2J^A \sum_i S_i^x S_{i+1}^x$. In a classical approximation, expected to be valid at high temperature, the frequency shift following Equation 6.7 can be evaluated exactly [200, 202]. With the exchange constants as free parameters, the shift along the x axis for the data of Crystal I is fitted in the range $0.5 \text{ K} \leq T \leq 3 \text{ K}$, giving $J/k_B \sim -300 \pm 200 \text{ mK}$ and $J^A/k_B \sim -9 \pm 4 \text{ mK}$, see Figure 6.6 (b) lower solid curve. The same parameters give a good match for the shift along the y axis, see Figure 6.6 (b) upper solid curve. Performing the same analysis on the data of Crystal II yields similar values $J/k_B \sim -235 \pm 180 \text{ mK}$ and $J^A/k_B \sim -7.5 \pm 3 \text{ mK}$, see Figure B.2 (b) in Appendix B.

As an alternative to fitting over this restricted temperature range, the data for Crystal I can also be fitted as a function of the effective spin temperature T_{eff} over the entire range $T_{\text{eff}} \leq 3 \text{ K}$, cf. lower dashed curve in Figure 6.6 (b). This yields similar values $J/k_B = -1200 \pm 500 \text{ mK}$ and $J^A/k_B = -10 \pm 3 \text{ mK}$. As before the same parameters give a good fit for the shift along y , see Figure 6.6 (b) upper dashed curve. The identical analysis is also done on the data extracted from the measurements of Crystal II, yielding similar values $J/k_B = -700 \pm 320 \text{ mK}$ and $J^A/k_B = -12 \pm 2 \text{ mK}$, see Figure B.2 (b) in Appendix B. Interestingly, in all cases the extracted anisotropic exchange is close to the dipole-dipole interaction $J^A/k_B = -3\mu_0 g^2 \mu_B^2 / 8\pi a^3 k_B \sim -10 \text{ mK}$ estimated from the molecular spacing $a \sim 7.1 \text{ \AA}$, found via X-ray diffraction. The deviation between fit and data presumably reflects the increasing importance of quantum correlations at low temperature and higher dimensionality of the interactions, neither of which is well captured by this one-dimensional model. Further, the imperfection of the measured or estimated spin-temperature, due to the bad thermalisation, is also reflected in the deviation of the fit to the data. The anisotropy axis in the spin resonance coincides with the long axis of the crystal, here labelled the x axis, but does not appear to correspond to any preferred direction in the X-ray diffraction structure, see Appendix B. The temperature dependence of the magnetic resonance field does not simply result from a demagnetising field, which would be weaker and would have the same sign for both orientations.

6.4 Conclusion

In this chapter the coupling between a microwave resonator and two individual crystals of a molecular ensemble both in an uncorrelated and AFM correlated state has been shown. The experiments described have already been published and the description followed Reference [59]. The anisotropy apparent in the magnetic resonance field was shown to arise due to AFM correlations in the crystals. Further, it was observed that the spins of the crystals stop thermalising before a phase-transition is reached. The resulting temperature dependence of the magnetic resonance can be described well by a simple one-dimensional uniaxial anisotropic antiferromagnetic model, yielding an anisotropic exchange energy comparable to the dipole-dipole interaction strength deduced from the crystal structure.

The crystal structure of Crystal I and Crystal II presumably exhibits a complex network of exchange interactions, but the circuit QED spin resonance techniques presented in this chapter, applied in future experiments, will enable measurements of spin systems with engineered interactions, for example molecular magnets in one-dimensional chains or higher-dimensional systems with well-defined exchange pathways [182, 183]. Magnetic resonance measurements on these molecules offer a way to extract spin correlation functions experimentally via Equation 6.7, thereby offering a platform to test theoretical predictions for quantum correlated systems. As a quantum memory, organic magnetic ensembles offer a high spin density, and therefore a strong ensemble coupling, with potential for chemical engineering of the spin system.

7

Carbon Nanotube Josephson Junctions Measured in DC and with RF Reflectometry

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Mesoscopic conductors/semiconductors can be probed in DC transport measurements, either in voltage- or current-bias. The tunnelling-barriers created by the electrical leads (source- and drain-contacts) to the conductor/semiconductor create a confinement potential for charge carriers. The electrostatic potential inside this potential well can be tuned with an applied gate voltage. The transport characteristics of such devices are explained and modelled by the theory of quantum

dots, see Chapter 3.

In this chapter experimental results of transport measurements of carbon nanotubes contacted by normal metals and superconductor-normal bilayers are presented. An emphasis is put especially on the devices contacted with superconductor-normal bilayers, since these effectively behave like Josephson junctions. The critical current of these devices is studied as a function of gate voltage in DC transport as well as radio frequency reflectometry measurements.

7.1 Device Fabrication

All measurements presented in this chapter utilise the same device geometry and measurement setup. Carbon nanotubes are initially grown via chemical vapour deposition on 10x10 mm Si/SiO₂ substrates. The substrate is then cleaved into smaller 5x5 mm chips and a first layer with 32 sets of 3 bond-pads and alignment markers is patterned via e-beam lithography and subsequently metalised with Ti/Au (10/60 nm). Afterwards, SEM imaging is used for locating and picking suitable carbon nanotubes. Each carbon nanotube is contacted with source- and drain-contacts, separated by $L = 300$ nm, and a side-gate is patterned next to it using e-beam lithography, see Figure 7.1 (c). Prior to evaporating the contacting metal, a cleaning step using a UV ozone etcher is performed in order to get rid of any PMMA residue on the carbon nanotube. Depending on the device either Pd (50 nm, normal) or a Pd/Al bilayer (4/80 nm, superconducting) is used as the contacting material.

The chip is mounted with PMMA in a sample board which allows radio frequency and DC electronic access to the devices, see Figure 7.1 (a). Previously selected devices (see Section 7.2) are then wire-bonded with Al wire to the sample board, see Figure 7.1 (b).

The superconductor-normal bilayer made from Pd/Al used here was developed and refined in previous work by *A. Nersisyan* [137] achieving highly transparent superconducting contacts to carbon nanotubes. In this initial work critical currents between $0.1 \text{ nA} < I_c < 8 \text{ nA}$ were observed. For the devices presented here the fabrication recipe was tweaked in order to improve the contacts, by changing the

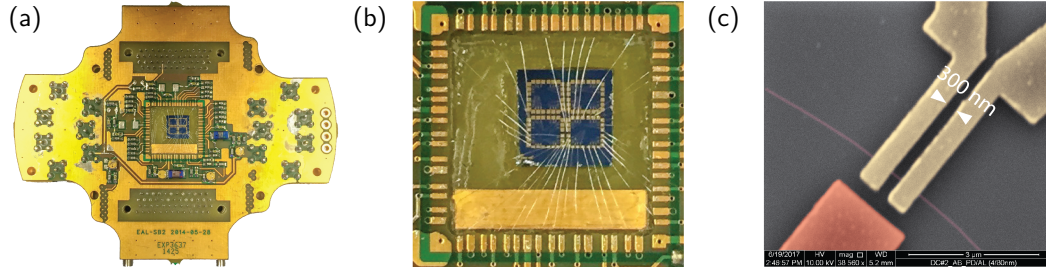


Figure 7.1: Images of PCB and device chip. (a) Image of the PCB with the bonded device chip. The PCB allows for DC and RF connectivity. (b) Image of the bonded device chip. Up to 24 devices are fabricated on a 5x5 mm Si/SiO₂ chip. Each contact (source, drain and gate) is wire-bonded with Al wire (thin silver lines) to the PCB. (c) False color SEM image of a single carbon nanotube (purple) with source- and drain-contacts (gold) and a side-gate (red). The channel length is $L = 300$ nm.

duration of the UV ozone cleaning step prior to contact metal deposition. This was observed to increase the device yield as well as decrease room temperature resistances of contacted carbon nanotubes, indicating that higher critical current carbon nanotube Josephson junctions should be achievable. As shown later in this chapter, several devices with critical currents in the 15 – 18 nA range were observed. It is worth pointing out that there are many variables that can affect the critical currents, and measurements of a much larger number of devices with systematic variation of fabrication parameters would likely be fruitful in refining the fabrication process. The carbon nanotubes used here were grown in a different furnace with a different recipe as opposed to the devices presented in Reference [137], very likely resulting in different quality nanotubes. Additionally, the metal deposition was performed in a home-built electron beam evaporator which was only recently fitted with a cryogenic pump, presumably improving the metal film quality due to the lower base and evaporation pressure. However, the crystal monitoring the evaporation rate was not calibrated on a regular basis, which will have an effect on the metal film thicknesses of the bilayer and hence directly impacting the induced superconducting gap Δ as well as the transparency of the contacts T_i .

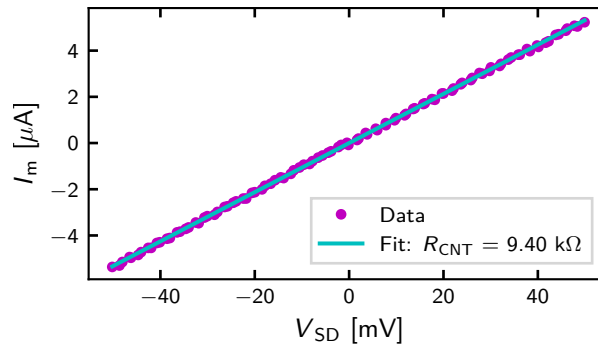


Figure 7.2: Room-temperature I-V curve of a carbon nanotube DC device with Pd/Al contacts. The I-V trace shows ohmic behaviour and is fit with a linear ohmic model (cyan curve), resulting in a resistance of 9.40 k Ω .

7.2 Room-Temperature Measurements/Selection

Prior to measuring carbon nanotube DC samples at cryogenic temperatures, each fabricated device is probed at room temperature on a DC probe station. For each device single I-V curves are recorded between their respective source- and drain-contacts with no voltage applied to the gate, see Figure 7.2. The I-V traces are used to pre-select the carbon-nanotube devices which are then wire-bonded to the sample board and measured at low temperature.

Room-temperature I-V traces of carbon nanotube devices can give insight into the fabrication yield of contacting the carbon nanotubes and the respective contact resistance. Furthermore, the I-V traces show either ohmic or semiconducting behaviour. Mainly devices with ohmic I-V traces (metallic/semi-metallic carbon nanotubes) are selected, as these devices usually show a fairly small semiconducting gap at low temperature.

Of about 150 fabricated carbon nanotube devices the contacting yield was $> 50\%$ and room-temperature resistances down to 7.1 k Ω have been observed, which indicates that some of these devices might be multi-wall rather than single-wall carbon nanotubes. About 60% of all contacted carbon nanotubes show resistances ≤ 25 k Ω and all of these conduct at low temperature. This is good quantitative evidence that the fabrication of the carbon nanotube devices, i.e. the contacting, is reproducible and reliable.

7.3 Carbon Nanotubes with Normal Leads

In this section DC transport measurements of carbon nanotubes contacted with normal metal (50 nm Pd) are presented. The motivation behind these measurements is to test the device fabrication, contact resistance and quality of the carbon nanotubes. As mentioned previously, see Chapter 3, the good wettability of Pd with respect to carbon nanotubes and a similar work function lead to low contact resistances in these devices.

In the case that the carbon nanotube is only weakly coupled to the leads, single electron transport can be measured through the quantum dot formed within the nanotube. The measurements presented in Figure 7.3 of a device described in Section 7.1, show clear signs of Coulomb blockade. The current I_m measured through the device as a function of the gate voltage V_G at a finite source-drain bias $V_{SD} = 5$ mV, shows peaks of high current and valleys of zero current, i.e. Coulomb blockade, see Figure 7.3 (a). For large positive gate voltages, $V_G > 30$ V, the current decreases completely to zero, which is an indication for p-type semiconductivity.

When V_{SD} is also tuned, in addition to V_G , and the current/conductance is plotted as a function of both, the peaks evolve in a triangular pattern and form the familiar Coulomb diamonds, see Figure 7.3 (b). The measurement shows the differential conductance map, i.e. stability diagram of the carbon nanotube quantum dot at $T = 10$ mK. The conductance of the device varies between 0 and $0.05G_0$, where $G_0 = 2e^2/h$ is the conductance quantum. Since the edges of the diamond that correspond to the different leads exhibit similar conductance, we can infer an almost symmetric coupling of the quantum dot to the two leads. In the area of conductance, resonances parallel to the diamond edges can be observed, which are due to charge transitions through excited states, indicated by purple arrows in Figure 7.3 (b). The conductance lines inside the Coulomb diamonds correspond to charge transport due to co-tunnelling effects, indicated by cyan arrow in Figure 7.3 (b). Usually the co-tunnelling should be independent of gate voltage. However, a gate-dependence has been observed in other experiments and attributed to the difference in coupling to the leads of the two doublets or excited states formed in carbon nanotubes [203–205].

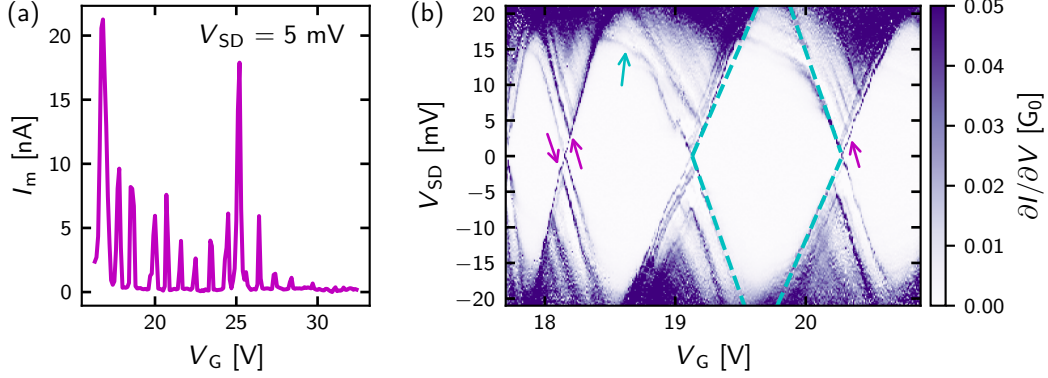


Figure 7.3: DC transport measurements of a single quantum dot in a carbon nanotube with Pd leads at $T = 10 \text{ mK}$. (a) Current I_m through the carbon nanotube as a function of applied gate voltage V_G at $V_{SD} = 5 \text{ mV}$. A sequence of Coulomb peaks is observed and in between them the current goes to zero. For $V_G > 30 \text{ V}$, the device is pinched off. (b) Stability diagram of a carbon nanotube device obtained by tuning V_G and V_{SD} . Coulomb diamonds are observed and one is traced by the cyan dashed line. Resonances parallel to the diamond edges are excited state transitions, indicated by purple arrows. The conductance inside the diamond is due to co-tunnelling effects, indicated by cyan arrow.

The intersection of the diamond edges ΔV_{SD} , where $\mu_{N-1} = \mu_s$ and $\mu_N = \mu_D$, can be used to estimate the addition energy of the quantum dot, i.e. $E_{\text{add}} = 26.9 \text{ meV}$. Additionally using the width of the Coulomb diamond ΔV_G , the gate lever arm can be estimated to be $\alpha_G = \Delta V_{SD} / \Delta V_G = 0.023$. Making use of the capacitance model for a quantum dot, described in Chapter 3, additional parameters can be extracted. The coupling capacitances of the leads, C_S and C_D , as well as the coupling capacitance of the gate, C_G , give an insight into how well the quantum dot is coupled to the electronic components. The width of the Coulomb diamond can be used to calculate the coupling capacitance of the dot to the gate, $C_G = |e| / \Delta V_G = 0.14 \text{ aF}$. The slopes of the diamond edges can be used to extract the coupling capacitances to the source- and drain-electrodes, $C_S = 2.7 \text{ aF}$ and $C_D = 3.1 \text{ aF}$. From this we can see that there is a slight coupling asymmetry between the two electrodes to the carbon nanotube, which explains the tilt of the Coulomb diamonds with respect to the horizontal axes in Figure 7.3 (b). The capacitance from the dot to the gate is roughly one-order of magnitude smaller than the capacitances to the leads. This is a direct result of the side-gate geometry and the very strong screening of the gate due to the source- and drain-contacts.

The resonances due to excited state transitions only occur when they fall into the bias window and their relaxation rate is smaller than the tunnelling rate through the dot, see Chapter 3. The excited state transitions in Figure 7.3 (b) indicated by the purple arrows, can be used to estimate the single particle excitation energy Δ , which is given by

$$\Delta = \mu_N^{(1,0)} - \mu_N^{(0,0)} = |e|\alpha_G \Delta V_G^{\text{ex}}, \quad (7.1)$$

where ΔV_G^{ex} is the distance in gate voltage between the ground and excited state conductance resonance. The measurement in Figure 7.3 (b) leads to $\Delta = 3.1$ meV, which can be used to estimate the charging energy of the quantum dot via $E_C = E_{\text{add}} - \Delta = 23.8$ meV.

7.4 Carbon Nanotubes with Superconducting Leads

In this section transport measurements of carbon nanotubes contacted with a superconductor-normal metal bilayer (Pd/Al, 4/80 nm) are presented. As mentioned earlier, the Pd/Al superconducting bilayer has been developed in previous PhD work by *A. Nersisyan* [137] and was optimised to achieve highly transparent contacts to carbon nanotubes, while also exhibiting a reasonably large superconducting gap. These carbon nanotube devices show regular quantum dot behaviour for biases larger than the superconducting gap Δ . However, for smaller biases the devices effectively behave like SNS Josephson junctions, which is due to the superconducting proximity effect. The devices are measured in two different configurations, i.e. in voltage bias and in current bias. The experiments described in the following sections were performed on the same device during a single cool-down, unless stated otherwise.

7.4.1 Voltage Bias Measurements: Coulomb Blockade and Conductance Oscillations

The device contacted with Pd/Al contacts was first measured in voltage bias. For $V_{\text{SD}}|e| > 2\Delta$ normal conduction behaviour is expected. The induced superconducting

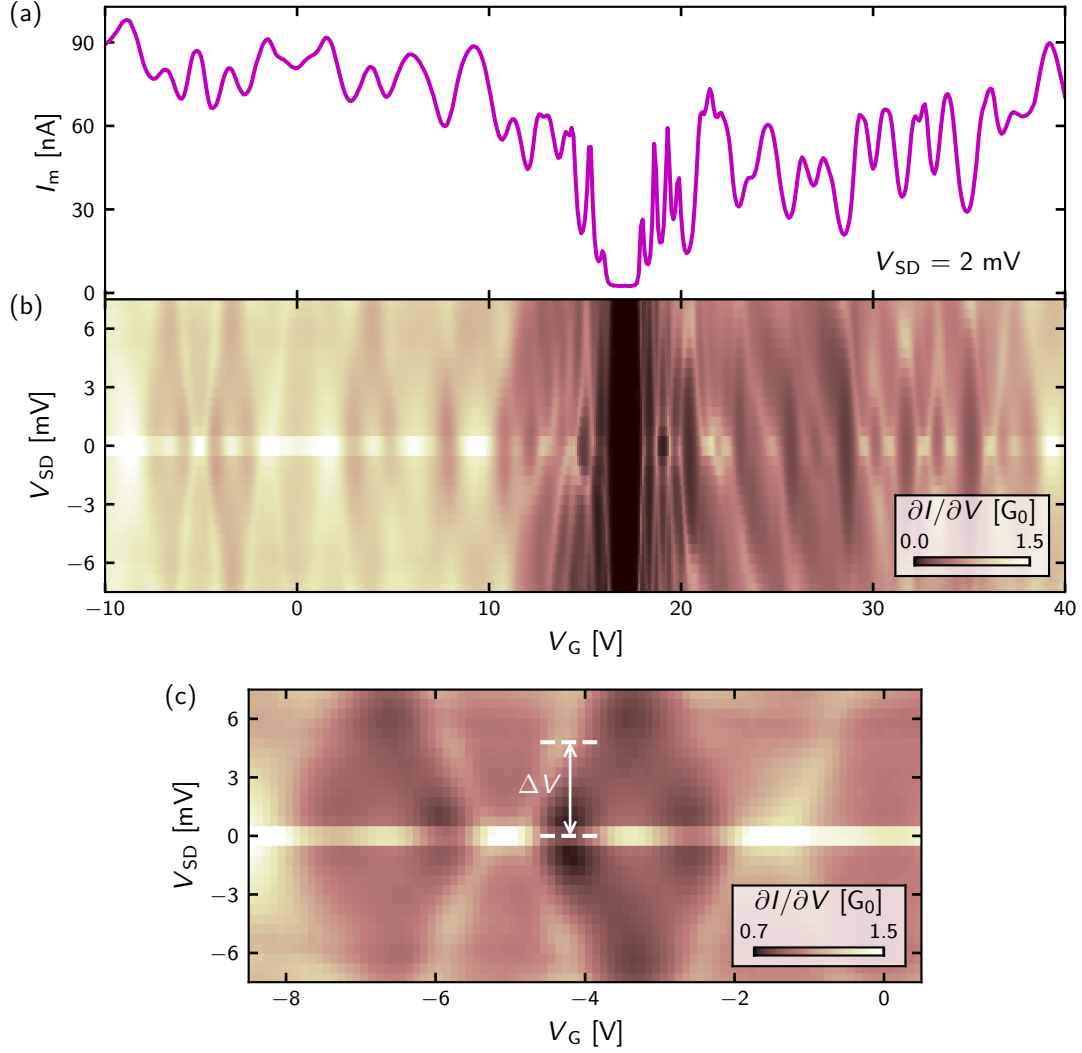


Figure 7.4: Voltage biased measurements of a carbon nanotube Josephson junction. (a) Current I_m through the carbon nanotube as a function of V_G at $V_{SD} = 2$ mV. For $V_G > 18$ V electron transport is observed, initially in the form of Coulomb blockade followed by conductance fluctuations. For $V_G < 16$ V hole transport is observed in the form of Coulomb blockade and conductance fluctuations. The semiconducting gap of the carbon nanotube is observed for $16 \text{ V} < V_G < 18 \text{ V}$. (b) Stability diagram of a carbon nanotube Josephson junction, measured by tuning V_G and V_{SD} . On either side of the semiconducting gap a few Coulomb diamonds can be observed. Further away from the semiconducting gap the transport is dominated by conductance oscillations with possible signatures of Fabry-Pérot type interferences. The high conductance around $V_{SD} = 0$ mV is due to the proximity induced supercurrent. (c) Zoom in of stability diagram in (b). Signs of Fabry-Pérot interferences can be observed with a resonance spacing of $\Delta V = 4.8$ mV. As in (b) the high conductance around zero bias is due to the supercurrent of the Josephson junction.

gap of the device was measured to be $\Delta \approx 90 \mu\text{eV}$, see Section 7.4.2. This is about three times smaller than the value for bulk Aluminium and also a reduction for thin

Al films, which is due to the proximity induced superconductivity in the bi-layer and the carbon nanotube. Voltage biased measurements are presented in Figure 7.4. Panel (a) of Figure 7.4 shows the measured current I_m through the carbon nanotube as a function of the gate voltage V_G at a finite voltage-bias $V_{SD} = 2$ mV at a temperature of $T = 10$ mK. From this measurement the current transport can be divided into two regimes which are separated by the semiconducting gap at $16 \text{ V} < V_G < 18 \text{ V}$. For $V_G < 16 \text{ V}$ the charge carriers contributing to the current are holes (p-type), whereas for $V_G > 18 \text{ V}$ the charge carriers are electrons (n-type). Close to the semiconducting gap both regimes exhibit Coulomb peaks in the current. When tuning V_G further away from the semiconducting gap, the current exhibits irregular oscillations on either side.

The stability diagram of the carbon nanotube device is shown in Figure 7.4 (b). The conductance of the device varies between 0 and $1.5G_0$. The highest conductance value is observed in the superconducting gap as here the proximity induced super-current contributes to the conductance. Similar as before, the semiconducting gap of the carbon nanotube can be observed by the most prominent feature of the stability diagram, i.e. for $16 \text{ V} < V_G < 18 \text{ V}$ where the conductance is 0. For $V_G > 18 \text{ V}$ Coulomb diamonds can be observed for the first few volts. At higher voltages the conductances exhibits oscillations as well as for $V_G > 29 \text{ V}$ again signs of Coulomb diamonds. For $V_G < 16 \text{ V}$ initially signs of Coulomb blockade, i.e. Coulomb diamonds can be observed. When V_G is tuned to values below 10 V the device becomes a lot more conductive and the stability diagram shows patterns of conductance oscillations. For $0 \text{ V} < V_G < 5 \text{ V}$ and negative gate voltages the stability diagram exhibits tentative signs of the checkerboard pattern usually produced by Fabry-Pérot interference [206]. Figure 7.4 (c) shows a zoom in of the stability diagram for $V_G < 0 \text{ V}$. Here the checkerboard pattern is more clearly visible and a Fabry-Pérot resonance spacing $\Delta V = 4.8 \pm 0.3 \text{ mV}$ is measured, corresponding to a channel length $L = 349 \pm 20 \text{ nm}$. This is close to the resonance spacing calculated from the device geometry $\Delta V = h\nu_F/(2Le) \approx 5.5 \text{ mV}$, where $L = 300 \text{ nm}$ is the length in the design and $\nu_F = 8.1 \times 10^5 \text{ m/s}$ [206]. This is a strong indication, that

the observed features result from Fabry-Pérot interference. In general the measured conductance appears to be higher in the p-type transport regime compared to the n-type transport regime, see Figure 7.4 (b). This is as expected from carbon nanotubes contacted with Pd, as Pd p-dopes the nanotube and hence provides a higher transparency for holes than for electrons.

7.4.2 Current Bias Measurements

7.4.2.1 Supercurrent and Andreev Reflections

Current bias measurements of SNS Josephson junctions can be used to determine their Josephson energy by measuring the critical current I_c . In Figure 7.5 current bias measurements of the carbon nanotube contacted with Pd/Al leads are presented. Here, the device is biased with I_{SD} from a current source, and the voltage drop V_m across the carbon nanotube is measured as a function of I_{SD} and gate voltage V_G . The differential resistance $\partial V/\partial I$ can then be obtained by differentiating each I-V trace numerically. Panel (a) and (b) of Figure 7.5 show the numerical differential resistance of the carbon nanotube as a function of I_{SD} and V_G in a region of n-type (a) and p-type (b) conduction. Both of the measurements exhibit resonances modulated by V_G which are due to multiple Andreev reflections of different order. The white region around $I_{SD} = 0$ nA, i.e. where $\partial V/\partial I = 0$ k Ω , is where the Josephson junction supports a supercurrent due to the proximity-induced superconductivity. The primary sharp resonance describes the critical current I_c of the junction which is modulated by V_G . The highest resonance marks the boundary to quasiparticle transport, which is observed in both (a) and (b) for $I_{SD} \gg I_c$.

The critical current observed in the n-type conduction can be modulated with the gate voltage from $I_c = 0$ nA up to a maximum of $I_c = 2.6$ nA. Across the whole gate region shown in Figure 7.5 (a) the average critical current is $\langle I_c \rangle \approx 0.4$ nA. Both of these quantities are significantly higher in the p-type conduction regime, shown in Figure 7.5 (b). Here the critical current can be tuned from $I_c = 0$ nA up to $I_c = 15.8$ nA. The average critical current observed in the p-type conduction regime is $\langle I_c \rangle \approx 5$ nA. The larger critical current in the p-type conduction regime compared

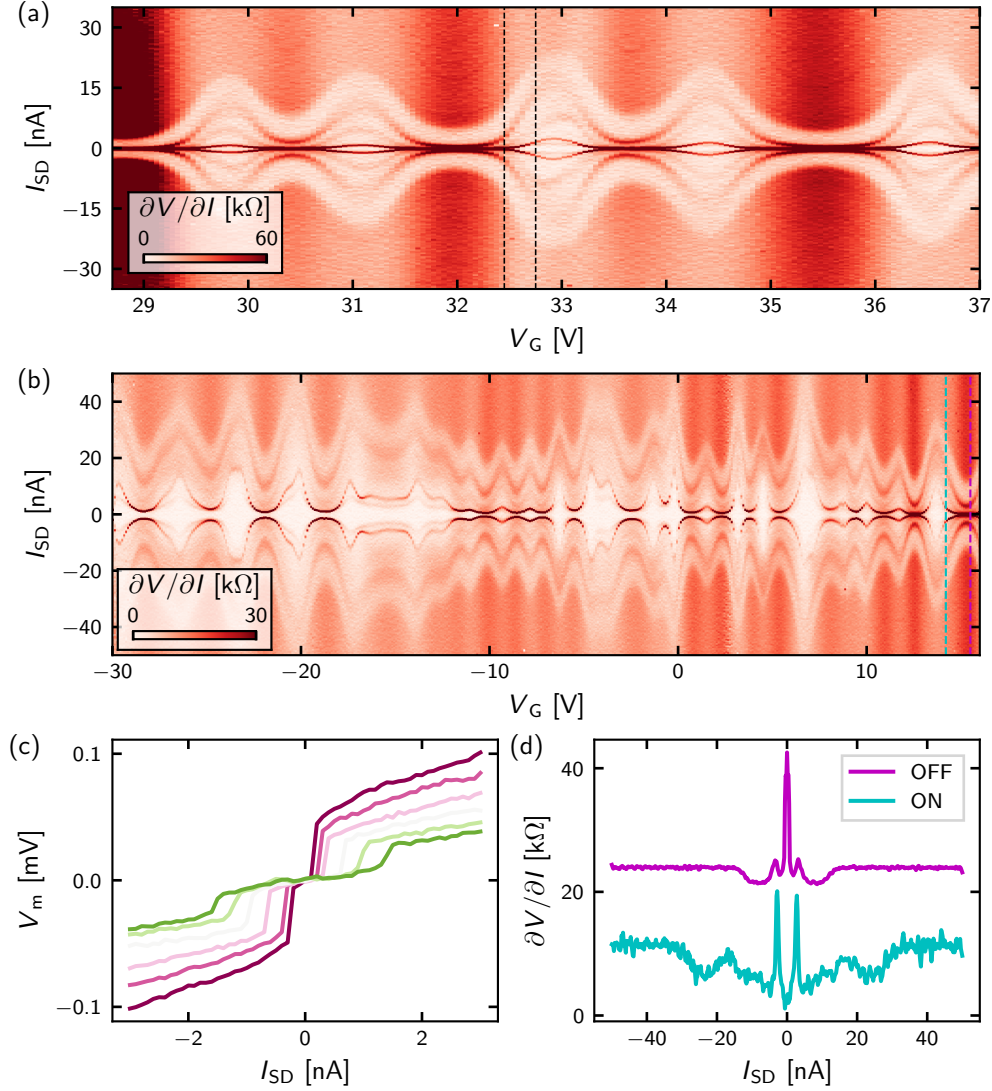


Figure 7.5: Current biased measurements of a carbon nanotube Josephson junction. (a) & (b) Differential resistance of the carbon nanotube as a function of the bias current I_{SD} and the gate voltage V_G for intervals of V_G in the n-type (a) and p-type (b) conduction regime. The resonances modulated with V_G are due to multiple Andreev reflections of different order. The first sharp resonance marks the critical current I_c of the Josephson junction. The white region near zero bias current, i.e. where $\partial V/\partial I = 0 \text{ k}\Omega$, is due to proximity-induced supercurrent flowing through the nanotube. The uniform region for large I_{SD} is due to quasiparticle transport. (c) I - V curves of the junction at various gate voltages (region marked in (a) with black dashed lines). The critical current I_c , at which a sudden transition to a finite resistance is observed, is tuned with V_G . (d) Differential resistance curves at two different gate voltages marked in (b), where either a supercurrent is observed or it is suppressed. This corresponds to 'On' and 'OFF' states of a supercurrent transistor. Curves are offset by 20 $\text{k}\Omega$ for clarity.

to the n-type conduction regime is to be expected, as I_c is linearly dependent on the transmission probability T_i for each conduction channel i of the leads. As

previously mentioned, Pd has higher transparency for holes which gives rise to the imbalance in I_c . However, the measured I_c is significantly lower than the estimated value for leads with transmission probability $T_i = 1$, $I_c^{\text{ideal}} = 2e\Delta/\hbar = 43.8 \text{ nA}$ (cf. Equation 3.11), where Δ is the induced superconducting gap of the carbon nanotube Josephson junction.

Single I-V traces at different V_G , shown in Figure 7.5 (c), exhibit a current at which a sudden transition to a finite resistance occurs. This is the critical current I_c of the carbon nanotube Josephson junction. The residual resistance, i.e. non-vanishing voltage, for $I < I_c$ is attributed to phase diffusion in an overdamped Josephson junction, see Section 7.4.2.2. The same effect leads to the rounding of the I-V curve around this critical current step. Since the critical current can be modulated by V_G from 0 to a finite value, this can be used similarly to a transistor in order to switch a supercurrent on and off [155], see Figure 7.5 (d). The resonance away from the critical current step occurs due to Andreev reflections.

Since in a current biased measurement the voltage drop across the device is measured, it is possible to interpolate the data in order to convert it to a voltage bias. The differential conductance converted from the measurements presented in Figure 7.5 (a-b) is presented in Figure 7.6. In both datasets high conductance around zero bias is attributed to the supercurrent. Additionally, Figure 7.6 (a-b) exhibit pronounced peaks at specific gate-independent V_{SD} . The energy above which quasiparticle transport dominates the conductance appears as a flat line at $V_{SD} = 2\Delta/e = 180 \text{ } \mu\text{V}$. Similarly, multiple Andreev reflections appear as gate-independent resonances at $V_{SD} = 2\Delta/ne$ with $n \in \mathbb{N}^+$. In this device only Andreev reflections with $n = 1, 2, 4$ are visible and are indicated with dashed lines in Figure 7.6. From the Andreev reflections and the boundary to the quasiparticle transport it is possible to extract the superconducting gap $\Delta \approx 90 \text{ } \mu\text{eV}$. This is slightly smaller than the value measured in similar devices $\Delta = 125 \text{ } \mu\text{eV}$ [137], which could be attributed to the contact transparency, calibration changes in the evaporator (i.e. film thicknesses vary) or spurious magnetic fields.

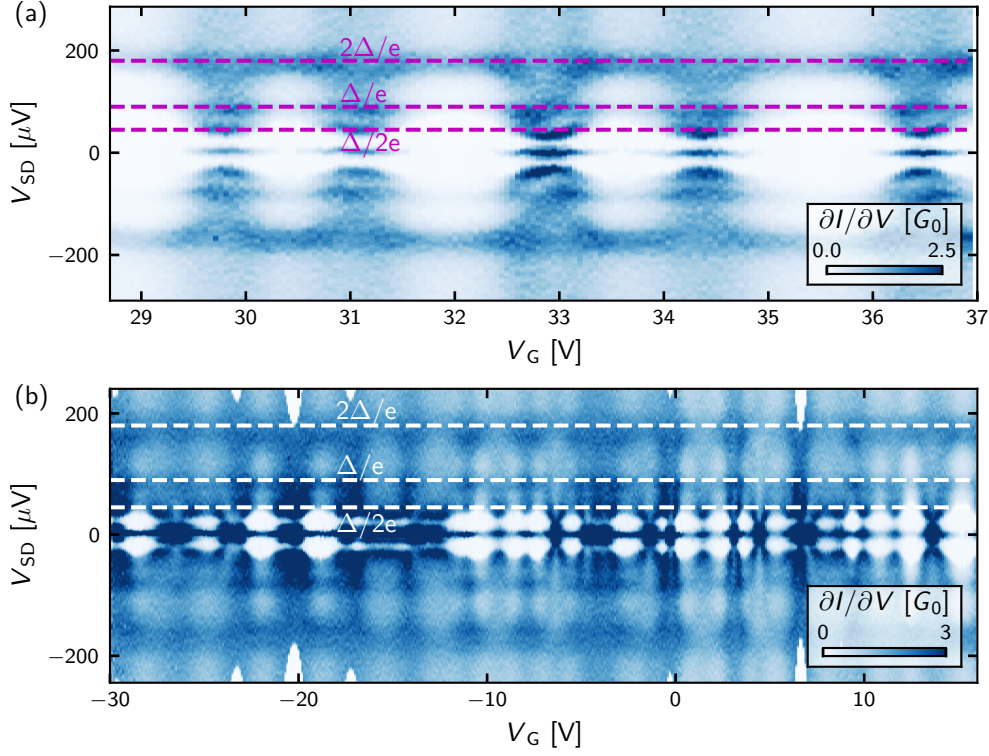


Figure 7.6: (a) & (b) Numerical differential conductance, converted from current biased measurements as a function of gate voltage and bias across the device. Multiple Andreev reflections appear as gate-independent resonances at $V_{SD} = 2\Delta/ne$, where Δ is the superconducting gap and $n \in \mathbb{N}^+$, however here only Andreev reflections with $n = 1, 2, 4$ are visible (indicated by purple (a) and white (b) dashed lines). The large conductance around zero bias is due to the supercurrent.

7.4.2.2 Residual Sub-Gap Resistance and Thermal Phase Diffusion

The measurements presented in Figure 7.5 show a residual differential resistance for $I < I_c$ in some of the supercurrent regions. A zoom-in of such a regime is shown in Figure 7.7 (a). Cutting along the blue dashed line in Figure 7.7 (a), the voltage drop across the device and the differential resistance can be plotted as a function of I_{SD} , see Figure 7.7 (b) and Figure 7.7 (c) respectively. In both cases the critical current shows up as a clear feature, marked by the purple dashed line. In the region around $I_{SD} = 0$ nA, between the two cyan dashed lines, no voltage drop across the device and no residual resistance is observed¹. Hence, the carbon nanotube Josephson junction carries a supercurrent in this regime. In the grey shaded region

¹It is worth mentioning that all residual resistances due to measurement lines, etc. are calibrated out of the data presented.

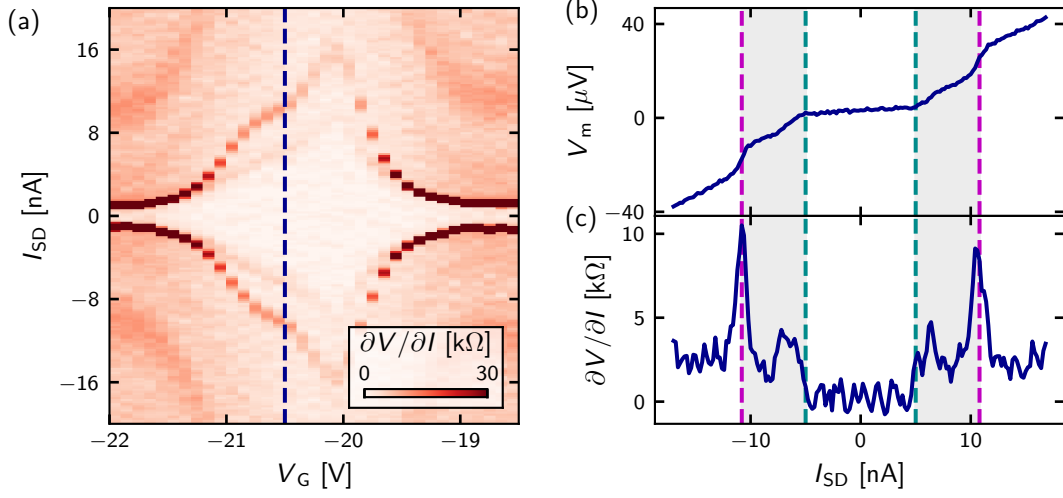


Figure 7.7: Sub-gap resistance in a carbon nanotube Josephson junction. (a) Zoom-in of differential resistance plot given in Figure 7.5 (b). The critical current step is clearly observable. Inside the supercurrent regime ($I_{SD} < I_c$) a finite residual resistance is observable. (b) I-V trace along dashed line in (a). (c) Differential resistance trace along dashed line in (a). (b) & (c) The critical current is indicated by the purple dashed line. The grey shaded area between the purple and cyan dashed line indicates the region with a finite sub-gap resistance, whereas between the two cyan dashed lines no residual resistance is observed.

of Figure 7.7 (b-c) a residual voltage or finite resistance is observed for $I_{SD} < I_c$.

This residual voltage or resistance can be attributed to thermally induced phase diffusion since the Josephson energy E_J is on the same order as the thermal electron energy (~ 100 mK). Due to the symmetry of I_c around zero bias, i.e. no hysteresis (see also Figure 7.7 (a-c)), the carbon nanotube Josephson junction is in the overdamped Josephson junction regime. For overdamped Josephson junctions it has been predicted by *Ambegaokar et al.* [207], that thermal noise can lead to phase diffusion which results in a non-vanishing junction voltage for $I < I_c$, giving rise to the residual differential resistance for $I < I_c$. This can be explained with the picture of the tilted washboard potential of the phase of a Josephson junction [99], see Section 2.5.1.1. At a finite current bias $I < I_c$ the thermal excitation is large enough to change the potential energy of the phase particle and in turn change the phase of the junction, which according to Equation 2.14 gives rise to a residual voltage, i.e. residual differential resistance. Additionally, the phase diffusion leads to a rounding of the I-V curve around I_c as the phase can already

vary for $I < I_c$. This behaviour has previously been simulated and observed in small capacitance Josephson junctions [208] as well as measured in carbon nanotubes and InAs nanowires contacted with superconducting leads [209, 210].

7.4.3 RF Reflectometry Measurements: Critical Current and Andreev Reflection

Radio frequency (RF) reflectometry measurements offer a highly sensitive method for electrostatic measurements [211]. Here, RF reflectometry is used to characterise a carbon nanotube Josephson junction as a function of I_{SD} and V_G , see Figure 7.8. The device measured in this section is similarly fabricated to the device presented in Section 7.4.1 and Section 7.4.2, but originates from a different fabrication run and chip. Panel (a) of Figure 7.8 shows the differential resistance of the carbon nanotube as a function of I_{SD} across a large region of V_G , very likely covering both the n-type as well as the p-type conduction regime. The white region around $I_{SD} = 0$ nA, i.e. where $\partial V / \partial I = 0$ k Ω , is where the Josephson junction carries a supercurrent due to the proximity induced superconductivity. The first resonance marks the point in bias current I_{SD} , i.e. the critical current I_c , where the Josephson junction turns normal. The critical current of the device can also be modulated with a gate voltage V_G , where $0 \text{ nA} < I_c < 18 \text{ nA}$. The maximum critical current of $I_c \approx 18 \text{ nA}$ observed in this device is slightly larger than in the device measured in Section 7.4.2, which as far as we are aware is the highest critical current observed in single Josephson junction devices based on carbon nanotubes to date. Previously carbon nanotube Josephson junctions in a SQUID architecture were reported to exhibit a single junction critical current of $I_c = 12 \text{ nA}$ [212]. A second faint resonance appears for $I_{SD} > I_c$ which is due to an Andreev reflection, cf. Figure 7.8 (c) cyan arrows. The more uniform region for $I_{SD} \gg I_c$ is where quasiparticle transport is dominant, cf. Figure 7.8 (c) shaded region. The region of high differential resistance between $0 \text{ V} < V_G < 10 \text{ V}$ likely stems from the semiconducting gap of the carbon nanotube. Similar to the device measured in Section 7.4.2, larger critical currents

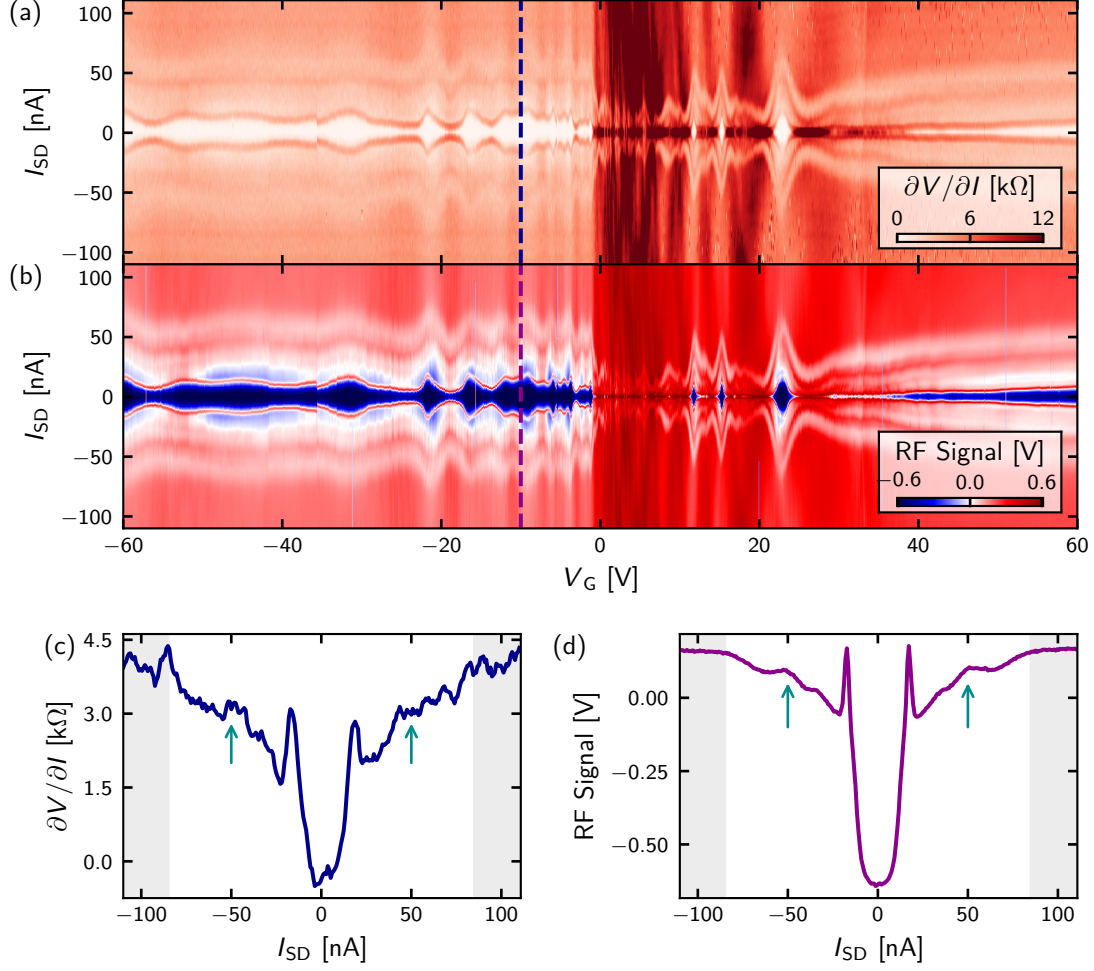


Figure 7.8: Current biased measurements of a carbon nanotube Josephson junction measured in DC transport (a) & (c) and RF reflectometry (b) & (d). (a) Differential resistance of the carbon nanotube as a function of I_{SD} and V_G . The white region near zero bias current where $\partial V/\partial I = 0$ kΩ is due to the dissipationless supercurrent flowing through the nanotube. The critical current I_c of the Josephson junction appears as the first sharp resonance. One faint resonance marks an Andreev reflection and the uniform region for large I_{SD} is due to quasiparticle transport. The darker region between $0 \text{ V} < V_G < 10 \text{ V}$ marks the band gap of the carbon nanotube. (b) Simultaneously measured RF reflectometry signal reflected from the carbon nanotube Josephson junction. All features observed in (a) also show up in the RF signal but with a higher contrast. (c) Differential resistance trace along the dashed line in (a) for $V_G = -10 \text{ V}$. I_c appears as two sharp peaks symmetrically around $I_{SD} = 0 \text{ nA}$. The faint signal from an Andreev reflection is indicated with cyan arrows and the region of quasiparticle transport is shaded grey. (d) RF signal trace along the dashed line in (b) for $V_G = -10 \text{ V}$. Two sharp symmetric peaks around $I_{SD} = 0 \text{ nA}$ mark I_c . The signal from an Andreev reflection is indicated with cyan arrows and the region of quasiparticle transport is shaded grey.

are found in the p-type conduction regime ($V_G < 0 \text{ V}$) compared to I_c observed in the n-type conduction regime ($V_G > 10 \text{ V}$).

Simultaneous to measuring the carbon nanotube in DC transport, the reflected RF signal of the carbon nanotube device can also be recorded², see Figure 7.8 (b). Here, an RF signal of frequency $f_{\text{RF}} = 261.2$ MHz and power $P_{\text{in}} = -58$ dBm is sent to the sample board and the reflection is recorded in a homodyne detection scheme, see Section 5.5 for details. The reflected signal as a function of I_{SD} and V_{G} exhibits the same characteristics as the DC measurement presented in Figure 7.8 (a). A line cut of the RF signal at $V_{\text{G}} = -10$ V is shown in Figure 7.8 (d), which exhibits two clear symmetric peaks around $I_{\text{SD}} = 0$ nA indicating the critical current of the Josephson junction. Additionally, the Andreev reflection shows up as a distinct increase in the RF signal, see Figure 7.8 (d) cyan arrows. The region of quasiparticle transport is also very apparent as a flat signal response after an initial increase due to the conductance change of the device, see Figure 7.8 (d) shaded region.

Comparing the two line cuts at $V_{\text{G}} = -10$ V of the differential resistance (Figure 7.8 (c)) and the RF signal (Figure 7.8 (d)), it is quite obvious that the RF reflectometry measurement offers an enhanced contrast for the critical current, the Andreev reflection as well as the quasiparticle transport as opposed to the DC measurement. This is due to the fact, that the RF reflectometry circuit is very sensitive to changes in the conductance or inductance of the measured device, cf. Section 5.5. Particularly the conductance changes very drastically around the critical current which leads to a strong response in the measured RF signal. Additionally, due to measuring at a finite RF frequency $1/f$ noise is reduced resulting in a better signal-to-noise ratio [213] compared to the DC measurement, see Reference [173] for an in depth discussion of noise reduction in RF reflectometry measurements.

7.5 Possible Superconducting Qubit Parameters

As one of the motivations for the work in this Chapter was to build a superconducting qubit based on a carbon nanotube Josephson junction (see Chapter 8) the feasibility of this endeavour is discussed here. The most integral part of a superconducting

²The RF signal is acquired with the same resolution in I_{SD} and is averaged the same amount as the DC signal.

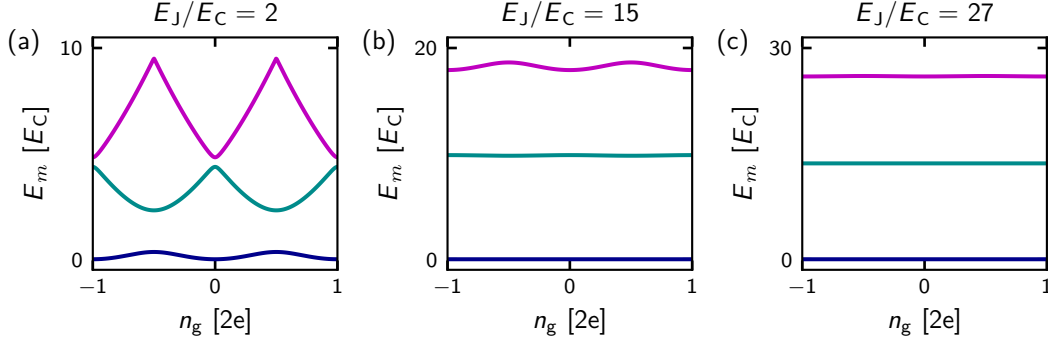


Figure 7.9: First three energy levels of a superconducting qubit based on the analytic solutions of the Cooper pair box (CPB) Hamiltonian, given by Equation 2.28, for different ratios of E_J/E_C as a function of the gate charge n_g . (a) Energy levels in the CPB limit for $E_J/E_C = 2$, where charge dispersion is very dominant. (b) Intermediate regime of the energy levels for $E_J/E_C = 15$, here only the third level exhibits a slight charge dispersion. (c) transmon regime of the CPB for $E_J/E_C = 27$. The energy levels are flat and hence resilient to charge noise.

qubit is a capacitively shunted Josephson junction and its parameters determine the energy scales of the qubit. The full Hamiltonian of a Cooper pair box (CPB), see Equation 2.27, includes two characteristic energies, the charging energy E_C and the Josephson energy E_J . The ratio of these two E_J/E_C determines the susceptibility of the qubit to charge noise. Hence ideally E_J/E_C should be large, i.e. $E_J/E_C > 20$ in the transmon regime. In Table 7.1 the design parameters of a superconducting qubit with a qubit frequency $f_Q = 5$ GHz are listed for different critical currents I_c of the Josephson junction. Additionally, Figure 7.9 shows the first three energy levels of a CPB for the three configurations given in Table 7.1. For the average critical current $I_c = 5$ nA obtained in the carbon nanotube Josephson junctions, the E_J/E_C ratio needed for a qubit frequency of $f_Q = 5$ GHz is quite low and would lead to a strong charge dispersion, i.e. a CPB type superconducting qubit. Additionally, the needed capacitance C_Σ is relatively small which is difficult to achieve in the design chosen for the readout resonators, cf. Section 4.2.2.2. However, for the highest critical current of $I_c = 15$ nA achieved in the carbon nanotube Josephson junctions $E_J/E_C = 15$, which is fairly close to the transmon regime. From Figure 7.9 (b) it is evident that for $E_J/E_C = 15$ the charge dispersion is already strongly reduced for the first two energy levels and hence favourable compared to $E_J/E_C = 2$. Further, $C_\Sigma = 38.7$ fF is easier to be achieved/fabricated in the chosen resonator geometry. In order to

I_c [nA]	E_J/h [GHz]	f_Q [GHz]	E_C/h [GHz]	C_Σ [fF]	E_J/E_C
5	2.48	5.0	1.12	17.3	2
15	7.85	5.0	0.50	38.7	15
20	9.94	5.0	0.36	53.8	27

Table 7.1: Superconducting qubit design parameters at a qubit frequency of $f_Q = 5$ GHz, for different critical currents I_c of the Josephson junction.

reach the transmon regime, i.e. $E_J/E_C > 20$, a critical current $I_c \approx 20$ nA would have to be obtained for the carbon nanotube Josephson junction. Potential avenues for increasing E_J in the carbon nanotube Josephson junctions will be discussed at the end of the next Chapter, see Section 8.4.

In the next Chapter a first implementation of a superconducting qubit based on a carbon nanotube Josephson junction will be presented. The qubit's charging energy is designed to be around $E_C/h \approx 500$ MHz, as this for high enough I_c will allow for operation as close as possible to the transmon regime. Since it has been observed in this Chapter that I_c can be modulated with V_G , this effect will be used to tune E_J and in turn f_Q of the qubit in the following experiments.

7.6 Conclusion/Summary

In this Chapter DC transport measurements of carbon nanotubes were discussed in detail. At first the device geometry, the fabrication and device pre-selection at room-temperature have been reviewed. Two types of devices contributed to the results from this Chapter, both of which used Pd as the main contacting material. This resulted in high transparency contacts with large conductance of the devices. The first device was solely contacted with Pd and its transport characteristics were explained. Additionally, device parameters such as coupling capacitances, gate-lever arm and addition energy of the quantum dot were extracted. The single quantum dot parameters are on the same order as previously measured with the exact same device geometry [137].

In a second device the carbon nanotube was contacted with a Pd/Al bilayer to study the Josephson junction behaviour due to the induced superconductivity.

The Josephson junction was found to be in the overdamped junction regime and exhibited gate modulated critical currents up to $I_c = 15.8$ nA, which is ~ 2 times higher than the critical current observed in similar devices previously measured in the group [137]. Additionally, multiple Andreev reflections could be observed and the induced superconducting gap was extracted $\Delta \approx 90$ μ eV. Further, a residual sub-gap resistance was observed in the device, which could be attributed to thermal phase diffusion.

Another device made from a carbon nanotube with Pd/Al superconducting leads was measured simultaneously in DC and with RF reflectometry. The critical current was studied over a wide range of gate voltages and exhibited modulation up to $I_c \approx 18$ nA, which as far as we are aware is the highest critical current reported in single carbon nanotube Josephson junctions to date. Comparing the DC measurements with the RF reflectometry measurements indicated that the RF reflectometry measurements yield a higher contrast for changes in the conductance of the device. This showcases that RF reflectometry is an interesting and faster measurement method for characterising novel types of Josephson junctions. Especially due to its large sensitivity to conductance changes in the device it could potentially be used for more in-depth studies of Andreev physics. Additionally, since the RF reflectometry circuit should also be susceptible to changes in the inductance of the device, measuring changes in resonance frequency of the circuit would allow for an indirect analysis of the Josephson inductance of SNS Josephson junctions.

In the last section of this chapter, possible parameters for a superconducting qubit based on a carbon nanotube Josephson junction were discussed. This showed that with the Josephson junctions measured in this chapter it should be possible to implement such a qubit. Although, it seems not possible to reach the transmon regime with the observed values of I_c , an intermediate regime of E_J/E_C already offers reduced charge dispersion. This implies a carbon-nanotube-based superconducting qubit is feasible to achieve and a first implementation of such a qubit is presented in the following chapter.

8

A Carbon-Nanotube-Based Superconducting Qubit

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Up to today, most circuit QED implementations for quantum information processing or the study of the underlying fundamental physics, utilise superconducting qubits based on standard Al/Al₂O₃ Josephson junctions (JJ). In this chapter a first realisation and thorough measurements of superconducting qubits with carbon-nanotube-based Josephson junctions are presented. This demonstrates a new type of superconducting qubit with a gate voltage tunable transition frequency.

8.1 Introduction

Conventional Al/Al₂O₃ Josephson junction qubits can be either fixed frequency (single JJ) or frequency tunable via a SQUID loop (two JJs). The tunability from these types of qubits arises from the fact that a magnetic flux threaded through the loop changes the Josephson energy of the SQUID, which in turn effects the transition frequency of the qubit. However, this flux tuning suffers from instabilities due to magnetic flux noise and does not allow operation in magnetic fields, i.e. heavy magnetic field screening needs to be undertaken in order to ensure good coherence.

Here, the tunable element of a superconducting qubit, i.e. the SQUID, is replaced with a carbon nanotube contacted with superconducting leads. The carbon nanotube effectively acts as a SNS Josephson junction. A gate allows for the tuning of the electrostatic potential inside the carbon nanotube which modulates the critical current in the junction, i.e. tunes the Josephson energy of the junction. Since this design only relies on voltages rather than magnetic fluxes, the qubit is not susceptible to magnetic flux noise and depending on the type of superconductor chosen for the leads, allows for operation in reasonable magnetic fields. Additionally, electric fields are much easier to localise compared to magnetic fields, which makes complex multi-qubit devices simpler to realise. Similar type of qubits, i.e. gatemon qubits, have been implemented before with InAs nanowires [50, 51], two-dimensional electron gases [214] and graphene [107]. However, the device presented here is a first implementation of a gatemon type superconducting qubit based on a carbon nanotube.

8.2 Device

The carbon nanotube superconducting qubit device is mounted in a purposely machined copper sample holder and wire-bonded to a specifically designed copper PCB, which allows for microwave and DC-voltage connections to the sample chip, see Figure 8.1 (a). The devices measured for contribution to this chapter are all based on the same layout, see Figure 8.1 (b). Each microwave chip consists of 10

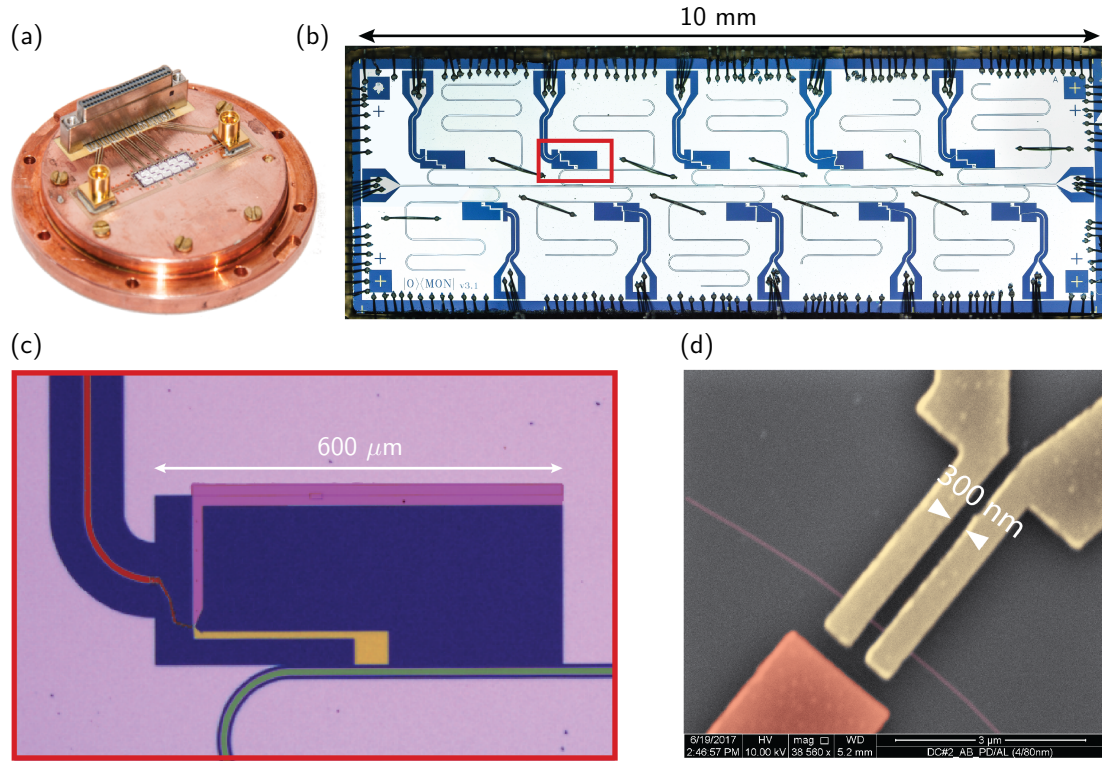


Figure 8.1: Carbon nanotube superconducting qubit device. (a) Image of the copper sample holder including bonded device chip in the centre. Microwave ports (gold) and the DC connection (silver) to the PCB are visible. (b) Optical image of the qubit device chip. A microwave transmission line in the centre addresses 10 multiplexed $\lambda/4$ resonators with different resonance frequencies. Each resonator has a single DC line and a window in the ground plane close to its electric field anti-node for qubit fabrication. (c) False color optical image of a single qubit with the island (gold) coupled to the resonator (green) and the other side shorted to ground (purple). A side gate (red) is provided via the DC line. (d) False color SEM image of a carbon nanotube (pink) contacted with two superconducting contacts (gold) separated by 300 nm and a side gate (red).

$\lambda/4$ resonators with different resonance frequencies, which can be addressed via multiplexed readout on a single microwave transmission line. Each resonator has a dedicated window in the ground plane close to its electric field anti-node, where the qubit can be fabricated. Additionally, each of these windows has its own DC line.

For the device fabrication, at first carbon nanotubes are grown via chemical vapour deposition on a Si/SiO₂ substrate. The microwave resonators are then patterned via UV-photolithography or e-beam lithography. Prior to metal deposition an oxygen plasma etch is done to ensure left-over carbon nanotubes are not shorting the resonators or the gate lines to ground. After lift-off the chip is imaged with an SEM in order to locate and pick a suitable carbon nanotube for the qubit.

The contacts to the carbon nanotube and islands of the qubit are then patterned with e-beam lithography, post development cleaned with a UV ozone cleaner and metalised with a Pd/Al (4/80 nm) bi-layer.

After fabrication the qubit devices are tested and pre-selected at room-temperature. For this the room-temperature resistance of the carbon nanotube Josephson junctions is measured on a DC probe-station. Working devices exhibit room-temperature resistances between $7 \text{ k}\Omega < R_n < 100 \text{ k}\Omega$, leading to desired critical currents of the junctions.

8.3 Measurements

The following sections describe the experiments to characterise a first implementation of a superconducting qubit based on a carbon nanotube Josephson junction. Three devices which showed similar characteristics were measured, i.e. *CNTQubit_v2_Q1* (v2_Q1), *CNTQubit_v2_Q5* (v2_Q5), *CNTQubit_v3_Q3* (v3_Q3). The device architecture was the same for all three of them, except that in v3_Q3 the coupling of the resonator to the transmission line was increased, see Section 4.2.1.2. This was done in order to better match the internal to the external Q-factor of the resonators which was necessary as left-over catalyst from the carbon nanotube growth significantly reduced the internal Q-factor of the resonators. The results presented in this chapter are from device *v3_Q3* and *v2_Q1* unless stated otherwise. Equivalent measurements of the other device exhibiting a similar behaviour can be found in Appendix C.

8.3.1 Resonator and Qubit Spectroscopy

The system is prepared in its ground state $|g, 0\rangle$ by cooling it to temperatures below 20 mK in a dilution refrigerator. As a first standard characterisation of the device, spectroscopy of the resonator is performed with the qubit in its ground state $|g\rangle$. For this measurement the RF microwave signal generator (cf. Section 5.4) is set to a fixed power of -50 dBm and its frequency is swept across the resonance frequency of one of the 10 resonators on the chip, see Section 4.2.2.2. The transmitted signal

is downconverted to an intermediate frequency (cf. Section 5.4) and its amplitude measured versus frequency. The resonance of a weakly driven damped harmonic oscillator exhibits a Lorentzian line shape (cf. Eq. 4.10 and Eq. 4.11). From this it is possible to extract the exact resonance frequency f_0 as well as the internal (Q_i) and external (Q_{ext}) quality factor of the resonator, see Section 4.2.2.2.

Since the resonator and the qubit are a coupled system and cannot be separated, f_0 depends on the detuning Δ of the qubit to the resonator as well as the state of the qubit. We assume that the qubit is cold enough, such that almost all of its population is in its ground state. However, due to the side-gate we are able to tune the frequency of the qubit. Therefore, resonator spectroscopy was performed as a function of the gate voltage applied to the carbon nanotube. Figure 8.2 (a) shows a colormap of such a measurement on device v3_Q3, where the gate voltage V_G is swept after taking a frequency sweep f_r of the transmitted amplitude S_{21} . For $V_G < -30$ V the resonance frequency of the resonator is $f_0 = 5.572$ GHz and relatively constant. Increasing V_G past -20 V, the resonator is shifting upwards in frequency for specific gate voltage values. For -15 V $< V_G < 5$ V the device seems to exhibit a quasi-periodic pattern. The shift upwards in frequency is a first indication that there is a qubit present with transition frequency below the resonator frequency. The strongest resonator shift observed was ~ 10 MHz.

In a second measurement the resonance frequency of the resonator is tracked while sweeping the gate voltage. In this case the RF microwave generator is set to the particular resonance frequency f_0 at each gate voltage and simultaneously a second generator is fed onto the input line in order to generate a second tone (i.e. qubit tone). While the qubit tone is swept in frequency the response of the resonator at its resonance f_0 is measured. The colormap of this measurement on device v3_Q3 is presented in Figure 8.2 (b). Whilst there is no evidence of a resonator response for $V_G < -30$ V, at higher gate voltages a resonator response is evident. The gate voltages at which there is a resonator response coincide exactly with the gate voltages of the resonator shifts shown in Figure 8.2 (a). This proves that there is a qubit present, which can be tuned in frequency between $2.8 \text{ GHz} < f_Q < 4.2 \text{ GHz}$.

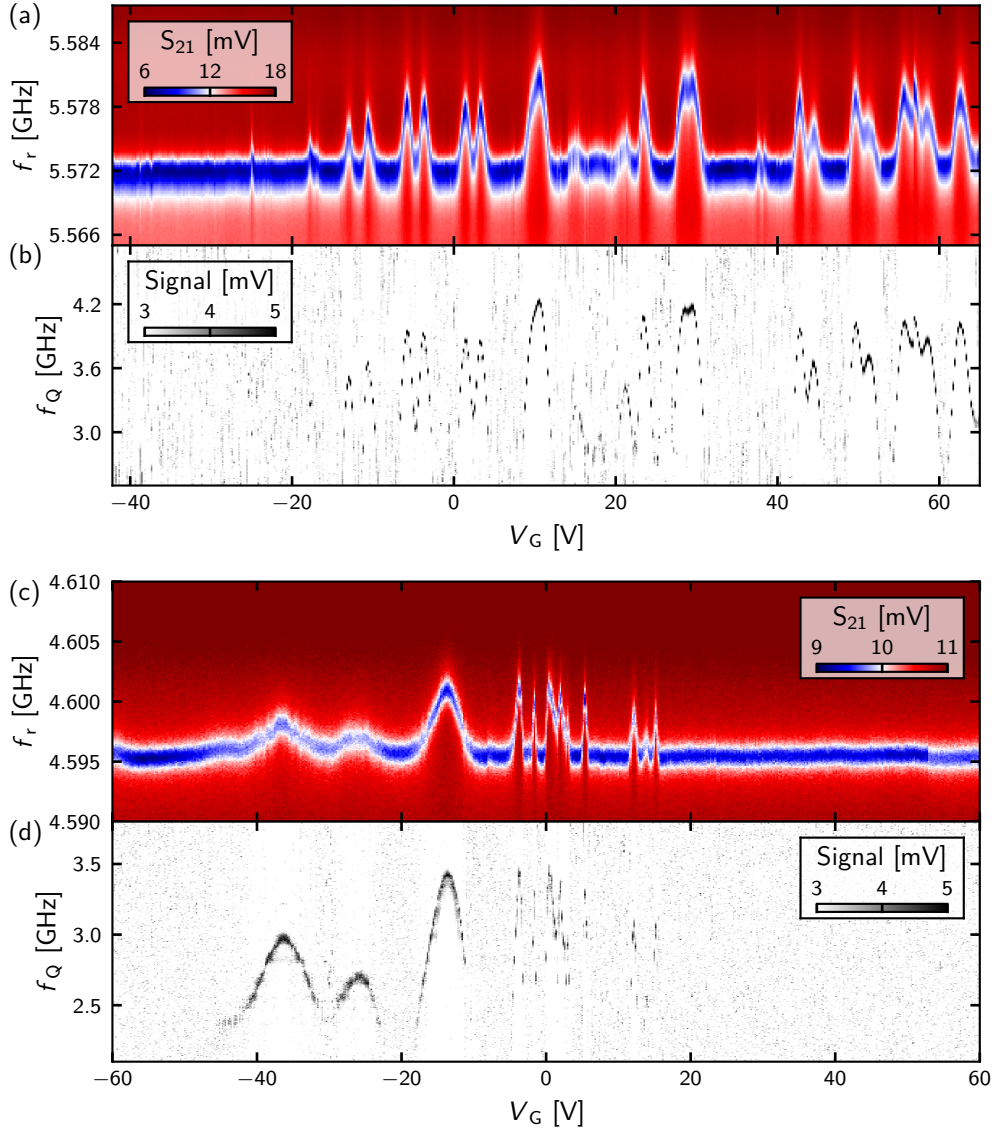


Figure 8.2: Resonator and qubit spectroscopy as function of applied gate voltage for devices v3_Q3 ((a) and (b)) and v2_Q1 ((c) and (d)). (a) Resonator spectroscopy of device v3_Q3 as a function of gate voltage with a 'bare' resonance frequency $f_0 = 5.572$ GHz. The resonator shifts upwards in frequency for distinctive gate voltages. (b) Qubit spectroscopy of device v3_Q3 as a function of gate voltage. A second tone, i.e. qubit tone, is applied to the resonator and its response at the resonance frequency for each gate voltage is measured. A distinctive response is observed at the same gate voltages for which the resonance of the resonator in (a) shifts upwards. (c) Resonator spectroscopy of device v2_Q1 as a function of gate voltage with a 'bare' resonance frequency $f_0 = 4.595$ GHz. Like in (a) the resonator shifts upwards in frequency for distinct gate voltages. (d) Qubit spectroscopy of device v2_Q1 as a function of gate voltage. An apparent resonator response is observed for gate voltages at which the resonator frequency is shifted in (c).

This qubit spectroscopy likely shows the f_{01} , i.e. ground state to first excited state transition of the superconducting qubit as a function of gate voltage.

Similar measurements as described above for device v3_Q3, were also performed on device v2_Q1 and presented in Figure 8.2 (c) and (d). The resonator of this device exhibits a 'bare' resonance frequency of $f_0 = 4.595$ GHz. The gate voltage dependence of f_r is flat for $V_G > 20$ V and exhibits upwards shifts when V_G is tuned below 20 V, see Figure 8.2 (c). The strongest resonance frequency shift was observed to be ~ 7 MHz. Further, the qubit spectroscopy presented in Figure 8.2 (d) shows no response for $V_G > 20$ V. However, for $V_G < 20$ V again a resonator response is visible at the same gate voltages as the resonance shifts of the resonator. Hence, in device v2_Q1 the ground to first excited state transition frequency can be tuned between $2.4 \text{ GHz} < f_Q < 3.5 \text{ GHz}$, cf. Table 8.1. A third qubit device was measured which exhibits similar behaviour, measurements can be found in Appendix C and the device behaviour is summarised in Table 8.1.

Since the qubits unfortunately cannot be tuned to cross the resonator, the coupling strength g between the microwave photons and the qubit has to be estimated using the dispersive shift χ of the resonator frequency. To first order χ , due to the coupling to the ground to first excited state transition of the qubit, is given by

$$\chi = \frac{g^2}{\Delta}, \quad (8.1)$$

where $\Delta = f_0 - f_Q$ is the detuning between the qubit and the resonator with $f_Q = f_{01}$. Using this expression and the data presented in Figure 8.2 (a) and (b) yields, averaged over all f_Q , $\langle g \rangle \cong 100$ MHz. For small f_Q the coupling strength is reduced and the minimum extracted $g = 76$ MHz. The opposite is true for higher f_Q , where a maximum of $g = 113$ MHz was found, cf. Table 8.1. The couplings observed in the other two devices are summarised in Table 8.1.

	f_r [GHz]	f_q [GHz]	$g_{\max}$ [MHz]	$\chi_{\max}$ [MHz]
v3_Q3	5.572	2.8 – 4.2	113	10
v2_Q1	4.595	2.4 – 3.5	88	7
v2_Q5	5.296	3.0 – 4.0	108	9

Table 8.1: Parameters extracted from resonator and qubit spectroscopy measurements for devices v3_Q3, v2_Q1 and v2_Q5.

8.3.2 Determining the Qubit Hamiltonian

The qubit spectroscopy, i.e. mapping out the $f_{01} = f_Q$ transition of the voltage tunable superconducting qubit was the first step in characterising the qubit. However, to fully describe the qubit Hamiltonian we would like to know E_J and E_C . A step towards further characterising the qubit is to measure the power-dependent qubit spectroscopy. For a power-dependence measurement the gate voltage is fixed and the resonator is measured only at its particular resonance frequency. The qubit tone is now swept over the f_{01} transition at increasing powers. Such a measurement is presented in Figure 8.3 (a). At low qubit drive powers, $P < -35$ dB, only a single peak is observed in the qubit spectroscopy, see Figure 8.3 (b) bottom trace. As mentioned before, this resembles the f_{01} transition of the qubit.

If the power of the qubit drive is increased to $P = -30$ dB, a more complicated multi-peak response is observed, which usually (i.e. for device v3_Q3) shows a relatively clear second peak at frequencies just lower than f_{01} , see Figure 8.3 (b) top trace. This is what is typically observed for transmon qubits and corresponds to the two photon transition from the ground state to the second excited state, i.e. $f_{02}/2$. However, this could also be a different transition due to the low E_J/E_C ratio and low anharmonicity of the qubit. Nevertheless, the certainty for it to be the $f_{02}/2$ transition is very high and we will assume this hereafter. The following analysis will give more insight as to why this assumption is likely to be correct. The additional broad transition at lower frequencies $f < f_{02}/2$, see Figure 8.3 (b) upper trace, could be another higher transition of the qubit energy spectrum due to the low E_J/E_C ratio and low anharmonicity.

At this point it is possible to extract f_{01} and $f_{02}/2$ at various gate voltages from data similar to Figure 8.3. These values are then used to calculate the anharmonicity between the first three energy levels via $\alpha = 2(f_{01} - f_{02}/2)$. By using a first order perturbation theory approach to a modified Cooper pair box Hamiltonian presented by *Kringhøj et al.* [114] the anharmonicity α and ground

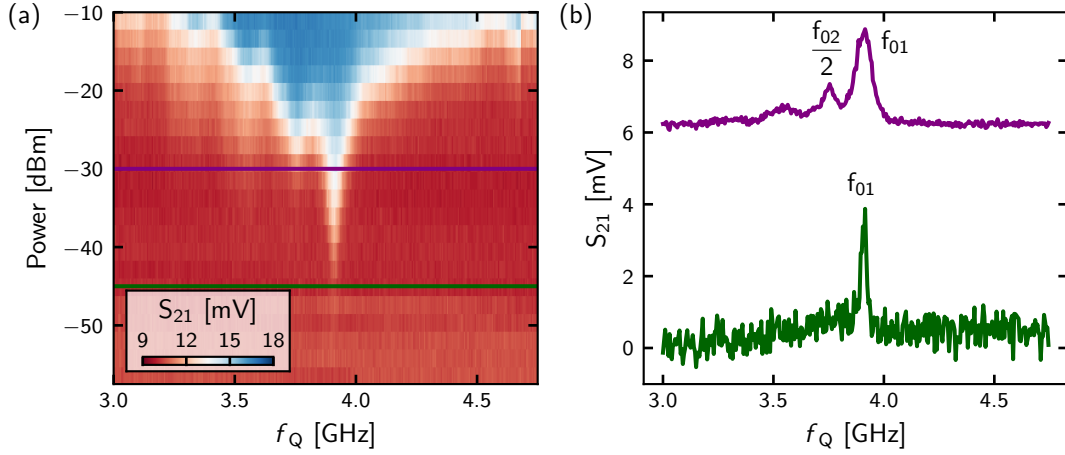


Figure 8.3: Qubit spectroscopy. (a) Amplitude response of the resonator as a function of frequency and power of the qubit drive tone. At low power only the f_{01} qubit transition is visible. At a drive power $P > -35$ dBm the two photon transition $f_{02}/2$ is also becoming visible. At significantly larger drive powers, the behaviour is more complex as the qubit has a low E_J/E_C ratio. (b) Single qubit spectroscopy traces along the cuts indicated in (a). The lower (green) trace, taken with $P = -45$ dBm, exhibits only the f_{01} transition of the qubit. The upper curve (purple, offset for clarity), taken with $P = -30$ dBm, exhibits a second sharp transition which is the $f_{02}/2$ transition of the qubit. The broader transition at lower frequency is likely an effect due to the low E_J/E_C ratio of the qubit.

to first excited state transition frequency f_{01} are given by

$$\alpha = E_C \left(1 - \frac{3E_J}{\Delta N} \right) \quad (8.2)$$

$$f_{01} = \sqrt{8E_J E_C} - E_C \left(1 - \frac{3E_J}{\Delta N} \right), \quad (8.3)$$

where $\Delta = 90 \mu\text{eV}$ is the induced superconducting gap of the nanotube Josephson junction (measured in Chapter 7) and N the number of channels contributing to the Cooper-pair transport, see Section 2.5.4 for details of the derivation. Using Equation 8.2 and Equation 8.3 together with the values extracted for f_{01} and α , the qubit parameters E_J and E_C can be calculated by solving the resulting set of equations. For the analysis presented here $N = 1$ was chosen, which is counterintuitive for carbon nanotubes known to exhibit four conduction channels. However, this was deduced to be the correct value according to the following analysis. If all channels are assumed to exhibit equal transmission $\mathcal{T}$, Equation 8.2 can be rearranged using $E_J = \frac{\Delta}{4} N \mathcal{T}$ such that $\mathcal{T} = \frac{4}{3}(1 - \alpha)$. The anharmonicity from the data can now be used to calculate $\mathcal{T}$ at each gate voltage (or at each f_{01}) resulting

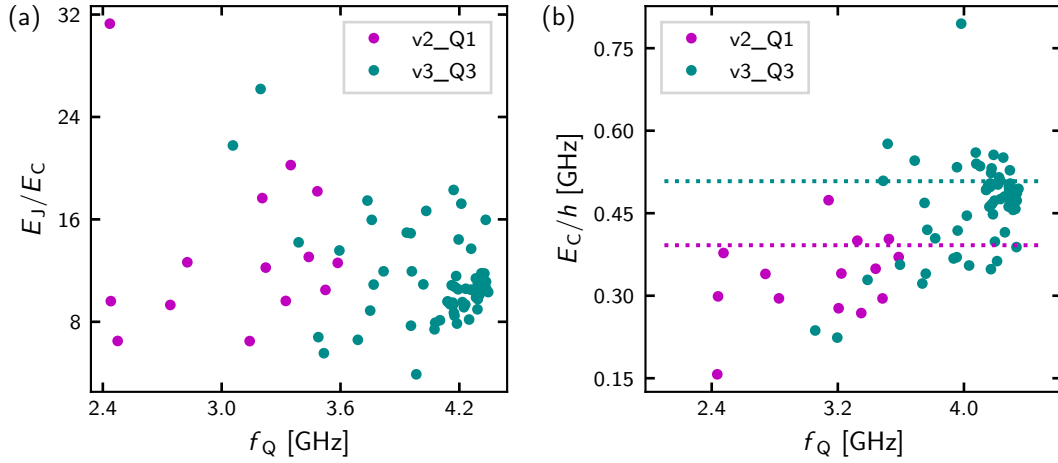


Figure 8.4: Qubit parameters E_J/E_C and E_C . (a) Extracted qubit E_J/E_C ratio as a function of f_Q for two different devices, i.e. v1_Q1 (purple points) and v3_Q3 (cyan points). (b) Extracted values for E_C as a function of f_Q for devices v1_Q1 (purple points) and v3_Q3 (cyan points). Dashed horizontal lines correspond to values of E_C determined via finite element simulations with *ANSYS Maxwell* for the exact device design.

in pairs of $[\mathcal{T}, f_{01}]$. Comparing these with $f_{01}(\mathcal{T}) = \sqrt{2\Delta E_C \mathcal{T} N} - E_C \left(1 - \frac{3}{4}\mathcal{T}\right)$ (resulting from rearranging Equation 8.3) for $N \in \{1, 2, 3, 4\}$ and E_C taken from electrostatic modelling, good agreement was only found for $N = 1$.

In Figure 8.4 (a) the E_J/E_C ratios extracted with the method mentioned above are presented as a function of qubit frequency $f_{01} = f_Q$. The extracted values accumulate around the mean $E_J/E_C \approx 11$ for v3_Q3 and $E_J/E_C \approx 13$ for v1_Q1. The spread in values is attributed to the error in extracting $f_{02}/2$ from the qubit power spectroscopy data as these can be noisy and sometimes exhibit a complicated multi-peak structure, making the peak distinction difficult. Additionally, the resolution in f_Q as well as in power had to be reduced due to finite measurement time. However, the qubit was designed with parameters yielding $E_J/E_C \sim 12$ in simulations, which restores confidence in the extracted mean values for E_J/E_C . In general, the average E_J/E_C ratio is rather low, i.e. $1 < E_J/E_C < 20$, which places the qubit just below the transmon regime ($E_J/E_C > 20$) but above a really "bad"¹ Cooper pair box regime ($E_J/E_C > 1$). Comparing $E_J/E_C \sim 12$ with the Cooper pair box energy level structure presented in Section 2.5.2 and Section 7.5, indicates

¹"Bad" in the sense that a low E_J/E_C ratio leads to a complex energy level structure with charge dispersion, which makes the qubit susceptible to charge noise.

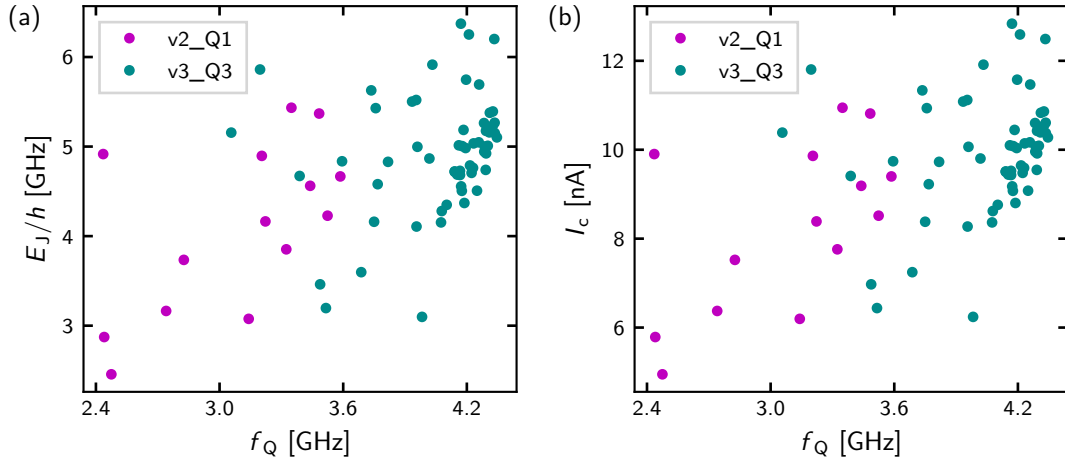


Figure 8.5: Parameters of the Josephson junction of the qubit. (a) Extracted values for E_J as a function of f_Q for devices v2_Q1 (purple points) and v3_Q3 (cyan points). As expected a linear trend is observable. (b) Critical current I_c of the qubit's Josephson junction, calculated from (a) following Equation 2.17. Values of I_c are comparable to values observed in Chapter 7.

that the first two energy levels should exhibit a vanishing charge dispersion. Only the third level is likely to display a slight charge dispersion. However, it is predicted that charge dispersion vanishes in channels where the transmission is approaching unity [215], which is the case for Josephson junctions made from carbon nanotubes. In this case the qubit presented here would exhibit almost no charge dispersion even though the E_J/E_C ratio is relatively low.

In Figure 8.4 (b) the extracted values for E_C are presented as a function of f_Q . The dashed horizontal lines correspond to values of E_C ($E_C^{v3-Q3} = 508$ MHz, $E_C^{v1-Q1} = 391$ MHz) calculated via finite element simulations using *ANSYS Maxwell* for the exact qubit design, see Section 4.3.1. For both qubit devices the extracted values are in close proximity to the simulated value. The spread in E_C is again attributed to the error in extracting $f_{02}/2$ as above. However, the charging energy could also be susceptible to excess charge build up in the substrate around the qubit device.

The second important qubit parameter is the Josephson energy E_J , which was also extracted with the method described above. Figure 8.5 (a) shows the extracted values of E_J as a function of f_Q . As expected E_J is increasing with increasing f_Q . Since v1_Q1 exhibits lower f_Q compared to v3_Q3 it also reaches lower E_J . The

spread in E_J is again attributed to the error of extracting $f_{02}/2$ from the power spectroscopy data. However, another reason for the distribution of extracted E_J values could also be phase diffusion in the Josephson junction, which was observed in DC measurements of similar Josephson junctions, see Section 7.4.2. Further, the Josephson energy can be used to calculate the critical current of the qubit's Josephson junction using Equation 2.17. The calculated values for I_c are presented in Figure 8.5 (b) and vary between $I_c = 5$ nA and $I_c = 13$ nA, with an average value $\langle I_c \rangle = 9.6 \pm 1.6$ nA across both qubit devices. These critical current values are comparable to values of I_c observed independently in DC measurements of carbon nanotube Josephson junctions, presented in Section 7.4. This proves the robustness and reproducibility of the fabrication of carbon nanotube Josephson junctions.

8.3.3 Qubit Lifetime and Coherence

Apart from determining the qubit Hamiltonian it is also necessary to characterise the qubit's lifetime and its coherence. There are multiple ways to perform this characterisation, however the most standard characterisation techniques are time resolved measurements using π and $\pi/2$ qubit pulses. For this Rabi oscillations are measured in order to determine the correct length for the π and $\pi/2$ pulses. However, in the qubits measured throughout this chapter different reasons prevented the visibility of Rabi oscillations. The resonator of qubit v2_Q1 exhibited Q_{int} lower than expected, which we attribute to catalyst residue of the carbon nanotube growth. Hence, $Q_{\text{int}} < Q_{\text{ext}}$ did not allow for Rabi oscillations to be visible. For the resonator of qubit v3_Q3 Q_{ext} was adjusted in the design, however a rather short T_1 of the qubit also did not allow for the visibility of Rabi oscillations. Hence it was necessary to make use of a different characterisation technique for each of these qubits. The techniques used to characterise T_1 and T_2 of the qubit are described below. Each of these measurements have been tested on a separate transmon qubit in a 3D-architecture in order to show that they yield the same result as standard techniques, see Appendix C.2.

8.3.3.1 Measuring T_1 with Chopped Microwave Pulses

In order to measure T_1 of the qubits, a pulse chopping method previously used for quantum dot charge qubits was borrowed and adjusted [216, 217]. In this method the resonator is continuously measured with low power at its resonance frequency f_r for a time of 100 μ s. Simultaneously the qubit drive tone is chopped up into a pulse train with a 50% duty cycle and for each measurement the pulse period τ is varied, see inset Figure 8.6 (a). For very short τ , i.e. $\tau \ll T_1$, the qubit drive excites the qubit into the first excited state and it has no time to relax. In the case of very long τ , i.e. $\tau \gg T_1$, the qubit gets excited and has time to relax. Therefore the measured signal is the time average of the qubit being in the ground and excited state, giving a signal of half the value found in the limit $\tau \rightarrow 0$. A single measurement following this procedure is presented in Figure 8.6 (a). Care was taken to filter out values of τ which are integer multiples of the downconversion intermediate frequency, $f_{IF} = 125$ MHz, as leakage through the IQ mixer would lead to false data points at these pulse periods. The data is normalised with respect to a measurement without the qubit drive tone turned on and fit with

$$S(\tau) = \frac{1}{2} + \frac{T_1(1 - e^{-\tau/(2T_1)})}{\tau} \quad (8.4)$$

where T_1 is the only free parameter. In this particular measurement shown in Figure 8.6 (a) the fit yields $T_1 = 117.3 \pm 5.8$ ns, which is one of the longer relaxation times measured in device v3_Q3.

The relaxation time T_1 was measured for a multitude of gate voltages, i.e. f_Q , for the two devices, see Figure 8.6 (b). Qubit v2_Q1 on average exhibits a longer $\langle T_{1,v2_Q1} \rangle = 151 \pm 71$ ns than qubit v3_Q3 where $\langle T_{1,v3_Q3} \rangle = 74 \pm 30$ ns, but qubit v2_Q1 is also lower in frequency which is in favour of a longer T_1 . The highest T_1 measured was $T_1 \sim 250$ ns in device v2_Q1 and $T_1 \sim 150$ ns in device v3_Q3. However, even for similar qubit frequencies qubit v2_Q1 exhibits a larger T_1 than qubit v3_Q3. This could be attributed to the carbon nanotubes being from two different growth batches, the carbon nanotube of qubit v3_Q3 being more disordered, a difference in contact resistance of the two devices or a change

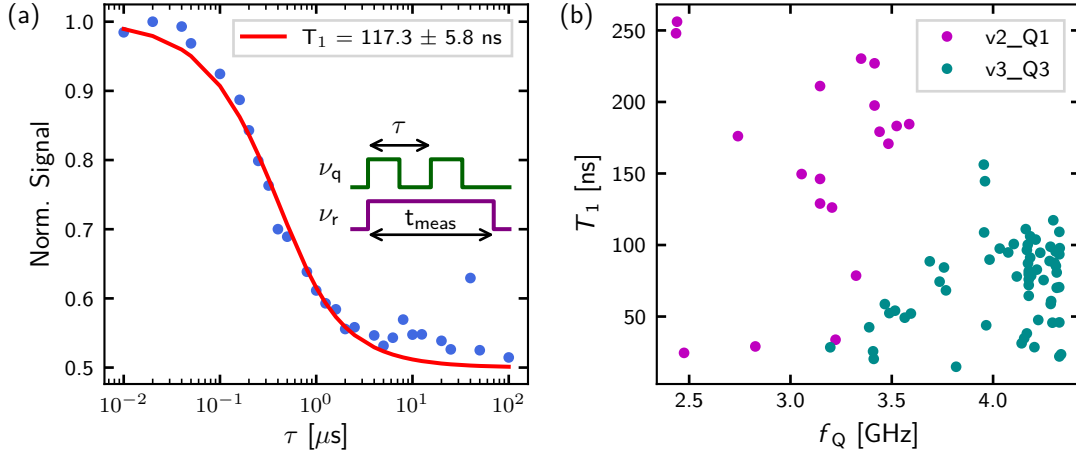


Figure 8.6: T_1 measurements. (a) A single T_1 experiment on device v3_Q3. Microwave pulses at the qubit frequency are chopped up into periods of length τ with a 50% duty cycle, whilst the resonator is continuously measured (see Inset for pulse sequence). The measurement response is normalised to the signal without a qubit drive tone and fit with Equation 8.4, yielding in this case $T_1 = 117.3 \pm 5.8$ ns. Inset: Pulse scheme (qubit pulse - green, measurement pulse - purple). (b) Measured T_1 as a function of qubit frequency f_Q for two devices v2_Q1 (purple) and v3_Q3 (cyan). The highest T_1 observed is $T_1 \sim 250$ ns in device v2_Q1.

made in the fabrication procedure for qubit v3_Q3. The change in fabrication made to device v3_Q3, was a secondary UV ozone clean after the last fabrication step (i.e. lift-off), which was envisioned to reduce the disorder on the carbon nanotube due to PMMA resist residue. The rather small relaxation times are attributed to dielectric induced charge noise.

8.3.3.2 Estimating T_2 from Qubit Spectroscopy

Since it was impossible to determine the parameters of a π -pulse for either of the qubit devices, it was not possible to measure the coherence time T_2 directly via Ramsey fringes. To our knowledge there is no other measurement that could be performed to directly test for the qubit's T_2 . However, an estimate, i.e. lower bound T'_2 , of T_2 can be found by measuring the linewidth of a low power qubit spectroscopy trace [14, 218], see Figure 8.7 (a). By fitting a Lorentzian with linewidth $2\delta\nu_{\text{HWHM}}$ to the qubit transition peak, T'_2 can be calculated via

$$2\pi\delta\nu_{\text{HWHM}} = \frac{1}{T'_2} = \left(\frac{1}{T_2'^2} + n_s \omega_{\text{vac}}^2 \frac{T_1}{T_2} \right)^{1/2} \quad (8.5)$$

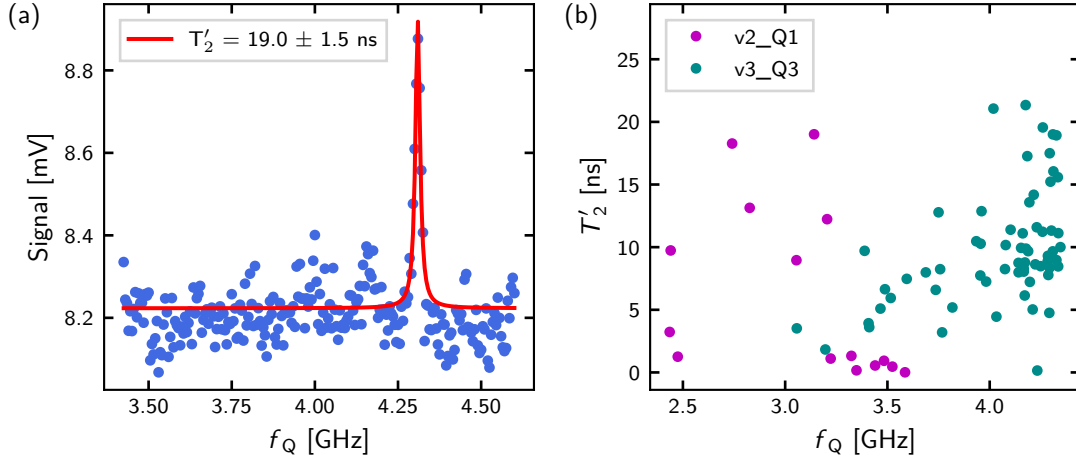


Figure 8.7: T'_2 measurements. (a) A single measurement of T'_2 on device v3_Q3. A low power qubit spectroscopy trace is fit with a Lorentzian (red curve) in order to estimate the linewidth and calculate T'_2 following Equation 8.5, yielding $T'_2 = 19.0 \pm 1.5$ ns. (b) Measured T'_2 as a function of qubit frequency f_Q for two devices v2_Q1 (purple) and v3_Q3 (cyan). The observed T'_2 s vary $0 < T'_2 \leq 25$ ns. In device v2_Q1 coherence is reduced at higher f_Q , whereas the opposite is true for device v3_Q3.

where $n_s \omega_{\text{vac}}^2$ is proportional to the microwave input power [14, 218]. Hence, at low qubit drive powers the linewidth should be the least broadened. This results in $T'_2 = 19.0 \pm 1.5$ ns for the data presented in Figure 8.7 (a).

The measurement and its analysis was repeated for different qubit frequencies f_Q , see Figure 8.7 (b). While T'_2 seems to be slightly increasing with increasing f_Q for qubit v3_Q3, this is not true for qubit v2_Q1. Coherence is highest at around $f_Q = 3$ GHz, with $T'_2 = 25$ ns for qubit v2_Q1, but significantly reduced at $f_Q = 3.5$ GHz. On average qubit v3_Q3 exhibits a longer $\langle T'_{2,v3_Q3} \rangle = 10 \pm 5$ ns compared to qubit v2_Q1 $\langle T'_{2,v2_Q1} \rangle = 6 \pm 6$ ns. Although these coherence times seem rather short, these only represent a lower bound for T_2 and potentially echoed T_2 measurements would reveal values much closer to the measured T_1 . However, the low coherence times could be attributed to dissipation due to dirty, disordered carbon nanotubes and residual resistance to the superconducting leads. Dissipation in nanoscale weak link Josephson junction oscillators, made from aluminium, was previously mentioned as a possible source of decoherence [219–221]. Short coherence times could also stem from the Andreev levels in the junction interacting with acoustic phonons [222, 223]. Additionally, the qubit linewidth could also be broadened due

to charge noise effecting the gate voltage applied to the carbon nanotube.

8.3.4 Signatures of Rabi Oscillations

Although it was not possible in the presented qubit devices to measure Rabi oscillations with separated qubit and measurement pulses, visibility of Rabi oscillations has been previously reported with weak continuous measurements while applying the qubit drive tone [224, 225]. A similar type of measurement was performed and some potential evidence of Rabi oscillations observed.

The measurements presented in the following paragraphs further investigated the time-resolved response of the measurement resonator, see Figure 8.8. Specifically, the difference between two cases was considered. One where a measurement pulse is sent to the resonator without a qubit drive pulse (Figure 8.8 (a)) and another one where during the pulse sent to the resonator a qubit pulse is switched on for 500 ns (Figure 8.8 (b)). In the second case it was possible to vary the amplitude V_{amp} of the qubit pulse. In both cases, the power of the measurement pulse was kept as low as possible in order to prevent the resonator pulse from interfering with the qubit. The measured time responses of the resonator for both pulse schemes, offset by 30 mV for clarity, are presented in Figure 8.8 (c). The top curve corresponds to the pulse scheme with no qubit drive pulse, i.e. $V_{\text{amp}} = 0$ V. Here a measurement tone is turned on for 2 μs and the turn on of the resonator is initially observed until it relaxes in its steady state population. When the pulse is turned off after $t = 2 \mu\text{s}$ the photon population of the resonator decays with its decay rate κ . The bottom curve of Figure 8.8 (c) corresponds to the pulse scheme with a 500 ns long qubit drive tone applied during the measurement pulse. The response initially exhibits identical behaviour compared to the resonator response without a qubit drive. However, when the qubit drive is turned on at $t = 0.5 \mu\text{s}$ the resonator exhibits a stronger magnitude response as the qubit is excited. The response due to the qubit drive pulse seems to display a period of an oscillation in the beginning until it reaches the steady state. When the qubit pulse is turned off, the resonator response due to the qubit pulse decays on a timescale comparable to T_1 . Eventually,

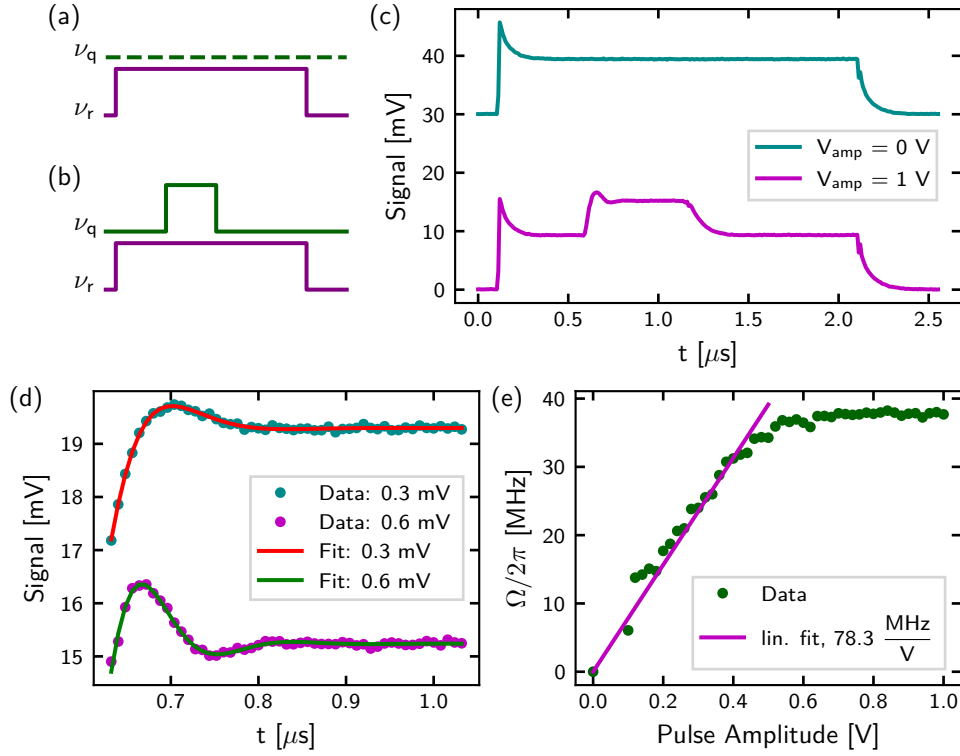


Figure 8.8: Signatures of Rabi oscillations in device v3_Q3. (a) Pulse scheme with a $2 \mu\text{s}$ long resonator pulse (purple) and no qubit drive pulse. (b) Pulse scheme with a $2 \mu\text{s}$ long resonator pulse (purple) and a 500 ns long qubit drive pulse in the middle of the measurement pulse. (c) Time-domain resonator response for pulse scheme in (a) top curve (cyan, offset by 30 mV due to clarity) and pulse scheme in (b) bottom curve (purple). (d) Zoom-in of the response of the resonator due to the qubit pulse for two different qubit drive amplitudes (offset due to clarity). The two responses exhibit an oscillation of different frequency, which is fit to an exponentially decaying sine function (solid lines). (e) Oscillation frequency $\Omega/2\pi$ as a function of the qubit pulse amplitude. For a pulse amplitude $< 0.6 \text{ V}$, $\Omega/2\pi$ is fit with a linear function (solid purple line) yielding a slope of 78.3 MHz/V .

once the resonator pulse is turned off after $t = 2 \mu\text{s}$ the resonator population again decays at a rate κ as for the case without a qubit pulse.

The amplitude of the qubit drive pulse can be varied from $0 \text{ V} < V_{\text{amp}} < 1 \text{ V}$. Further examining the resonator response due to the qubit pulse, it is obvious that the initial oscillation is only present for the first $150\text{-}200 \text{ ns}$ after the qubit drive pulse is turned on, see Figure 8.8 (d). Additionally, varying the qubit pulse amplitude results in a modulation of the oscillation frequencies of this initial response to the qubit drive pulse. Figure 8.8 (d) shows a close up of the response due to the qubit drive pulse for two different amplitudes (offset for clarity). The top curve, i.e. for

a lower qubit drive pulse amplitude, exhibits a slower oscillation compared to the bottom curve for double the qubit pulse amplitude. Both of these measurements are fit with an exponentially decaying sine curve, each yielding a frequency $\Omega/2\pi$ of the respective oscillation. This analysis can be done for a sweep of the qubit drive pulse amplitude V_{amp} . The extracted $\Omega/2\pi$ is plotted against the qubit pulse amplitude in Figure 8.8 (e). For pulse amplitudes < 0.6 V, $\Omega/2\pi$ linearly increases with a slope of ~ 78.3 MHz/V, determined from a linear fit (solid purple line). This linear dependence on qubit drive amplitude is a characteristic feature of Rabi oscillations. For pulse amplitudes > 0.6 V, $\Omega/2\pi$ saturates at just below 40 MHz which may be explained as being due to the low anharmonicity of the qubit (the qubit will be driven into higher energy states at drive rates greater or equal to the anharmonicity).

8.4 Summary/Conclusion

The experiments described in this Chapter demonstrate a first implementation of a voltage tunable superconducting qubit based on a carbon nanotube Josephson junction. Similar qubits have been previously implemented with Josephson junctions based on InAs nanowires [50, 51], two-dimensional electron gases [214] and graphene [107]. The qubit presented here is of comparable geometry and exhibits similar gate voltage behaviour. In the beginning of the Chapter the geometry and fabrication of the device were discussed. As a first measurement the resonator and simultaneous qubit spectroscopy was presented, which showed clear evidence of microwave photon to qubit coupling with an average coupling strength of $g = 100$ MHz. The coupling strength is comparable to state of the art circuit QED experiments with any type of transmon qubit. The qubit frequency was tunable between $2.8 \text{ GHz} < f_Q < 4.2 \text{ GHz}$, which is on the lower end of the frequency spectrum in common implementations of circuit QED. Usually these use the frequency spectrum between 4 GHz and 8 GHz, as most of the microwave components are readily available and well understood at these frequencies.

In a second experiment the qubit spectroscopy was observed as a function of qubit drive power for a multitude of gate voltages, i.e. qubit frequencies. This was

used in order to extract the f_{01} and the $f_{02}/2$ transition frequency of the qubit. With these values the anharmonicity was calculated and a first order perturbation theory approach to a modified Cooper pair box hamiltonian was used to extract the important qubit parameters E_J , E_C , their ratio E_J/E_C and I_c of the Josephson junction. The perturbation approach used and first introduced in Reference [114] leads to reasonable results which agree quite well with simulations of the actual qubit device and previous independent DC measurements of carbon nanotube Josephson junctions. However, *Whisler et al.* [226] mention that a more accurate modelling of proximitised Josephson junction qubits, i.e. the Josephson non-linearity, is necessary as their theoretical results deviate from the model used by *Kringhøj et al.* [114]. Nevertheless, the current analysis is still assumed to be valid as a supposedly more accurate model is still outstanding research and the results agree well with simulations and previously measured devices. The extracted values for E_J/E_C place the qubit in an intermediate regime, which is below the transmon regime but away from a bad Cooper pair box regime. Charge dispersion should not affect the first two energy levels and will only have a minor effect on the third level. Interestingly, *Averin* [215] predicted that charge dispersion vanishes for channels for which the transmission approaches unity. This could have a beneficial effect in the qubit presented here.

Further to determining the essential qubit parameters, the lifetime T_1 and coherence T_2 of the qubit were characterised. The T_1 measured in the two proof of concept devices in this chapter is very short compared to other already more advanced gatemon-type qubits. However, the first implementation of such devices with InAs nanowires also suffered from a low $T_1 = 500 - 800$ ns [51]. Advances in the fabrication of InAs nanowire qubit devices lead to a drastic improvement resulting in $T_1 = 5 - 20$ μ s [227, 228], which is similar to state of the art flux-tunable transmon qubits based on Al/Al₂O₃ Josephson junctions [79]. New realisations of gatemon type qubits based on proximitised two-dimensional electron gases show $T_1 \sim 2$ μ s [214]. Like for the measured T_1 , similarly T_2 measured in other gatemon type qubits is already more advanced. However, what seems promising is that

almost all of the measured T_2 are T_1 limited, i.e. an improvement in T_1 will also most likely improve T_2 . For the InAs nanowire qubits initially $T_{2,\text{echo}} \sim 900 \mu\text{s}$ was reported [51], which was then improved an order of magnitude [227]. For the proximitised two-dimensional electron gas qubit $T_{2,\text{echo}} = 2.2 \mu\text{s}$ was reported [214]. Very recently another voltage tunable superconducting qubit was realised with a Josephson junction based on graphene encapsulated in hexagonal Boron Nitride (hBN), exhibiting a very broad transition linewidth of 350 – 425 MHz [107]. Due to the broad linewidth no further measurements of relaxation and coherence times as well as Rabi oscillations were reported for the graphene-based superconducting qubit.

Due to the low relaxation time of the qubit it was not possible to observe standard time resolved Rabi oscillations. However, signatures of Rabi oscillations were observed in a weak continuous measurement while applying a qubit drive tone. The observed oscillation in the resonator response while the qubit is driven, has a linear dependence on the drive amplitude and doubles if the amplitude is doubled. This is a strong indication that the observed oscillations are indeed Rabi oscillations.

For future work it would be desirable to increase E_J such that the qubit can be operated in the transmon regime. This will be achieved by increasing the critical current of the carbon nanotube Josephson junction which is thought to be realised by different aspects of the fabrication of the devices. At first a recalibration of the Pd/Al bilayer could lead to a larger induced superconducting gap and in turn a higher critical current. Reducing the channel length of the carbon nanotube between the superconducting leads would favour a more ballistic transport and probably also increase I_c . In general cleaner nanotubes in the device will greatly benefit the critical current. One could either think about a stamping scheme, where the nanotube is placed across the leads as the last step of the fabrication and hence preventing the nanotube from getting "dirty" during all the lithography and imaging steps. Similarly, as a last fabrication step the carbon nanotubes could be grown directly on the chip. However, this limits the choices of superconductors and other metals used in the device fabrication as these need to withstand the high temperatures of the nanotube growth. A possible easier approach to a clean carbon nanotube would

be to encapsulate the carbon nanotube in hexagonal Boron Nitride (hBN) as it is already quite common in graphene device fabrication. The improved cleanliness and hence increased E_J such that the transmon regime is reachable would most likely also benefit the coherence and relaxation time of a qubit based on a carbon nanotube. Encapsulating the carbon nanotube in hBN might reduce limitations in the coherence due to the substrate but could also add a great deal of problems.

9

Conclusion and Prospects

In this thesis, experimental work on different types of hybrid quantum devices employing superconducting circuits, spin ensembles and carbon nanotubes was presented, including the successful realisation of a carbon-nanotube-based superconducting qubit.

The first experiments presented in Chapter 6 studied the interaction of microwave photons with an organic spin ensemble in an uncorrelated and correlated state. Microwave transmission spectroscopy with the magnetic field tuned and detuned from the magnetic resonance of the spin ensemble, allowed for the extraction of all the energy decay rates as well as the coupling rate of the system. This showed that strong coupling was achieved to the correlated state of the spin ensemble and high cooperativity coupling to the uncorrelated state. The coupling strength as well as the magnetic resonance field was further investigated as a function of temperature and external magnetic field direction. The analysis of the field direction dependent magnetic resonance illustrated that the anisotropy observed in the temperature dependence of the magnetic resonance is explained by the onset of antiferromagnetic correlations in the spin ensemble. The anisotropy in the temperature dependence of the magnetic resonance was further investigated with a one-dimensional model of a uniaxial anisotropic antiferromagnet. This resulted in a prediction of the anisotropic

exchange energy present in the spin ensemble, which was in good agreement with the dipole-dipole interaction estimated from the crystal structure of the ensemble.

Hybrid quantum circuits based on spin ensembles have potential use in quantum information processing as quantum memories [34, 35] as well as for microwave-to-optical conversion [177]. Antiferromagnetic ensembles exhibit many benefits such as higher spin density and no net magnetisation as opposed to paramagnetic and ferrimagnetic ensembles respectively. Additionally, heterostructures made from antiferromagnetic materials could potentially combine spintronic and magnonic functionalities [181]. In order to use these materials for such complex purposes it is essential to understand the range of interactions governing antiferromagnetic systems. Organic magnets can be engineered as model systems with clearly defined magnetic interactions [182, 183], which can be probed in a hybrid circuit QED architecture in the interest of testing magnetic models of different dimensions [184].

In Chapter 7 the transport characteristics of carbon nanotubes embedded in circuits made from normal metals and superconductors were investigated. In both type of devices clear signs of Coulomb blockade could be observed. The carbon-nanotube-superconductor hybrids exhibit a superconducting gap around zero-bias voltage as predicted by the superconducting proximity effect. Probing these devices in a current biased measurement showed that they effectively behave like SNS Josephson junctions, where the critical current and in turn the Josephson energy can be modulated with an externally applied gate voltage. Additionally, multiple Andreev reflections were observed. The same behaviour of a carbon nanotube Josephson junction was discovered when measured with RF reflectometry. In light of the Josephson junction behaviour of the carbon nanotube devices with superconducting leads, the feasibility of realising a superconducting qubit was discussed at the end of the chapter. Such a qubit has been realised in Chapter 8.

Aside from the application as a Josephson junction in a superconducting qubit, these types of voltage tunable Josephson junctions can also be used as a switch to turn the coupling between a qubit and a microwave resonator on and off [229]. In a more elaborate approach, the Andreev Bound states present in the carbon

nanotube Josephson junction and responsible for the supercurrent transport can be used as a new type of qubit implementation, where the qubit state is encoded in two Andreev levels of the junction [223, 230]. Carbon nanotubes electronically accessed with normal metal leads can be used to define double quantum dots and implement a spin-valley qubit [231], which due to the strong spin orbit coupling [232] could be interfaced with superconducting microwave resonators, much along the lines of similar experiments with other quantum dot devices [39–49].

Finally, the results on carbon nanotube Josephson junctions were used in order to realise a carbon-nanotube-based superconducting qubit, which is presented in Chapter 8. Qubit spectroscopy measurements were performed as a function of gate voltage applied to the carbon nanotube Josephson junction, resulting in a modulation of qubit frequency ranging from qubit transitions completely switched off to qubit transitions with frequencies between 2.8 – 4.2 GHz. High-power qubit spectroscopy in combination with a model taking the different junction characteristics into account allowed the extraction of the qubit parameters E_J and E_C . The extracted values of E_J and E_C compare very well with values from measurements in Chapter 7 and finite element simulations respectively. Further, the relaxation and coherence time of the carbon nanotube qubit was analysed as a function of qubit frequency, resulting in an average $T_1 \approx 100$ ns and $T_2' \approx 10$ ns across all qubit frequencies. Weak continuous time resolved measurements showed potential signs of Rabi oscillations in the resonator response to a qubit drive of varied amplitude.

Voltage tunable superconducting qubits have also been realised with Josephson junctions made from InAs nanowires [50, 51], InAs/InGaAs heterostructures [214] as well as graphene [107, 233]. The carbon-nanotube-based superconducting qubit realised in Chapter 8 adds to the list of potential candidates for this hybrid architecture commonly referred to as gatemon qubits. Future research directions of gatemon qubits can lead towards new coupling schemes for qubits or even qubit architectures [234]. Proof of concept of tunable coupling to microwave resonators mediated by a voltage tunable Josephson junction has already been presented [229]. Mainly chased in InAs and InSb nanowires [235] contacted with superconductors

but also predicted in carbon nanotubes [236–238] are Majorana fermions which could be used for topological quantum computing [239]. Proposals have been made to interface a topological qubit with regular superconducting qubits [240–243], where gatemon type qubits could become beneficial. Depending on the type of superconductor used for implementing a gatemon it is possible to operate these in significant magnetic fields [107, 228], which would allow for possible interesting experiments with spin ensembles, e.g. used as quantum memories.

Modified versions of the different hybrid superconducting circuits presented in this thesis have potential to be combined to an extensible architecture for quantum information processing in the future. This would make it possible to harness each building block for the task it performs best at. Additionally, hybrid quantum devices can lead to further insight and understanding of the fundamental principles of quantum mechanics governing the various systems interacting with each other.

Part IV

Appendices



Appendix: Fabrication Recipes

A.1 NbTiN Resonator Fabrication

The NbTiN coplanar waveguide resonators used in the experiments presented in Chapter 6 were fabricated using the following recipe.

Process Step	Description
Chip preparation	Quartz chip pre-sputtered with NbTiN film Acetone ultrasonic bath for 5 min, 50°C (2x) Rinse in acetone and IPA N ₂ blow dry chip carefully Prebake on hotplate for 2 min at 120°C Cooldown for 5 min
Spin Coating	Cover entire chip in S1813 Spin for 2 s at 500 rpm Spin for 40 s at 7000 rpm Bake on hotplate for 90 s at 115°C
Exposure and Development	Expose in maskaligner with 45 mJ/cm ² Develop in MF-319 for 90 s Stop development in DI water for 60 s Hardbake on hotplate for 30 min at 125°C
Etching	Dry etch in Oxford Instruments Plasmalab 80 SF6 @ 13.5 sccm, O2 @ 20.0 sccm, 200 W, 21 mTorr for 4 min 30 s
Chip Cleaning	Strip resist in Remover 1165 for 60 min at 80°C Rinse in acetone and IPA N ₂ blow dry chip

A.2 Al Resonator Fabrication

The microwave resonator chips used in the experiments presented in Chapter 8 were either fabricated with following recipe or with ebeam lithography, see next section.

Process Step	Description
Chip Preparation	Dice Si wafer into 10x10 mm chips Acetone ultrasonic bath for 10 min at 50°C (2x) Rinse in acetone and IPA N ₂ blow dry Clean in piranha etch (3:1, sulfuric acid:hydrogen peroxide) Dilute and wash in DI water Rinse in acetone and IPA N ₂ blow dry
CNT Growth	for qubit devices CNT growth, for only resonators skip this step
Spin Coating	Cover entire chip in AZ 5214E Spin at 500 rpm for 10 s Spin at 3000 rpm for 30 s Bake on hotplate for 50 s at 110°C
Exposure and Development	Expose in maskaligner with V+H contact, with 40 mJ/cm ² Bake on hotplate for 120 s at 120°C Flood expose in maskaligner (no mask) with 200 mJ/cm ² Develop in AZ 726 MIF for 90 s Stop development in DI water for 60 s N ₂ blow Dry Oxygen plasma etch for 60 s, 50 W at 0.5 mbar
Evaporation and Lift-Off	Evaporate 100 nm of Al at 0.5 nm/s Lift-Off in acetone bath for 60 min at 50°C Rinse in acetone and IPA N ₂ blow dry

A.3 E-Beam Lithography and Carbon Nanotube Contacting

For large features, such as bondpads, alignment markers and microwave resonators, down to a feature size of roughly ~ 500 nm, the following recipe was used. If the chip has already been processed before, i.e. metal evaporated or carbon nanotubes

grown, the entirety of the chip preparation step is skipped.

Process Step	Description
Chip Preparation	Dice Si wafer into 10x10 mm chips Acetone ultrasonic bath for 10 min at 50°C (2x) Rinse in acetone and IPA N ₂ blow dry Clean in piranha etch (3:1, sulfuric acid:hydrogen peroxide) Dilute and wash in DI water Rinse in acetone and IPA N ₂ blow dry
Spin Coating	Cover entire chip in PMMA 495K A6 Spin at 500 rpm for 10 s Spin at 6000 rpm for 50 s Bake on hotplate for 5 min at 175°C Cover entire chip in PMMA 950K A3 Spin at 500 rpm for 10 s Spin at 6000 rpm for 50 s Bake on hotplate for 5min at 175°C
Exposure and Development	Expose in ebeam writer with 757 $\mu\text{C}/\text{cm}^2$, Aperture 3 and beam current 10 nA Develop in 3:1 IPA:MIBK for 45 s Stop development in IPA for 30 s N ₂ blow Dry Oxygen plasma etch for 60 s, 50 W at 0.5 mbar
Evaporation and Lift-Off	Evaporate 100 nm of Al at 0.5 nm/s Lift-Off in acetone bath for 60 min at 50°C Rinse in acetone and IPA N ₂ blow dry

For features smaller than 500 nm the following recipe was used and a pitch of roughly ~ 100 nm could be achieved. This recipe is mainly used for contacting the carbon nanotubes, especially when short channel lengths are required.

Process Step	Description
Spin Coating	Cover entire chip in PMMA 950K A6 Spin at 500 rpm for 10 s Spin at 6000 rpm for 50 s Bake on hotplate for 10 min at 175°C
Exposure and Development	Expose in ebeam writer with 757 $\mu\text{C}/\text{cm}^2$, Aperture 1 and beam current 100 pA Develop in 3:1 IPA:MIBK for 45 s Stop development in IPA for 30 s

	N ₂ blow Dry UV Ozone clean for 60 s
Evaporation and Lift-Off	Evaporate 4 nm of Pd at 0.25 nm/s Evaporate 80 nm of Al at 0.5 nm/s (For only Pd contacts: Evaporate 50 nm of Pd at 0.25 nm/s) Lift-Off in acetone bath for 60 min at 50°C Rinse in acetone and IPA N ₂ blow dry

B

Appendix: Strong Coupling of Microwave Photons to Antiferromagnetic Fluctuations in an Organic Magnet

B.1 Measurements of Crystal II

The measurements of Figure 6.2, Figure 6.3 and Figure 6.6 in Chapter 6 were rerun with a second sample (Crystal II). The measurements and the extracted parameters are similar to the results presented in Chapter 6.

Figure B.1 shows measurements and fits similar to Figure 6.2 in Chapter 6. Extracted parameters are given in Table 6.1. Figure B.2 shows measurements and fits similar to Figure 6.6 and the extracted parameters are mentioned in Chapter 6.

B.2 Number of Coupled Radicals in the Crystal

The Crystal I was measured to have a length of $365 \mu\text{m}$. The effective coupled width of the crystal is assumed to be $w + 2s = 60.6 \mu\text{m}$. Additionally, a penetration depth of the vacuum magnetic field of $4 \mu\text{m}$ is assumed [244]. This results in a coupled crystal volume of $V = 1 \times 10^{-7} \text{ cm}^3$. Taking the density of molecules from the diffraction data (Figure B.3) as $\rho_V \sim 2 \times 10^{21} \text{ cm}^{-3}$, then the number of coupled radicals in Crystal I is $N = \rho_V V \sim 1.7 \times 10^{14}$.

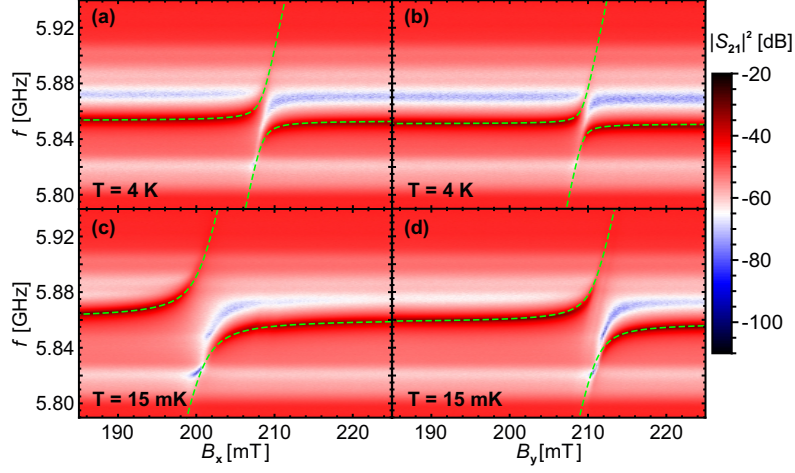


Figure B.1: Transmission as a function of external magnetic field $B_{x,y}$ and resonator probe frequency f , measured at two different temperatures using Crystal II. Superimposed on each panel are fits (dashed lines) using Equation 6.1 in Chapter 6.

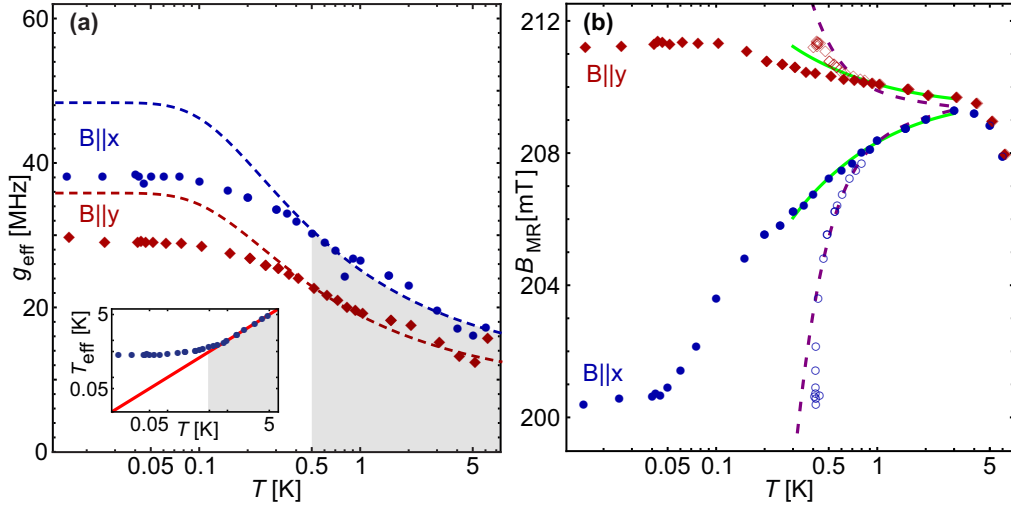


Figure B.2: Temperature dependence of Crystal II. (a) Temperature evolution of g_{eff} for $\mathbf{B}$ applied along x and y . Above $T \sim 500$ mK, the data agree with a PM model [dashed lines, fit to Equation 6.5 over the shaded temperature range]. Inset left: Effective spin-temperature calculated with Equation 6.5 (points). (b) Filled symbols: Resonance magnetic field along x and y as a function of temperature. As temperature decreases, the resonance magnetic field moves away from its paramagnetic value. At intermediate temperatures, both branches are fit by a spin chain model (solid green curves). Unfilled symbols: B_{MR} along x and y as a function of effective temperature according to Equation 6.5. The data for $T \leq 3$ K is fit by a spin chain model (dashed curve).

B.3 Growth and Structure of the Crystal

All reagents including 2,2-Diphenyl-1-picrylhydrazyl (DPPH; CAS: 1898-66-4), n-hexane, (99+%), and toluene, (99+%), were purchased from Sigma Aldrich or Fisher Scientific and used without further purification. Low temperature X-ray diffraction data were collected from Crystal II using an Oxford Diffraction (Rigaku) SuperNova diffractometer. Data were reduced using CrysAlisPro and solved using SIR92 [245] within the CRYSTALS suite [246, 247]. The structure was refined using full-matrix least-squares with CRYSTALS to give the unit cell shown in Fig. B.3. Disordered solvent in each unit cell was modelled as one toluene molecule. In general, hydrogen atoms were visible in the difference map and refined separately using soft restraints prior to inclusion in the final refinement using a riding model [248]. There was no evidence of protonation of the central nitrogen atom in each molecule (N2 in Fig. B.3). Comparison of the final refinement with that generated using PLATON/SQUEEZE [249] suggested that the disordered toluene model was a good fit to the electron density. Full refinement details are given in the Supporting Information (CIF file). Crystallographic data (excluding structure factors) have been deposited with the Cambridge Crystallographic Data Centre (CCDC 1475359) and will be available via http://www.ccdc.cam.ac.uk/data_request/cif.

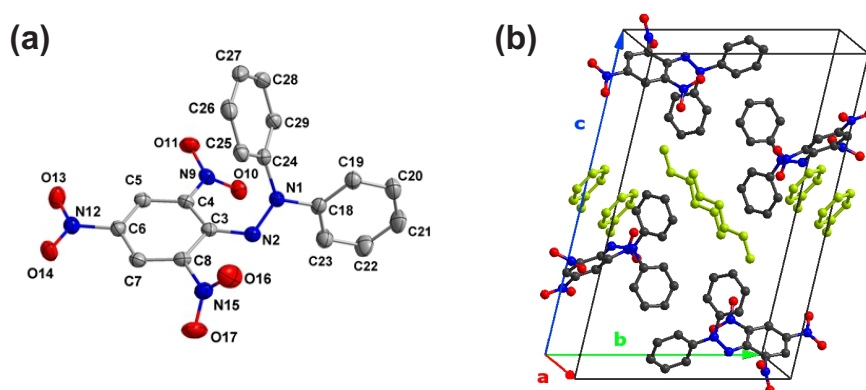


Figure B.3: (a) Molecular structure of DPPH with displacement ellipsoids drawn at 50% probability. Color code: carbon (grey), oxygen (red), nitrogen (blue), hydrogen not shown. (b) Unit cell of the measured crystals containing four DPPH molecules, one hexane, and one toluene molecule per cell, with the latter two shown in green. The toluene exhibits crystallographic disorder (not shown). Lattice vectors **a**, **b**, **c** are marked. Lattice constants are $|a| = 7.4 \text{ \AA}$, $|b| = 13.5 \text{ \AA}$, $|c| = 21.7 \text{ \AA}$.

B.4 Bulk Susceptibility Measurements

B.4.1 High Temperature Measurements

Magnetisation $M(T)$ was measured as a function of temperature (and converted to susceptibility by the relation $\chi(T) = M(T)/H$) using a Quantum Design MPMS 7 T SQUID. Polycrystalline samples of DPPH were loaded into gelatine capsules, mounted in a plastic drinking straw, and affixed to the end of a stainless steel/brass rod. The sample was cooled from 300 K to 1.8 K. Results are shown in Fig. B.4.

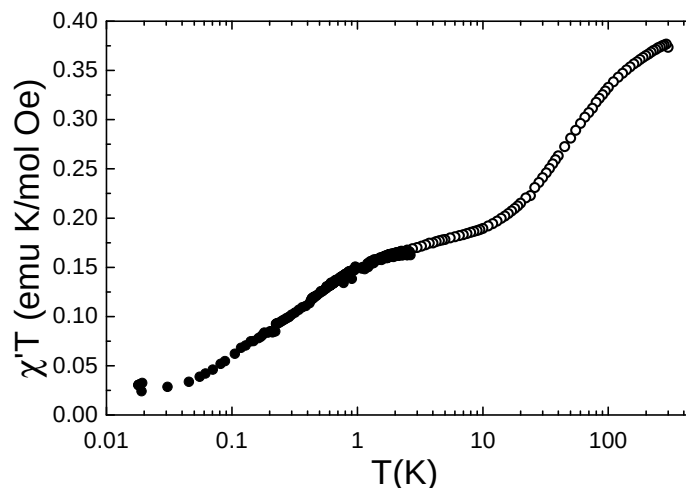


Figure B.4: Magnetic susceptibility of a polycrystalline sample of DPPH. Data is plotted as the χT product, which was corrected for the diamagnetic background. High- and low-temperature measurements are marked with open and filled circles respectively.

Near room temperature the product χT is constant at a value 0.37 emu/K mol Oe, as expected for a paramagnet with $S = 1/2$ and $g = 2.0$ [250, Eq. (A.8)]. Below ~ 100 K, the susceptibility decreases, consistent with antiferromagnetic spin interactions. The additional structure in the data observed on cooling may reflect different values of the intermolecular exchange along different crystallographic directions, not included in the chain model in the main text.

B.4.2 Low Temperature Measurements

To measure susceptibility below 1.8 K, we used a 30 μm micro-SQUID susceptibility meter [251]. Polycrystalline samples of DPPH were attached with Apiezon N

grease to a SQUID loop. The sample was cooled from 3 K to 16 mK in a dilution refrigerator and an AC magnetic field of amplitude 0.25 Oe was applied with its frequency varied from 0.3 Hz to 200 kHz.

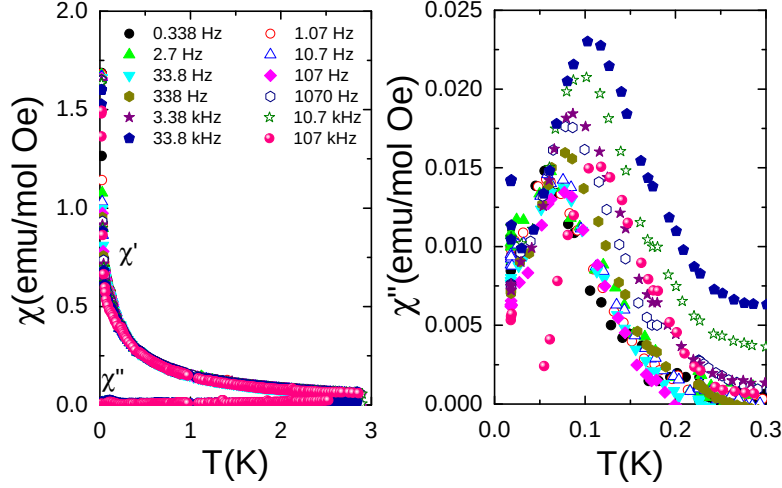


Figure B.5: Magnetic susceptibility of a polycrystalline sample of DPPH for $T \leq 3$ K, measured with twelve different ac-magnetic field frequencies. Data is shown for the in-phase, χ' , and out-of-phase, χ'' , component.

Figure B.5 shows representative results of the in-phase, χ' , and out-of-phase, χ'' , component of the susceptibility χ . Below ~ 1.7 K the product $\chi'T$ is decreasing, which is consistent with antiferromagnetic interactions. However, no evidence of a phase transition (in the form of a peak in $\chi'(T)$) could be detected, indicating that any transition from PM to AFM occurs below 16 mK. From the peak in $\chi''(T)$ it is evident that there is an onset of slow relaxation below 150 mK.

B.5 Muon Spin Relaxation Measurements

To further exclude long-range magnetic ordering we carried out muon-spin relaxation measurements down to $T = 19$ mK. Measurements were performed at the Swiss Muon Source, Paul Scherrer Institute using the LTF instrument. Roughly 100 mg of polycrystalline DPPH was attached with vacuum grease to a thin silver foil, which in turn was attached to a silver plate mounted on the cold finger of the dilution refrigerator. In a μ^+ SR experiment spin polarised muons are implanted into the

sample. The measured quantity is the asymmetry $A(t)$, which is proportional to the spin polarisation of the muon ensemble.

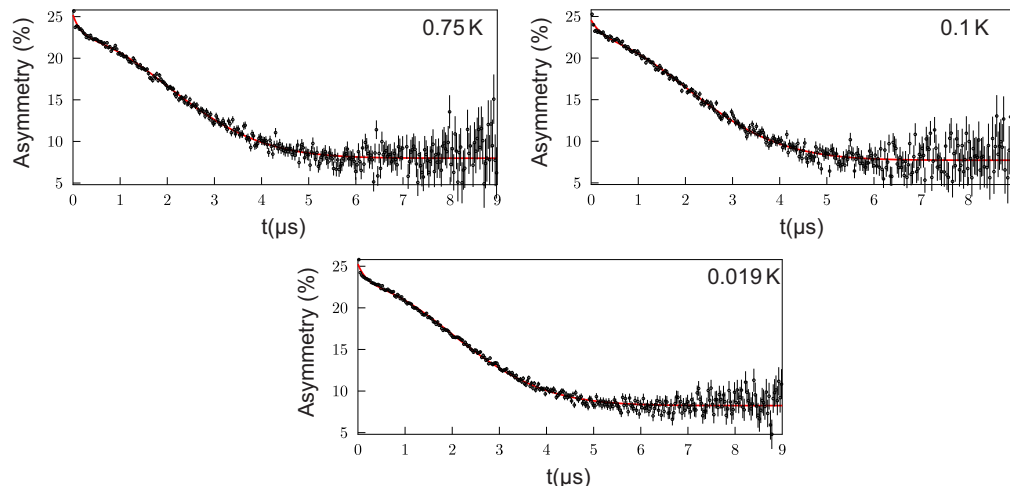


Figure B.6: Example asymmetry as a function of time for a polycrystalline sample of DPPH , measured at three temperatures

This asymmetry at each temperature (Fig. B.6) shows a broadly Gaussian relaxation, which we attribute to the magnetism of randomly oriented quasi-static nuclear spins. Significantly, the spectra are quite temperature independent up to $T = 0.8$ K and we do not observe any spontaneous oscillations in the asymmetry or a shift in the baseline that might be expected for a transition to a regime of long range magnetic order. We therefore conclude that the system is magnetically disordered down to $T = 19$ mK, with electronic moments fluctuating outside the muon time window, causing them to be motionally narrowed from the spectra.

B.6 Modelling the Temperature-Dependent Frequency Shift

In this section, the model leading to the fits in Figure 6.6 (b) is explained. For this purpose the spin chain Hamiltonian given in Equation 6.6 is used.

Because exchange interaction between spins leads to hybridisation of the eigenstates, the magnetic resonance frequency in general shifts away from the paramagnetic value as the crystal is cooled into its antiferromagnetic phase. At high temperature, spin correlations are negligible and the magnetic resonance frequency

is given by the single-spin Zeeman splitting. At lower temperatures, as nearby spins become correlated, there is an exchange shift. In the antiferromagnetic phase, the magnetic resonance frequency corresponds to the energy of a long-wavelength magnon.

For this one-dimensional model, the shift calculated in perturbation theory (Equation 6.7) was evaluated in Reference [200]:

$$\delta f_{\parallel} = -4 \frac{J^A}{h} \frac{\langle S_i^x S_{i+1}^x - S_i^y S_{i+1}^y \rangle}{\langle S^x \rangle / N} \quad \text{for } \mathbf{B} \text{ along } x \quad (\text{B.1})$$

$$\delta f_{\perp} = 2 \frac{J^A}{h} \frac{\langle S_i^y S_{i+1}^y - S_i^x S_{i+1}^x \rangle}{\langle S^y \rangle / N} \quad \text{for } \mathbf{B} \text{ along } y. \quad (\text{B.2})$$

where N is the number of spins in the crystal. Thus, given the anisotropic exchange strength J^A , the measured frequency shift is a direct measure of temperature-dependent correlation functions.

Extracting the exchange parameters J and J^A from data requires a knowledge of the correlation functions in Equations B.1-B.2. Even in this comparatively simple model, these are not known. However, we can estimate them following Reference [200] by making two additional assumptions:

1. We rely on the smallness of $\mathcal{H}'$ compared to $\mathcal{H}^0$ to approximate

$$\langle \dots \rangle \approx \langle \dots \rangle_0. \quad (\text{B.3})$$

In other words, the thermal expectation values in Equations B.1-B.2, which are determined by the entire Hamiltonian $\mathcal{H}$, can be replaced by thermal expectation values $\langle \dots \rangle_0$ under the unperturbed Hamiltonian $\mathcal{H}^0$.

2. We use a classical spin model in which the quantum mechanical spin operators are replaced by classical vectors [202].

Because of the rotational symmetry of $\mathcal{H}^0$, assumption (1) immediately leads to the equivalence of the two sets of expectation values in Equations B.1-B.2, implying that the frequency shifts along the two directions should differ by a factor -2. This is in fair agreement with the data of Figure 6.6, supporting this model.

Assumption (2) reflects the correspondence of quantum and classical pictures at high temperature. It allows the expectation values to be evaluated by expanding two-spin correlators at finite field in terms of higher-order correlators at zero field [200]. The result is (to lowest order in J^A/J):

$$\delta f_{\parallel} = \frac{2J^A g\mu_B B}{5hJv} \left\{ \frac{2+uv}{1-u^2} - \frac{2}{3v} \right\} \quad (\text{B.4})$$

$$\delta f_{\perp} = -\frac{\delta f_{\parallel}}{2}, \quad (\text{B.5})$$

expressed in terms of dimensionless parameters

$$u(J, T) \equiv \coth w - 1/w \quad (\text{B.6})$$

$$v(J, T) \equiv -1/w \quad (\text{B.7})$$

$$w \equiv \frac{2JS(S+1)}{k_B T}. \quad (\text{B.8})$$

The fit curves in Figure 6.6 derived from Equations B.4-B.5 are then:

$$B_{\text{MR}}(\parallel) = \frac{h(f_0 + \delta f_{\parallel})}{g\mu_B} \quad (\text{B.9})$$

$$B_{\text{MR}}(\perp) = \frac{h(f_0 + \delta f_{\perp})}{g\mu_B} \quad (\text{B.10})$$

with $g = 2$. The failure of the fits at low temperature presumably reflects the increasing importance of quantum correlations, neglected by assumption (2) above, or of interactions beyond one dimension. Thus, to the extent that our spin-chain approximation is correct, the data of Figure 6.6 provide a measurement of the spin correlation function that is more accurate than provided by the calculation relying on assumption (2) above.

C

Appendix: A Carbon-Nanotube-Based Superconducting Qubit

C.1 Additional Qubit Measurements

In this section measurements from an additional qubit device are presented, i.e. resonator and qubit spectroscopy from device *CNTQubit_v2_1_A_Q5*. Additionally, coherence measurements from a transmon qubit in a 3D-architecture are presented and two different measurement techniques compared.

C.1.1 Resonator and Qubit Spectroscopy

Figure C.1 shows the resonator and qubit spectroscopy described in Chapter 8.3.1 for one more qubit device. The behaviour is similar to the devices described in the main text. Here, the resonator spectroscopy exhibits upwards shifts in the resonance frequency at specific gate voltages. Simultaneously measured qubit spectroscopy shows a clear magnitude response at the same gate voltages, indicating the presence of a qubit with given ground-state $|g\rangle$ to excited-state $|e\rangle$ transition frequency, cf. analysis in Section 8.3.1.

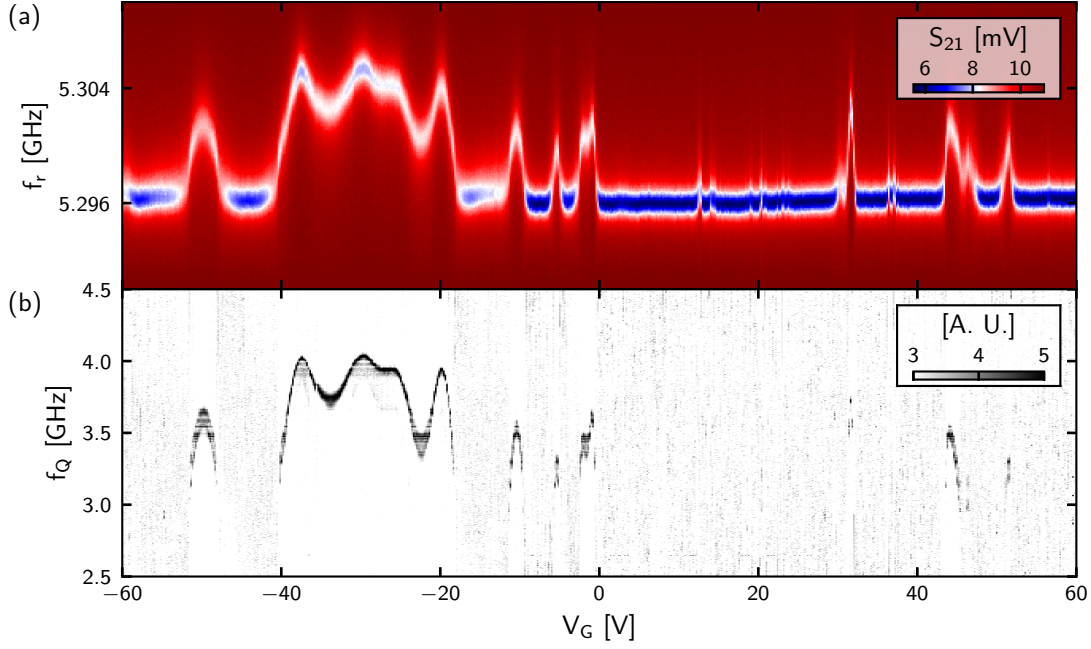


Figure C.1: Resonator (a) and qubit (b) spectroscopy as a function of applied gate voltage for device *CNTQubit_v2_1_A_Q5*.

C.2 Comparison of Coherence Measurement Techniques

In this section lifetime and coherence measurements of a transmon qubit in a 3D-cavity are presented. The measurements are performed in two different ways and their results compared, see Figure C.2. At first the qubit lifetime and coherence is assessed with the standard time-domain approach, using π - and $\pi/2$ -pulses. For this, a qubit pulse of varying length t is sent to the cavity at a fixed power and immediately after a measurement pulse is sent, in order to read the state of the qubit. This results in Rabi oscillations, see Figure C.2 (a). These oscillations are then used to determine the pulse length of a π - and $\pi/2$ -pulse, needed for the further analysis. In order to now determine T_1 of the qubit, a π -pulse is sent to the cavity and after the wait time t the state of the qubit is measured. Varying the wait time t results in an exponential decay of the qubit's excited state, see Figure C.2 (b). This data can be fit with an exponential decay resulting in a $T_1 = 3.2 \mu\text{s}$. To measure the decoherence time T_2 of the qubit, two $\pi/2$ -pulses separated by a time t are sent to the qubit and immediately after the second pulse the qubit state is measured

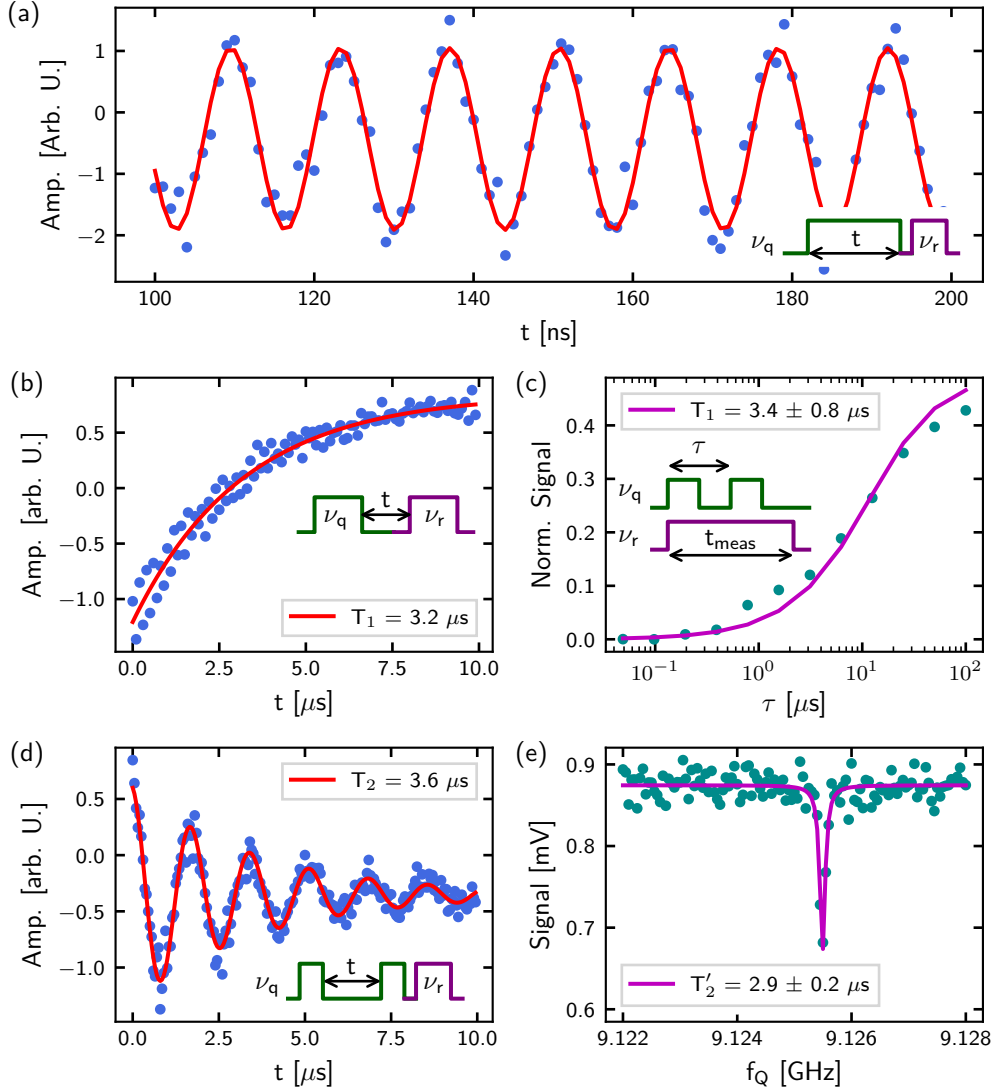


Figure C.2: Comparison of T_1 and T_2 measurement techniques for a transmon qubit in a 3D-cavity. (a) Rabi oscillations of the transmon qubit with sinusoidal fit (red curve). Inset: Pulse scheme to measure Rabi oscillations (qubit pulse - green, measurement pulse - purple). (b) Measurement of T_1 with a standard π -pulsed measurement. Red curve is an exponential decay fit, yielding $T_1 = 3.2 \mu\text{s}$. Inset: Pulse scheme to measure T_1 (qubit pulse - green, measurement pulse - purple). (c) Measurement of T_1 via chopped qubit microwave pulses with fit (purple curve) resulting in $T_1 = 3.4 \pm 0.8 \mu\text{s}$. Inset: Pulse scheme to measure T_1 with chopped microwave pulses whilst continuously measuring the resonator (qubit pulse - green, measurement pulse - purple). (d) Measurement of T_2 via standard Ramsey fringes, with fit to an exponentially decaying sine resulting in $T_2 = 3.6 \mu\text{s}$. Inset: Pulse scheme to measure Ramsey fringes (qubit pulse - green, measurement pulse - purple). (e) Estimating T_2 with a measurement of T_2' . A Lorentzian (purple curve) is fit to the low power qubit spectroscopy and its linewidth extracted.

(Ramsey interferometry). If the qubit pulse is slightly (~ 5 MHz) detuned from the qubit frequency and the pulse delay t is varied, this results in exponentially

decaying Ramsey fringes, see Figure C.2 (d). These Ramsey fringes can be fit with an exponentially decaying sine function, resulting in a $T_2 = 3.2 \mu\text{s}$.

The above described method is the standard way of characterising a qubit's lifetime T_1 and coherence time T_2 . However, in certain scenarios it is not possible to tune up a π -pulse through Rabi oscillations, which might either be due to very low coherence of the qubit or due to a mismatch between the internal and external quality factor of the measurement resonator. For the carbon nanotube superconducting qubits described in Chapter 8, both of these are the case. Hence, a different measurement technique for measuring T_1 and estimating T_2 has been used. Here, it is shown that both of these techniques lead to the same result.

In order to measure T_1 a method is borrowed from the field of quantum dot charge qubits, where microwave pulses at the qubit frequency are chopped up into a 50% duty cycle and the resonator is continuously measured [216, 217]. The period τ of the qubit pulse sequence is increased until only one period fits into the measurement cycle. The longer the qubit pulse period τ the more time the qubit has to relax. The resulting decay is normalised to the signal without a qubit drive on and fit to

$$S(\tau) = \frac{1}{2} - \frac{T_1(1 - e^{-\tau/(2T_1)})}{\tau}, \quad (\text{C.1})$$

see Figure C.2 (c)¹. This results in $T_1 = 3.4 \pm 0.8 \mu\text{s}$, where the value extracted from the standard pulsed measurement is within the error. The inaccuracy in the fit and the large error is due to the qubit's T_1 being quite long and the signal not being able to tail off completely due to restrictions in the measurement acquisition time.

The coherence time T_2 can be estimated from the low power linewidth of the qubit spectroscopy [218]. For this the linewidth is fit with a Lorentzian for different qubit drive powers. For low enough powers the linewidth should not change and $1/T_2' = 2\pi\delta\nu_{\text{HWHM}} = (1/T_2^2 + n_s\omega_{\text{vac}}^2 T_1/T_2)^{1/2}$ [14, 218]. This indicates that $T_2' < T_2$ and that T_2' is a lower bound for T_2 . By fitting the lowest power qubit spectroscopy with a Lorentzian, see Figure C.2 (e), we get $T_2' = 2.9 \pm 0.2 \mu\text{s}$, which is fairly close to the value measured with Ramsey fringes and gives a reasonable lower bound for T_2 .

¹It is worth mentioning that Eq. C.2 has a minus sign instead of a plus sign, cf. Eq. 8.4, as the measurement is done in reflection rather than in transmission.

Given the above results, we can conclude that the measurement techniques used in Section 8.3.3 in order to estimate the qubit's lifetime and coherence time are valid alternatives over the standard pulsed approach.

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