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Abstract

Vanadium in silicon carbide has been proposed as a platform for quantum communications due to its bright emission in the telecom O-band. However, several of the key properties for the application of this spin center as a qubit or quantum memory have not yet been characterized. In this thesis, the spin polarization, spin-lattice relaxation, and hyperfine structure of the vanadium center are examined using laser spectroscopy at cryogenic temperatures. It is demonstrated that optical polarization of the electron spin is possible, achieving contrasts in excess of 90%. At 100 mK, spin lattice relaxation times of over 20 seconds are observed, showing great promise for quantum memory applications. Further, a two-laser spectroscopy experiment is presented which reveals the defect's rich hyperfine structure.

Zusammenfassung

Vanadiumdefekte in Siliziumkarbid werden aufgrund ihrer hellen optischen Übergänge im Telekom O-Band als vielversprechende Qubitkandidaten für Anwendungen in der Quantenkommunikation gehandelt. Diese Masterarbeit ist ein Teil der Bestrebungen, diese Annahmen zu überprüfen und tiefere Einblicke in die Spineigenschaften dieser Defekte zu gewähren. Durch optische Spektroskopie an Defektensembles bei sehr niedrigen Temperaturen konnte gezeigt werden, dass es möglich ist, den Elektronenspin optisch zu polarisieren, wobei Kontraste von über 90% erreicht wurden. Bei 100 mK wurde eine T_1 -Relaxationszeit des Elektronenspins von knapp einer halben Minute beobachtet. Vielversprechend für quantentechnologische Anwendungen ist ebenfalls, dass bei Temperaturen rund um ein Kelvin Relaxationszeiten im Sekundenbereich gemessen wurden. Darüber hinaus ist es gelungen, mithilfe von Zwei-Laser-Spektroskopie die komplexe Hyperfeinstruktur der Defekte sichtbar zu machen.

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1 Introduction

In the last century, advances in physics and their implementation in technology have changed the human way of life at a faster pace than ever before. Information and communication technologies in particular have arguably led to the most dramatic changes in recent times, comparable to the invention of the printing press. However, they are still limited by the laws of classical physics. Currently, quantum physics and quantum information technologies are seen as a possible next big leap in innovation. While the impact of a universal quantum computer on the world is impossible to predict, quantum cryptography could present a solution to the very prevalent problem of secure communication and privacy in a more and more interconnected world, employing already validated schemes such as quantum key distribution (QKD) [1, 2].

Both distributed quantum computing and QKD will require a quantum network or quantum internet, similar to the classical internet, to connect any two entities that would like to exchange or transmit quantum information. In principal, this exchange would be carried out using photons much like in classical networks. There, transmission losses due to scattering and absorption in optical fibers lead to an exponential decrease in transmission probability with distance, which is counteracted by amplifying the optical signals along the channel. The very nature of quantum information, however, prohibits amplification, and a quantum internet would need different approaches to overcome this obstacle [3]. Satellites have shown to be very powerful in establishing a quantum connection between distant points on earth, as they take advantage of reduced optical losses in the vacuum of space [4, 5]. While this approach will be useful for long range connections, to establish a network within metropolitan areas and interconnecting individual users with information hubs, a fiber based network is believed to be more practical [5]. Such a network would be based on entangled nodes as elementary building blocks, which would serve two basic purposes.

Firstly, to compensate the higher losses photons suffer in fiber they would act as quantum repeaters in point to point connections [6]. To illustrate the fundamental mechanism, one can consider four quantum nodes A, B, C and D that are connected through optical fiber in series. The repeater sequence starts by establishing entanglement between two distant nodes A and B, while at the same time entangling C and D, forming elementary quantum links. Now, performing an entanglement swapping operation on the nodes B and C, results in entanglement between the nodes A and D, therefore extending the quantum link. This process could be repeated over many nodes to create an arbitrarily long quantum link that is only limited by the coherence time T_2 of the quantum node [7, 8]. Placing several repeater nodes at each point in the link that would constantly generate entanglement, and employing entanglement purification and quantum error correction on them, is expected to recoup the higher optical losses in fiber and outperform satellites at regional scales [4].

Secondly, quantum nodes would act as a quantum memory, storing quantum information for on demand readout. Such nodes could construct a quantum network,

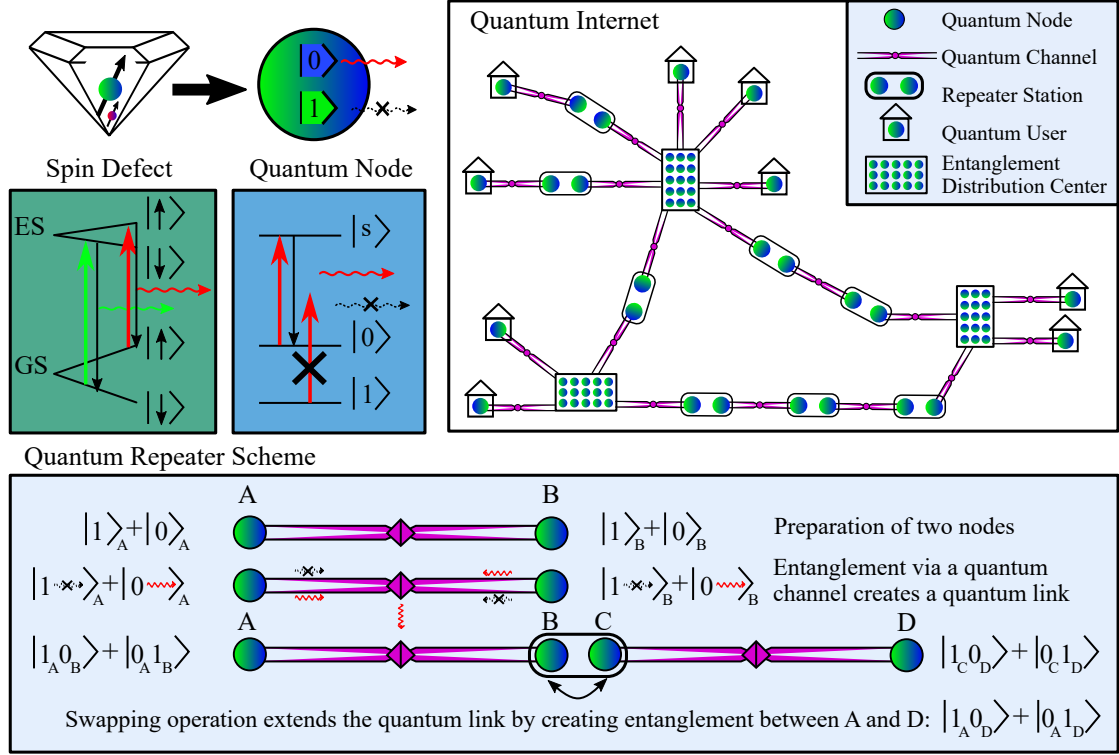


Figure 1: Possible implementation of a node based quantum network: A color defect in crystal with an electron (blue green) and a nuclear (red blue) spin, that has spin dependent optical transitions, can form a qubit or quantum node. The node requires states $|0\rangle$ and $|1\rangle$ for quantum information and a state $|s\rangle$ which can be used for optical readout of the qubit. In the bottom panel a simplified working principle of a quantum repeater is shown. First, a quantum link between two nodes, connected via a quantum channel, is established. This is followed by entanglement swapping to extend the quantum connection. An illustration of a quantum network can be seen in the top right panel. Here each node (blue green circle) represents tens or hundreds of nodes which would be used in parallel.

where quantum repeater enhanced entanglement links connect quantum end users to interconnected entanglement distribution centers, consisting of thousands of quantum nodes that would use the aforementioned operations to rout entanglement and create a quantum link between any two parties in the network [4, 9]. An illustration showing a possible realization of a quantum network can be seen in fig. 1, and while the displayed entanglement method (photon - no photon), which is chosen for its simplicity, is very susceptible to photon loss, more robust schemes, such as time to polarization conversion, can be used to create heralded entanglement, relying on the same fundamental mechanisms [10]. Similar node based optical entanglement architectures have also been proposed as a possibility to build a quantum computer [11].

A quantum node would therefore need to meet certain requirements. It would have to operate in the near infrared telecom band for optimal transmission in fiber, and contain a qubit with a good fidelity, controllability, and long coherence times ($T_2 \gtrsim 10$ ms). As with any real life application, practical aspects also come into play, such as scalability, manufacturing and cost. Currently many types of qubit platforms are being investigated, but so far no single platform has met all the parameters required [5]. One very promising system are solid state spin (or sometimes color) centers, which consist of a spin system formed by an impurity in the crystal. Such a spin system is created by individual defects in the lattice, usually with two or more electronic spin states that allow for gate operation and state specific optical readout and initialization. Furthermore, the quantum state can be swapped into the defect's own, or even adjacent atoms', nuclear spin allowing for long term quantum information storage [9, 12]. The most famous example is the nitrogen vacancy (NV) center in diamond, that has not only allowed researchers to investigate many fundamental questions posed by quantum physics, but also expanded the possibilities of magnetometry [13, 14]. While the NV center and similar systems have shown to be very viable for sensor applications and some quantum computing approaches, they are not considered ideal candidates for the emerging field of long distance quantum communication, limited by high optical losses of the corresponding wavelengths and low optical emission into the zero phonon line (ZPL) [9].

This has sparked a hunt for new solid state qubits, with bright near infrared (NIR) transitions, long lasting coherence, good controllability and a proven manufacturing process [15]. Many different host crystal materials, such as diamond and silicon, are currently being investigated for defects and their quantum properties. One material that has gained momentum recently is silicon carbide (SiC), which in many aspects has similar properties to diamond. Its large band gap of over 3 eV [16] gives ample room for optical transitions, making it a promising host. The neutral vanadium defect V^0 , also referred to as V^{4+} , is a spin 1/2 system that could meet the requirements [17, 18]. It has bright optical ZPL transitions in the telecom O-band ($\lambda = 1260$ nm – 1360 nm), which could prove particularly useful, as this band is currently not used for long distance communication between major hubs. This would allow quantum and classical information to use the same infrastructure, by multiplexing and demultiplexing the different bands at internet exchange points [19].

However, there are still many open questions when it comes to the quantum properties of this defect. One important parameter for the quantum coherence time T_2 of a solid state qubit, is the electron spin lifetime T_1 . It gives a theoretical upper bound for the coherence time ($T_2 \leq 2 T_1$) of such a defect, and also a good estimate of the T_2 timescales actually observed ($T_2 \approx (0.1 - 0.5) T_1$) [12, 15]. Even though other defects in SiC have been shown to have long electron spin lifetimes, such as molybdenum ($T_1 = 2.4$ s at 2 K) [20], for vanadium no spin lifetime has been observed so far, even below liquid helium temperatures at around 3 K [18]. Other key questions regard the charge stability of the defect, as well as the hyperfine level structure and its optical behaviour and polarizability, which are all prerequisites for the realization of a solid state qubit.

This thesis aims to answer some of these open questions and to evaluate whether vanadium in silicon carbide has potential as a qubit. To achieve this, three of the numerous defect sites were investigated through different methods of optical spectroscopy. Those measurements were conducted at temperatures in the millikelvin range, since cooling a crystal to such cold temperatures has been shown to increase the lifetime of the electron spin immensely (up to several hours) [21].

First, resonant photo-luminescence spectroscopy measurements were performed under different conditions. Further, the charge state stability of the defects was examined by means of bleaching and subsequent spectroscopy. Next, optical spin pumping and polarization was demonstrated, which was used to measure the spin lattice relaxation rate Γ_1 ($1/T_1$), over a wide range of temperatures, from 100 mK to 1.9 K. Finally, two laser spectroscopy was employed to reveal the hyperfine structure of the defects, with unprecedented resolution. All of the measurements presented were performed on ensembles of vanadium in silicon carbide wafer material.

In the following chapters, this thesis will first give background information on the physics used herein, followed by an explanation of the experimental apparatus built and used during the research. The results of the experiments will be presented and discussed in chapter 4, where all measurement schemes will also be elaborated on. Finally, the gained knowledge will be interpreted and placed within the context of current research, and further steps necessary towards developing the V^0 defect in SiC as a qubit will be summarized in the last chapter.

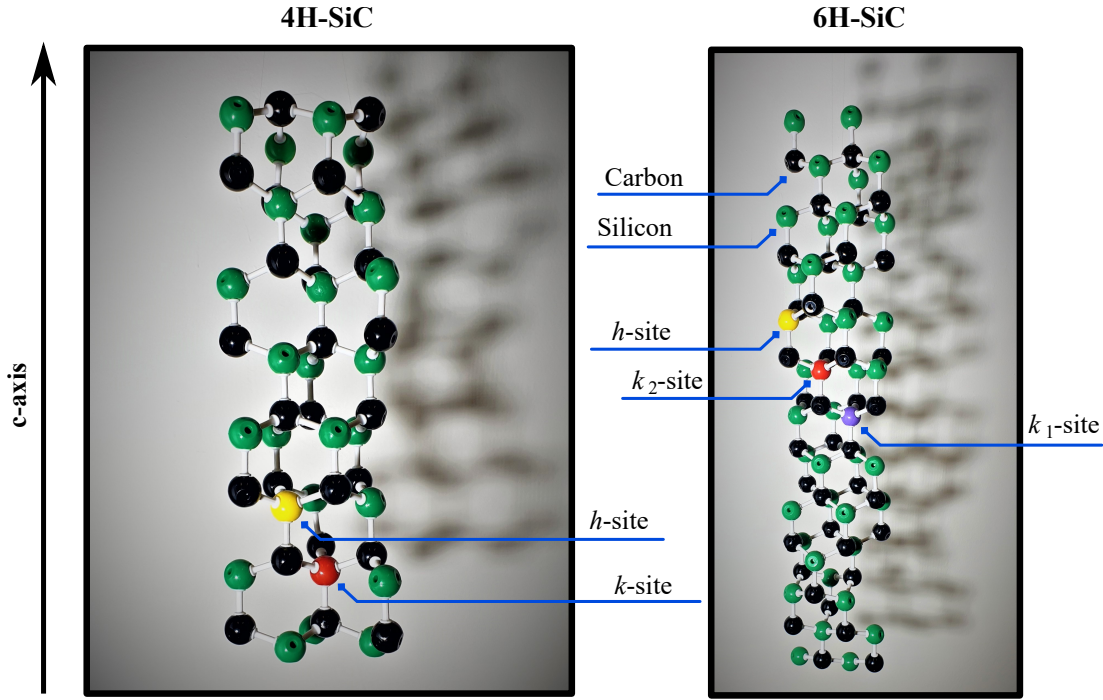


Figure 2: Photograph of crystal models of the 4H and 6H polytypes of silicon carbide with a length of two unit cells in the *c*-axis direction. Carbon is represented by black spheres and silicon by green ones. The colored spheres show the possible locations a silicon substitute can occupy, yellow for quasihexagonal sites (*h*-site) and red and purple for the quasicubic sites (*k*- or *k*₁- and *k*₂ - sites respectively).

2 Background

2.1 Silicon Carbide

Silicon carbide (SiC), also referred to as carborundum, is a semiconductor made of silicon and carbon. Together they form a crystal which can be considered as a closely packed stacking of double layers of Si and C atoms, with a distance of 1.89 Å. They are bond together by strong tetrahedral sp^3 bonds that are 88% covalent and 12% ionic. The double layers can stack in three possible relative configurations, named A, B and C, which allows for infinitely many stacking arrangements, also known as polytypes [22].

Even though SiC has been known for almost 200 years, it has taken until the 1990s for the manufacturing process to mature, to produce wafer grade bulk material. Its extreme stiffness makes it not only a good abrasive, but is very useful as a substrate for telescope mirrors and other applications that would otherwise require diamond [23]. Compared to diamond it has a higher refractive index of around 2.7. As a semiconductor it is known for its high band gap (up to 3.3 eV).

Semiconductor applications of silicon carbide include UV detectors, high voltage diodes, as well as radiation resistance electronics and integrated circuits that can operate at very high temperatures. To alter the electrical properties, often dopants are added, the most common ones being nitrogen to create n-type SiC and aluminium or boron for p-type. Recently, research into the optical and spin properties of defects has gained momentum, as there are strong indications that SiC is an advantageous platform for solid state qubits [17].

2.1.1 Polytype Properties

Out of the infinitely many possible polytypes that can be created by stacking the layers A, B and C, in this thesis the 4H and 6H polytypes were used. 4H SiC has a stacking of ABAC of the silicon and carbon bilayers and can be seen in fig. 2 LEFT. This results in unit cells that contain 8 atoms and makes it 50% hexagonal, meaning that half of the atoms experience a hexagonal local environment. 6H SiC has a bigger unit cell of 12 atoms and is layered ABCACB (see fig. 2 RIGHT), leading to a hexagonality of only 33.3% [22].

Even though both polytypes are made up of the same elements and held together by the same bonds, the different geometries lead to rather different properties. While the band gap of 4H SiC is 3.26 eV, 6H SiC has a reduced band gap of 3.02 eV. Also the electron mobility is significantly higher in the 4H polytype and less anisotropic than in 6H [16].

As this thesis studies the neutral vanadium V^0 defect that substitutes for a silicon atom, there are 5 different sites that it can occupy. In 4H SiC, this kind of symmetry creates two possible sites a dopant can occupy, one quasihexagonal h - and a quasicubic k -site, resulting in two versions of the defect with distinct properties. The 6H polytype has 3 possible locations for such a substitutional defect, one quasihexagonal h -site and two quasicubic sites, called k_1 and k_2 .

2.2 Vanadium Hamiltonian

The neutral V^0 defect in silicon carbide belongs to the group of transition metal (TM) defects that substitute a silicon atom in the lattice. As the vanadium atom is bound to other atoms within a lattice, its orbital dynamics are mostly influenced by its interaction with these neighbouring atoms and their potentials. The energy levels of the defect are therefore significantly different from those of a free vanadium atom. Four of its electrons form bonds with adjacent carbon atoms and can be considered part of the valence band (hence the alternative name V^{4+}). The defect therefore has one unbound electron with spin=1/2. This electron, if it were influenced only by the coulomb forces from the nucleus, would occupy the fivefold degenerate d-orbital [24]. Like in other solid state defects, this is used as a starting point. Using group theory and symmetry approaches, combined with experimental findings, a Hamiltonian of the following form was derived for the vanadium defect in silicon carbide:

$$H = H_{TM} + V_{CR} + H_{SO} + H_{Hf} + H_Z. \quad (1)$$

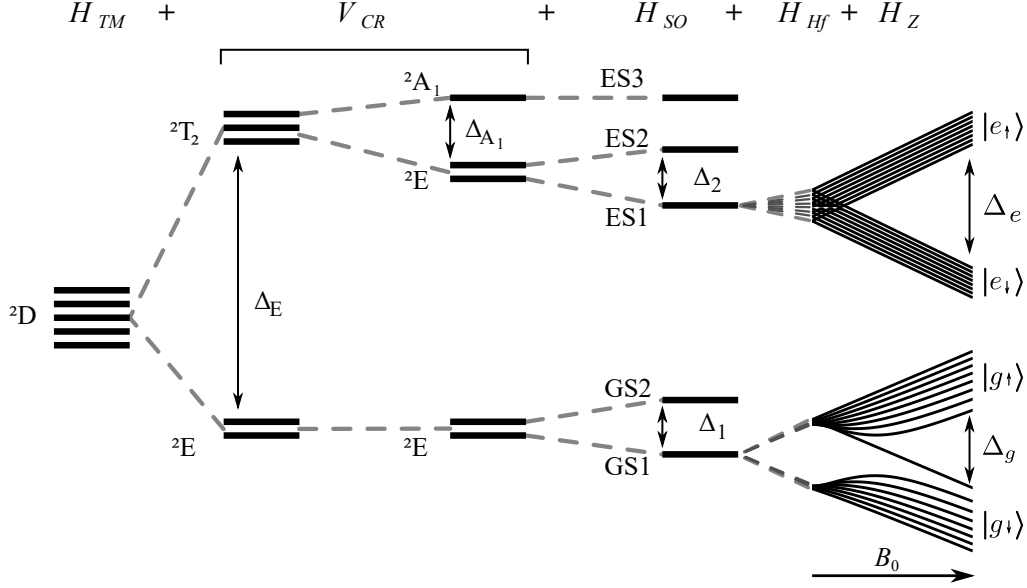


Figure 3: Schematic of the energy levels of the vanadium electron in silicon carbide, resulting from the Hamiltonian shown in eq. (1). The fivefold degenerate d-orbital is first split by the crystal potential and then by spin orbit coupling into five orbital states. For the GS1 and the ES1 the effects of the hyperfine interaction and the Zeeman effect are further visualized, resulting in 16 Zeeman states for each orbital level.

Here, the first term H_{TM} describes free atom electron d-shell orbital wave function, which is then further extended by the crystal potential of the lattice V_{CR} , spin orbit coupling H_{SO} , hyperfine coupling H_{HF} and finally the Zeeman effect H_Z [25]. The main contribution to the electron's energy comes from $H_{TM} = \frac{p^2}{2m_e} + V_{TM}$, with V_{TM} the Coulomb potential of the nucleus and $\frac{p^2}{2m_e}$ the kinetic energy of the electron. This energy contribution, however, can be ignored for a description of the internal orbital dynamics, as long as the electron is always localized within the d -orbital.

2.2.1 Crystal Potential

The crystal potential contribution can be split into two components, which come from lattice geometry. First, the spherical symmetry of the d -orbital is broken by the tetrahedral symmetry (T_d) of the lattice where the defect is embedded. This lifts the fivefold degeneracy into a ground state orbital doublet (^2E) and an excited state orbital triplet ($^2T_2$), separated by a splitting Δ_{T_d} . Next, the tetrahedral symmetry is further reduced to a C_{3v} point defect symmetry group, which splits the excited state orbital triplet ($^2T_2$) into an orbital doublet (^2E) and an orbital singlet state ($^2A_1$), separated by an energy Δ_{A_1} [25]. This would be sufficient to explain the effect of a tetrahedral symmetry, found in the 3C polytype of silicon carbide. However, in the polytypes investigated in this thesis, which exhibit hexagonality to some degree, there is a trigonal symmetry contribution by second and third nearest neighbours, which is resolved by replacing Δ_{T_d} with $\Delta_E = \eta\Delta_{T_d}$. In this case η is a factor that

can be varied to adjust for the splitting of the different sites [26].

2.2.2 Spin Orbit Interaction

Another contribution is the effect of spin-orbit coupling which arises from the interaction between the orbital and the spin degree of freedom of the electron:

$$H_{SO} = \lambda_{kl} \mathbf{L} \cdot \mathbf{S} . \quad (2)$$

It is comprised of the spin operator $\mathbf{S} = \frac{\hbar}{2} \vec{\sigma}$, the orbital angular momentum operator $\mathbf{L}$ and the reduced coupling matrix element λ_{kl} , giving the strength of the coupling for each particular defect, taking into account the influence of the lattice symmetry and nearby potentials. This splits the ground and excited ${}^2E$ orbital doublet states further, creating four so-called Kramers doublet states [25]. Such states are described by Kramers theorem, stating that for a system with an odd number of electrons, potential and kinetic energy as well as spin-spin and spin-orbit interactions are invariant through time reversal, in the absence of an external magnetic field [27].

As a result each Kramers doublet is a new twofold degenerate orbital state, comprised of two combinations of spin and orbital degree of freedom for which $\mathbf{L}_z \cdot \mathbf{S}_z$ are equal. In ground states this leads to a splitting of Δ_1 , and in the excited state doublet Δ_2 [25]. Together with the contributions to the Hamiltonian discussed above, these terms lead to a splitting of the d-orbital into five orbital states, as shown in fig. 3.

In the Kramers doublet ground states, the lower state will from now on be referred to as ground state 1 (GS1) and the upper state as ground state 2 (GS2). Similarly, in the excited state the Kramers doublets resulting from the splitting of the ${}^2E$ orbital doublet state shall be called excited state 1 (ES1) and 2 (ES2), with ES1 being the lower state. Finally, ${}^2A_1$ will be referred to as excited state 3 (ES3).

2.2.3 Hyperfine Interaction

The last intrinsic contribution to the Hamiltonian, which is independent of external magnetic or electric fields, is the hyperfine interaction H_{HF} . It accounts for the effect of the nuclear spin ($I=7/2$) on the electron ($S=1/2$). This can be implemented in a simple form using the electron $\mathbf{S}$, the nuclear $\mathbf{I}$ spin operators and the hyperfine tensor $\mathbf{A}$ [18]:

$$H_{HF} = \mathbf{S} \cdot \mathbf{A} \cdot \mathbf{I} . \quad (3)$$

This approximation neglects interactions with neighbouring nuclear spins, even though neighbouring atoms can have non-zero nuclear spins. However, their natural abundances for Si and C are very low [28, 29]. During the writing of this thesis a refined Hamiltonian was published, including Fermi contact terms and orbital angular momentum coupling to the nuclear spin [25].

2.2.4 Zeeman Effect

Exposing the system to a constant external magnetic field B_0 will lead to a shift in energy, known as the Zeeman effect. The spins couple to the external field which

leads to the Zeeman Hamiltonian contribution in the form of

$$H_Z = \mu_B B_0 \cdot g_{state} \cdot S - \mu_N g_N B_0 \cdot I , \quad (4)$$

with μ_B being the Bohr magneton, g the electron g-factor, μ_N the nuclear magneton and g_N the nuclear g-factor. It is important to note that for electrons in a lattice the g-factor is not necessarily ≈ 2 , like for free electrons, but can deviate significantly depending on the site, orbital and direction of B_0 , showing strong anisotropy [30]. In this thesis, as magnetic fields will only be applied parallel to the c-axis, the state specific g-factor g_{state} will refer to the parallel component of the electron's effective g-factor g . For many descriptions of the effect of external magnetic fields, the nuclear component of the Zeeman Hamiltonian can be neglected as its contribution is smaller by around 4 orders of magnitude compared to the coupling of the electron spin to the field [25]. Combined with the hyperfine interaction this leads to a splitting of each orbital state into 16 Zeeman states for $B_0 \neq 0$ with a field dependence, also illustrated in fig. 3.

2.2.5 Summary

At $B_0 = 0$ the five degenerate states in the d-orbital given by H_{TM} are split into five states by the effect of the crystal lattice potential V_{CR} and spin orbit coupling H_{SO} . These are further split by the electron spin $S=1/2$ and nuclear spin $I=7/2$ interaction given by H_{HF} . Applying a magnetic field lifts the degeneracy of the electron spin and gives rise to 16 different energy levels in each of the orbital states. Ignoring the nuclear spin's contribution, the shift of each electron state energy is

$$\Delta_{Ze} = \mu_B B_0 \cdot g \cdot S \approx \frac{14 \text{ GHz}}{\text{T}} \cdot B_0[\text{T}] \cdot g \cdot \left(\pm \frac{1}{2} \right) , \quad (5)$$

with g being a specific constant for each site and orbital level. In this simplified level scheme the electron spin state with higher energy is $|\uparrow\rangle$ and $|\downarrow\rangle$ is the state with lower energy. This leads to a splitting between $|\uparrow\rangle$ and $|\downarrow\rangle$ of $2\Delta_{Ze} \approx 14 \frac{\text{GHz}}{\text{T}} \cdot B_0[\text{T}] \cdot g$ as an approximation for the electron spin Zeeman effect.

Importantly, the g-factors for the ground state and excited state of an optical transition are not necessarily identical. This can be used to separate the optical transitions according to the electron spin state. As the ground and excited state shift roughly according to eq. (5), but with different g-factors, the energy difference between spin conserving optical transitions ($\Delta_{optical}$) is given by

$$\Delta_{optical} \approx \mu_B B_0 \cdot (g_{ES} - g_{GS}) \approx \mu_B B_0 \cdot \Delta g \approx \frac{14 \text{ GHz}}{\text{T}} \cdot B_0[\text{T}] \cdot \Delta g . \quad (6)$$

Here g_{ES} and g_{GS} are the excited and ground state g-factors. This can be used to separate the spin conserving optical transitions depending to the electron spin state by applying a magnetic field, as long as $\Delta g \neq 0$.

	4H- α	6H- α	4H- β
GS1-ES1 (nm)	1278.8	1308.6	1335.3
GS1-GS2 (GHz)	529	524	43
ES1-ES2 (GHz)	181	-	167
τ (ns)	≈ 167	≈ 50	≈ 45
g_{GS1}	1.748	1.749	1.870
g_{ES1}	2.24	2.24	2.03

Table 1: Overview of known properties of the defects investigated in this thesis taken from [18, 30, 31]. The values of the g -factors displayed are the parallel components, as those were relevant in this thesis.

2.3 Vanadium sites in Silicon Carbide

In 4H and 6H silicon carbide there are different geometries in which a vanadium atom can substitute a silicon atom. The site assignment of the observed optical resonance to a lattice defect site ($k\ k1\ k2\ h$) is not yet well established, with existing literature coming to contradictory conclusions [18, 32]. For this reason the unassigned site names α , β , and for 6H also γ , will be used in this thesis, in accordance with previous research into the optical properties of the neutrally charged vanadium defect V^0 . All these sites have been shown to emit light when illuminated with above-band gap blue light and also when excited with a resonant laser, with photons emitted into the optical phonon sideband (PSB) [18]. Early experiments also showed that it is possible to burn spectral holes in the defects resonance by means of two-step photoionization, where an excited electron absorbs a second photon, and is then trapped in some other acceptor level in the crystal [33]. A schematic of the electronic levels, and the resulting possible optical excitations, for the $V^0 - \alpha$ defect of interest can be seen in fig. 4.

The following sections will present the current state of knowledge for the defects investigated, i.e. the 4H- α and - β , and the 6H- α site, as established by previous research. Important defect parameters used in this thesis are summarized in table 1.

2.3.1 4H Alpha

For the 4H alpha defect (4H- α), two ground states (GS1 and GS2), as well as two excited states (ES1 and ES2) have been identified, but an ES3 has not been observed. An optical transition between GS1 and ES1 was found to have a zero phonon line wavelength of 1278.8 nm, showing strong emission into the phonon sideband with wavelengths $\lambda > 1300$ nm. The two Kramers doublet ground states are separated by 529 GHz, but only the parallel component of the g -factor of GS1 has been identified ($g_{GS1} = 1.748$). Optical excitation leads to pumping into the excited states with an optical lifetime of $\tau = 167$ ns, where the two excited states are separated by 181 GHz. For the state ES1, the g -factor along the c -axis is $g_{ES1} = 2.24$, deviating from g_{GS1} and making it possible to separate the optical transition energies by Zeeman splitting [18].

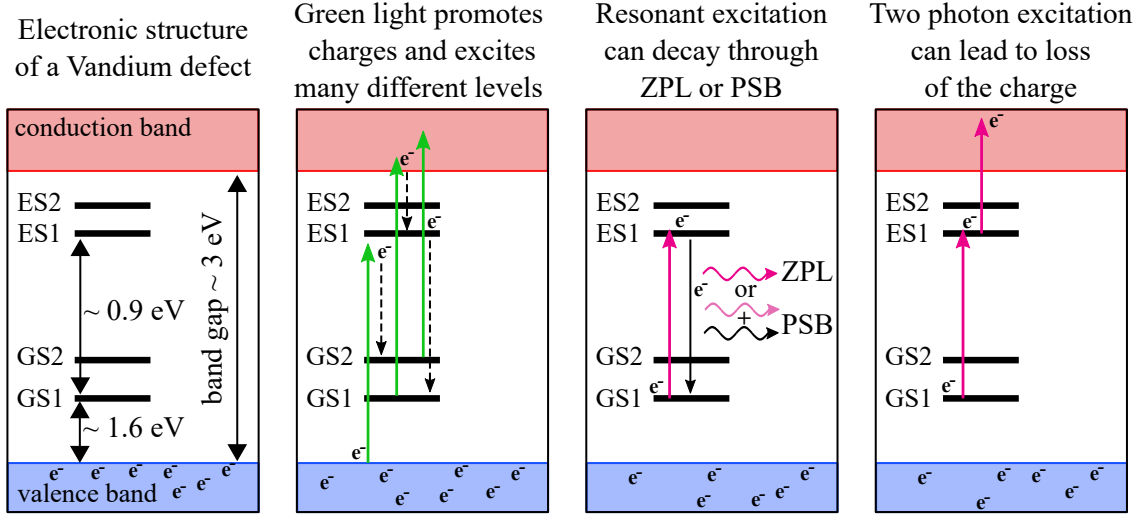


Figure 4: Schematic of the electronic structure of an example vanadium defect (e.g. 6H- α). Above band-gap illumination (in this case green) can promote charges from the valence band to the neutral charge state (V^0), as well as drive many defect transitions in the crystal. Resonant excitation drives a specific transition, which can decay through the zero phonon line (ZPL), or the phonon sideband (PSB), where a lower energy photon is emitted together with one or more phonons. If an electron in the excited state absorbs a second photon, the electron can be lost to the conduction band.

2.3.2 4H Beta

A different site geometry for the 4H beta defect (4H- β) leads to very different properties, compared to its site neighbour Alpha, only displaying one excited state (ES1) and two Kramers ground states with a splitting GS1-GS2 of 43 GHz. The optical GS1-ES1 transitions zeron phonon line has a wavelength $\lambda = 1335.3\text{ nm}$ with a shorter optical lifetime of $\tau = 45\text{ ns}$. All the parallel components of the electron g-factor are known for both ground states ($g_{GS1} = 1.87$ and $g_{GS2} = 2.035$) and the excited state $g_{ES1} = 2.03$ [18].

2.3.3 6H Alpha

Of the three defects in 6H silicon carbide, only the alpha site (6H- α) will be discussed in this thesis. It shows similar properties as its counterpart in 4H, with two Kramers doublet ground states (GS1 and GS2) separated by 524 GHz and two excited states (ES1 and ES2) with a splitting of 167 GHz. The optical GS1-ES1 transition has a wavelength of $\lambda = 1308.6\text{ nm}$ and an optical lifetime of $\tau = 108\text{ ns}$. For the ground state 1, the g-factor is known to be $g_{GS1} = 1.749$ and in the ES1 $g_{ES1} = 2.24$ [18], just like in the 4H Alpha site, also making it possible to split the spin states' optical spin conserving transition by applying a magnetic bias field.

2.4 Spin Lattice Relaxation

This chapter will discuss the theory and mechanisms of spin lattice relaxation and follows the book "Electron paramagnetic resonance of transition ions", by A. Abragam and B. Bleaney [27], closely.

The Zeeman effect splits the energy levels of a spin $S = \frac{1}{2}$ system in the presence of a magnetic field B_0 , by $\Delta E = h\nu = \mu_B g B$. This results in a higher energy spin up $|\uparrow\rangle$ and lower spin down $|\downarrow\rangle$ state, creating an effective two level system. Applying an oscillatory magnetic field of the corresponding frequency ν drives transitions between the two states, where a transition from $|\downarrow\rangle$ to $|\uparrow\rangle$ corresponds to the absorption of a photon and the inverse process to an emission. For an ensemble with population N , the ratio between the instantaneous populations ($n_{|\uparrow\downarrow\rangle}$) can be described by a spin temperature T_S :

$$\frac{n_{|\uparrow\rangle}}{n_{|\downarrow\rangle}} = \exp\left(-\frac{h\nu}{k_B T_S}\right), \quad (7)$$

where k_B is the Boltzmann constant and $n_{|\uparrow\rangle} + n_{|\downarrow\rangle} = N$. The populations become equal for $T_S \rightarrow \infty$ and a population inversion $n_{|\uparrow\rangle} > n_{|\downarrow\rangle}$ formally corresponds to a negative T_S . If the spins are coupled to a thermal bath of temperature T_0 with infinite heat capacity the equilibrium population ($N_{|\uparrow\downarrow\rangle}$) distribution is given by:

$$\frac{N_{|\uparrow\rangle}}{N_{|\downarrow\rangle}} = \exp\left(-\frac{h\nu}{k_B T_0}\right), \quad (8)$$

again with $N_{|\uparrow\rangle} + N_{|\downarrow\rangle} = N$. When the spin populations are out of equilibrium, over time the spins will relax into the thermal equilibrium, hence $T_S \rightarrow T_0$. In a solid state system, the thermal bath temperature is given by the temperature of the crystal and $T_S \rightarrow T_0$ holds if the thermal capacity of the crystal is far bigger than the spin heat capacity, or if the crystal is connected to a thermal bath. Unlike optical transitions, for which relaxation into the ground state is dominated by spontaneous emission, for this type of relaxation in solids, whose energy corresponds to radio frequencies, this process is negligible. Relaxation in solids is mostly induced by lattice vibrations if $T_S \neq T_0$:

$$\frac{d(n_{|\downarrow\rangle} - n_{|\uparrow\rangle})}{dt} = \frac{1}{T_1} \{ (N_{|\downarrow\rangle} - N_{|\uparrow\rangle}) - (n_{|\downarrow\rangle} - n_{|\uparrow\rangle}) \}. \quad (9)$$

Here the characteristic time constant T_1 is the spin-lattice relaxation time, also referred to as electron spin lifetime, and equal to the inverse of the spin relaxation rate $\Gamma_1 = 1/T_1$. Solving eq. (9) gives the behaviour of the instantaneous spin population distribution over time for a given initial spin population $(n_{|\uparrow\downarrow\rangle})_0$:

$$(n_{|\downarrow\rangle} - n_{|\uparrow\rangle})(t) = (N_{|\downarrow\rangle} - N_{|\uparrow\rangle}) + \{ (n_{|\downarrow\rangle} - n_{|\uparrow\rangle})_0 - (N_{|\downarrow\rangle} - N_{|\uparrow\rangle}) \} \exp(-t/T_1). \quad (10)$$

Setting $(N_{|\downarrow\rangle} - N_{|\uparrow\rangle}) = c$ and $n_{|\uparrow\rangle} + n_{|\downarrow\rangle} = N = 1$ gives the following expression for the spin $|\downarrow\rangle$ population

$$n_{|\downarrow\rangle}(t) = (1 - c)(1 - \exp(-t/T_1)) + c = (1 - c)(1 - \exp(-t \cdot \Gamma_1)) + c, \quad (11)$$

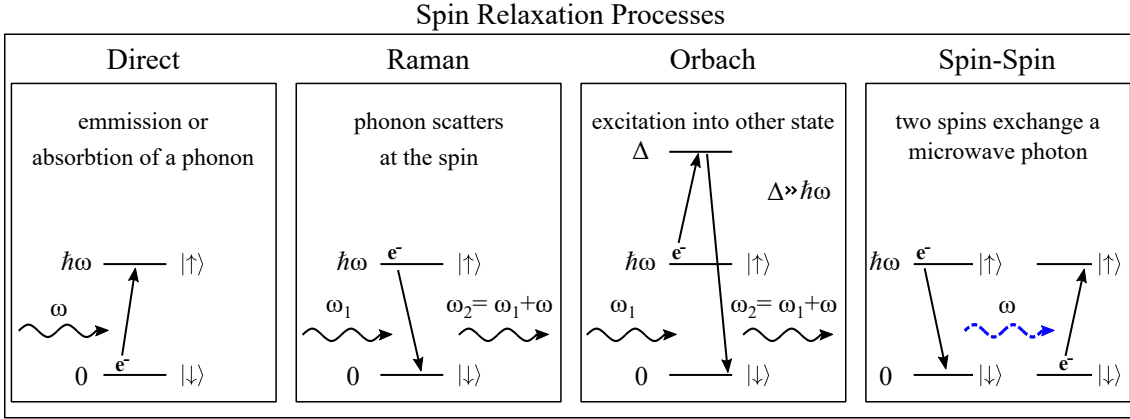


Figure 5: Schematic of common spin lattice relaxation processes through which an electron spin polarization can be lost. For the direct process, an electron can flip its spin by either absorbing or emitting a resonant phonon with energy corresponding to the splitting of the spin states ($\hbar\omega = \hbar g\mu_B B_0$). A Raman process occurs when a non resonant phonon scatters at the spin and flips it. The scattered phonon's energy then changes by $\pm\hbar\omega$. In the presence of a far higher energy level (Δ), the Orbach process can cause the electron to lose its spin polarization by being excited into the higher state, and decaying into the other spin state. Two spin systems in close proximity can also exchange a quantum of energy directly between each other causing a spin-spin or flip-flop interaction.

which is the form of equation used to analyse the experiment in the later chapters of this thesis.

As transitions between the spin states can only be induced by an oscillatory magnetic field, we need to explain how these can be induced by lattice vibrations. Early considerations involving phonon induced modulations of the ions' dipole proved to be insufficient to estimate the observed relaxation rates. Current theory relies on spin-lattice relaxation by the vibrational modulation of the ligand field. A way to describe the crystal electric field distortions, is by expanding the crystalline potential V :

$$V = V^{(0)} + \epsilon V^{(1)} + \epsilon^2 V^{(2)} + \dots \quad (12)$$

Here $V^{(0)}$ is the zero order static crystal potential of eq. (1), followed by additional higher order potential terms created by a phonon-induced fluctuating strain ϵ . This does not cause a direct interaction with the electron spin, but works similar to how the static crystal potential splits the energy levels of a spin defect and influences the spin-orbit coupling (see section 2.2). Analogous, a fluctuating crystal potential can induce transitions between these levels, coupling to the spin indirectly. The most common lattice vibration relaxation mechanisms for electron spins in solid states, described by quantized lattice vibrations known as phonons, are illustrated in fig. 5 and will be discussed in the following sections.

2.4.1 Direct Process

The first type of relaxation to consider is the direct process, where an electron either absorbs or emits one single lattice phonon with energy $\omega = g\mu_B B_0$. For Kramers doublets this interaction is only possible at non zero magnetic fields. In the presence of an external field B_0 or a local nuclear magnetic moment [26], the states no longer possess time reversal symmetry. Then a direct relaxation is allowed, with the following dependence:

$$\Gamma_{1 \text{ direct}} \propto B_0^5 \coth \left(\frac{g\mu_B B_0}{2k_B T_0} \right) . \quad (13)$$

This relation describes the direct process well, until $g\mu_B B_0 \ll k_B T_0$, with T_0 being the temperature of the thermal bath or crystal and assuming that the splitting due to the external magnetic field is bigger than the hyperfine splitting.

2.4.2 Raman Process

The direct process involves only one phonon of energy ω to flip the electron spin, however two phonon relaxation is also possible. One such mechanism is the two phonon Raman process, relying on an analogous process to optical Raman scattering. It involves a non resonant phonon with energy ω_1 being (virtually) absorbed, followed by an immediate (virtual) emission of a phonon with energy ω_2 . This requires two lattice phonons that fulfill $\omega_1 - \omega_2 = \pm\omega$ in order to maintain energy conservation, where the energy difference ω is either absorbed or emitted by the electron. Even though this is a second order process, the fact that there are far more phonons that can partake in this process than in the direct process makes it the dominant process at temperatures above 1 K for transitions in the GHz range as it scales strongly with temperature:

$$\Gamma_{1 \text{ Raman}} \propto T_0^n . \quad (14)$$

The value in the exponent n is defect and lattice dependent with values reported ranging from 5 to 11 with $n = 9$ being a typical value for Kramers doublet electrons.

2.4.3 Orbach Process

In an Orbach process, a high energy phonon with energy ω_1 excites the electron from the $|\uparrow\rangle$ or $|\downarrow\rangle$ ground state level to an external much higher level with energy Δ , followed by a subsequent emission of a second phonon with energy ω_2 . As the electron decays back into the ground state it can end up in the other spin state. Involved phonons have to fulfill the condition $\omega_1 - \omega_2 = \pm\omega$, but compared to the Raman process they have much larger energies. The Orbach process has a strong temperature dependence, as it relies on the occupation of phonons with energies that can excite the electron into a higher energy state ($\omega_{12} \approx \Delta$). For temperatures $k_B T_0 \ll \Delta$ the behaviour leads to a contribution to the relaxation rate that can be approximated by:

$$\Gamma_{1 \text{ Orbach}} \propto \exp \left(\frac{-\Delta}{k_B T_0} \right) . \quad (15)$$

Unlike the direct process, which has a strong dependence on the magnetic field, and the Raman process that has a weak dependence, for the Orbach process the splitting of the $|\uparrow\rangle$ and $|\downarrow\rangle$ states is virtually irrelevant, as it relies only on the higher state's energy Δ and the thermal population of high energy phonons.

2.4.4 Phonon Bottleneck

The spin lattice relaxation rate of an ensemble can also be affected if the phonon temperature is different from the thermal bath temperature T_0 . If, for example, the spin population is inverted ($n_{|\downarrow\rangle} < n_{|\uparrow\rangle}$) and T_S becomes negative, every phonon mediated transition from $|\uparrow\rangle$ to $|\downarrow\rangle$, will add energy ω to the phonon band. When the spin heat capacity of the system is equal or bigger than the phonon heat capacity and/or the rate at which phonons can transfer their energy to the thermal bath is significantly smaller than the rate at which the electrons relax, the relaxation of the electron spins will heat the phonon lattice. More lattice vibrations with energies resonant to the spin transition will therefore be present in the crystal, and interact with other spins, an effect called the phonon bottleneck. This is especially relevant at very low temperature $h\nu > k_B T_0$, since most vibrational modes should be frozen out, where spins should mostly relax via the direct process, spontaneously emitting phonons. Initially, such a system would relax with a rate Γ_1 , but as more and more resonant phonons are emitted and stay in the crystal, stimulated emission can start to take place, an effect known as the phonon avalanche which causes a rapid depolarisation of the electron spins into a mixture $n_{|\downarrow\rangle} = n_{|\uparrow\rangle}$. After that, both the phonon and electron system relax towards the bath temperature T_0 at the combined rate of the two systems with the bath.

2.4.5 Spin-Spin Interaction

The spin relaxation processes mentioned so far were based on spin-phonon interactions with a uniform spin temperature within a crystal. However, it is also possible for spins to interact with other spins in the lattice by means of dipole-dipole or flip-flop interactions. If T_S is raised only locally within a crystal area, or even for a subensemble of spins, excited $|\uparrow\rangle$ spins can relax into neighbouring spins by exchanging quanta of energy $\omega\hbar$. As a result the spin polarization of the subensemble can be lost by spin diffusion. This can lead to fast depolarization as heat is transferred into "cold" spins, decreasing T_S of the observed spins, before releasing this heat into the lattice. Ideally, with infinitely narrow transition resonances, only spins with the same resonance frequency ω could partake in such an exchange, but since all linewidths are finite, even spins with different frequencies can be involved in the process, as long as the resonances have some overlap, leading to cross relaxations. Another way spin depolarization can be accelerated is by magnetic noise induced by other spins. Quantum fluctuators, such as fast flipping spins or tunnelling charges, adjacent to a spin, can interact with the electrons, destroying polarization locally at a faster rate. These processes could, like a phonon bottleneck, lead to a non exponential behaviour in the recovery of spins to equilibrium by a stretched exponential instead:

$$n_{|\uparrow\rangle}(t) \propto \exp\left(-\sqrt{t/T_1}\right) . \quad (16)$$

Spin-spin interaction type relaxations are highly reliant on the distance between two spins, being more dominant in crystals with high spin concentrations ($\sim 10^{19} \frac{1}{\text{cm}^3}$), making it hard to extract a potential single spin relaxation rate in such samples [34].

3 Experimental Implementation

This chapter will discuss and explain the experimental apparatus that was used for the investigation into the cold temperature behavior of the vanadium defect in SiC. During this thesis, the entire experiment, of which an overview schematic can be seen in fig. 6, was built together with Thomas Astner, starting with only the dilution refrigerator. The general purpose of the experiment was to conduct different kinds of optical spectroscopy measurements at low temperatures. This meant preparing the excitation laser light in a proper manner, sending it to a sample that was investigated under different conditions, collecting the emission and extracting the data from it. As can be seen in the schematic, the setup has grown quite big over the year of research, which is why the description has been split into four different sections.

3.1 Laser Path

Most of of the experimental setup revolves around the laser light used for excitation of the defects and its preparation. This section explains the path of the light that is sent to the sample and goes into the details of the components used, as well as their functionality. To make use of the advantages of telecom emitters, most of the optics are run in fibers. Two types of fibers were used, single mode optical telecom fibers (SMF28) as well as special polarization maintaining panda (PM-panda or just PM) fibers, optimized for wavelengths around 1310 nm.

3.1.1 Laser

The laser source used is a tuneable telecom O-band fiber laser (EXFO T100S-HP). It has a range from 1260 nm-1360 nm and can output from -6 dBm up to 13 dBm optical power, which corresponds to 0.25 mW-20 mW. Its narrow linewidth of around 400 kHz and its ability to sweep the wavelength mode-hop free made it a good choice for examining O-band emitters. The laser is placed on a floating optical table to reduce vibrations and is controlled remotely by the measurement software. Using a polarization maintaining fiber the laser light is routed to our fiber optics rack (fig. 6 Fiber Rack). There it is connected to a PM-isolator (ISO), to avoid any backscattering into the laser. Next, 10% are split off by a 90:10 PM-Panda splitter for monitoring purposes, whereas the majority of the light goes to the electro-optical-modulator (EOM).

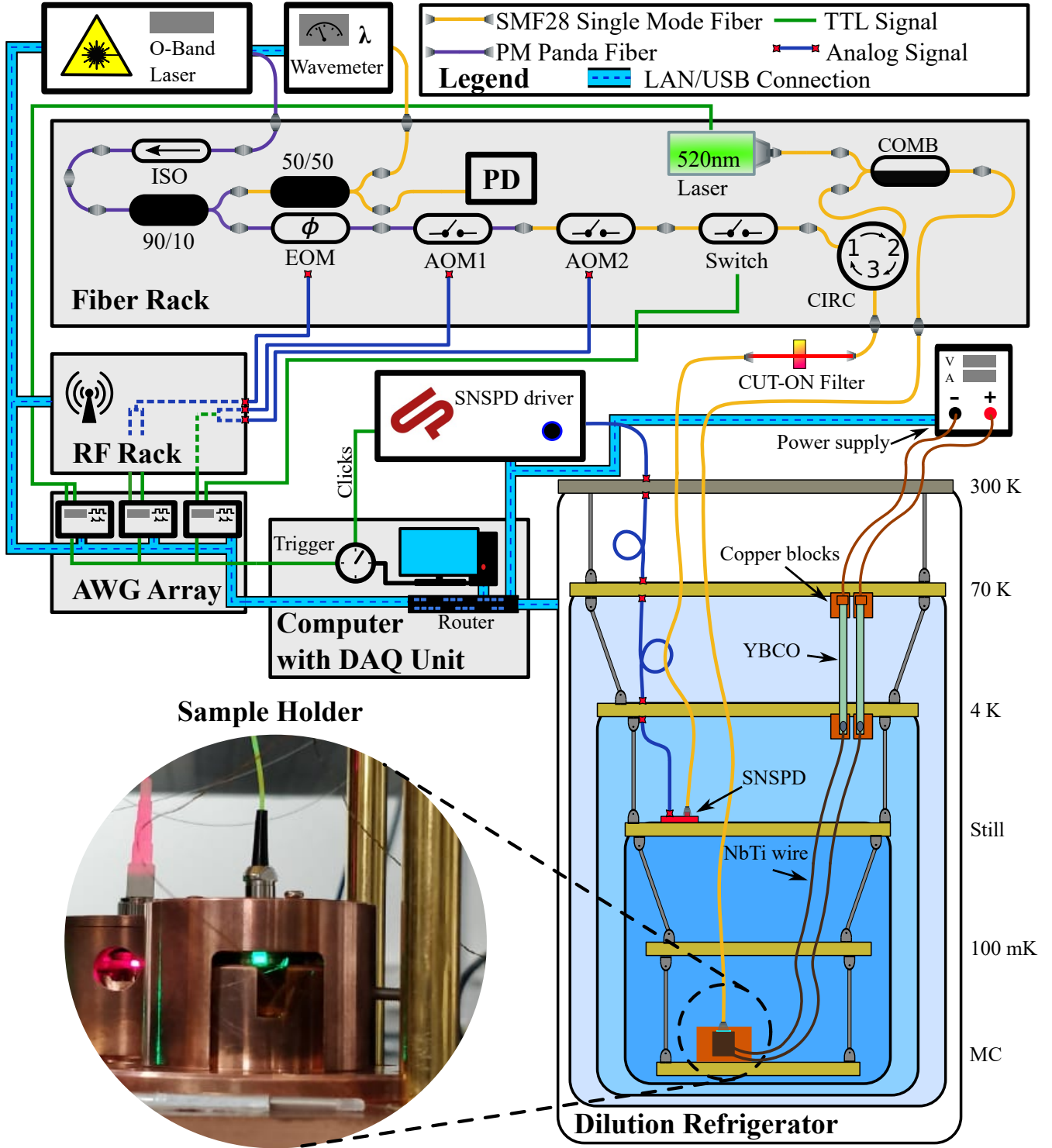


Figure 6: Schematic of the experimental setup. The source of the resonant infrared (IR) light is a tunable O band laser. Light goes through PM fiber (purple) into the **Fiber Rack** to the Isolator (**ISO**), to a **90/10** tap where 10% of the light is tapped off and monitored by a wavemeter and a photodiode (**PD**). 90% are guided to an electro optic modulator (**EOM**), used to create sidebands. Next there is a 160 MHz acoustooptic modulator (**AOM1**), followed by a 80 MHz AOM (**AOM2**), followed by a mechanical fiber switch to **switch** the light on and off. After this the light goes into the circulator (**CIRC**) port 1 and out in port 2. Green illumination (520 nm) from a laser diode is combined with the IR light in the combiner (**COMB**) and couples into a fiber leading to the sample in the sample holder on the mixing chamber plate inside the cryostat. Light emitted from the sample is picked up by the same fiber and goes in the reverse direction along the IR path through the combiner and the exits the circulator at port 3. From there all the reflected laser light is filtered out by an edge pass (**CUT-On**) filter (1300 or 1350 nm respectively) and only the emitted photons go to the **SNSPD** on the **Still** plate. A power supply is connected to the dilution refrigerator where copper wires are connected to high temperature superconductor strips on the **70K** stage leading down to the **4K** stage. From there a superconducting wire connects the current line to the solenoid in the sample holder. Arbitrary waveform generators (**AWG array**) control the green laser and the high speed switches in the **RF Rack** where RF signals are generated and switched to be sent to the **Fiber Rack**. The entire experiment is connected via LAN or USB to a computer with an integrated measurement card, which acts as a trigger and timetagger for measurement sequences.

3.1.2 Monitoring

The light used for monitoring is split by a 50/50 splitter, where half of the light goes to a photodiode (PD) for monitoring the power. The other 50% of laser light is sent to a wavemeter (HighFinesse WS6-200 IR-I), which uses a Fizeau-type interferometer to measure the wavelength with an accuracy of 120 MHz in the telecom O-band. The wavemeter made it possible to stabilize the laser for measurements that required long term wavelength stability and to reduce the jitter to less than 1 picometer (over several weeks).

3.1.3 Phase Modulation

The 90% port of the splitter enters a 5 GHz electro optical modulator (EOM) or phase modulator (Optilab PM-1310-5), specified for a wavelength of 1310 nm, which is also interfaced through a PM-panda fiber. The phase modulator uses a lithium niobate crystal to shift the phase of the light by applying a DC voltage. This is because the refractive index of the crystal depends on the local electric field, making it possible to change the time light needs to pass through such a crystal by changing the applied field. In this particular model a 3 V DC voltage corresponds

FPI Measurement of the sideband modulation

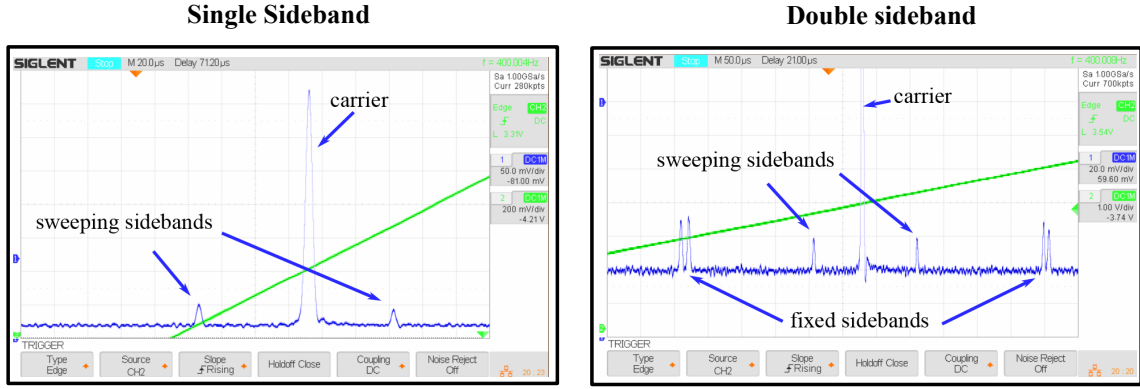


Figure 7: Measurement of the sidebands created by the **EOM** using an Fabri-Perot interferometer with a free spectral range of 1 GHz and an oscilloscope. In green the sweep voltage of the piezo driver is shown and the optical photodiode voltage is shown in blue. LEFT: Modulation of a single sideband pair. RIGHT: The scheme used with two pairs of modulated sidebands, one pair fixed and one pair sweeping. The copies of the fixed sidebands are due to the free spectral range (FSR) not being exactly 1 GHz.

to roughly a π -shift in the phase of the light. The DC phase shift was tested using an interferometer with polarization quadrature readout to ensure proper functionality of the device. An EOM can also be used with AC voltage, thus creating a sinusoidal shift to the phase of the electric field:

$$Ae^{i\omega t + i\beta \sin(\Omega t)} \approx A(e^{i\omega t} + \frac{\beta}{2}e^{i(\omega+\Omega)t} - \frac{\beta}{2}e^{i(\omega-\Omega)t}) . \quad (17)$$

This creates sidebands relative to the carrier, but shifted by the frequency $\pm\Omega$ of the AC voltage and with amplitudes proportional to the initial field amplitude A and the strength of the shift β . The amplitude of the phase shift or sidebands respectively depends on the power of the microwave (MW) input and increases as the MW power increases. Unfortunately, as the MW power increases, so does the amplitude of higher order harmonics of the sideband. In the experiment we used two tunable microwave sources (AimTTi TGR6000 and Rohde und Schwarz SMF100-A), to drive the EOM and create sidebands in a range from 10 MHz up to 5 GHz. The MW signals can be switched on and off individually using TTL signals by coaxial high isolation switches (ZASWA-2-50DR+), were then combined into one SMA cable, amplified by a broadband amplifier and then fed to the EOM voltage input. The sidebands' shapes, positions, and amplitudes were verified using a Fabry-Perot interferometer (Toptica FPI 100, FSR=1 GHz $\pm$ 1%) as can be seen in fig. 7. The phase modulator was used for the two-laser spectroscopy experiments, and no voltage was applied for other experiments, thus not modulating any phase and having no effect aside from input losses.

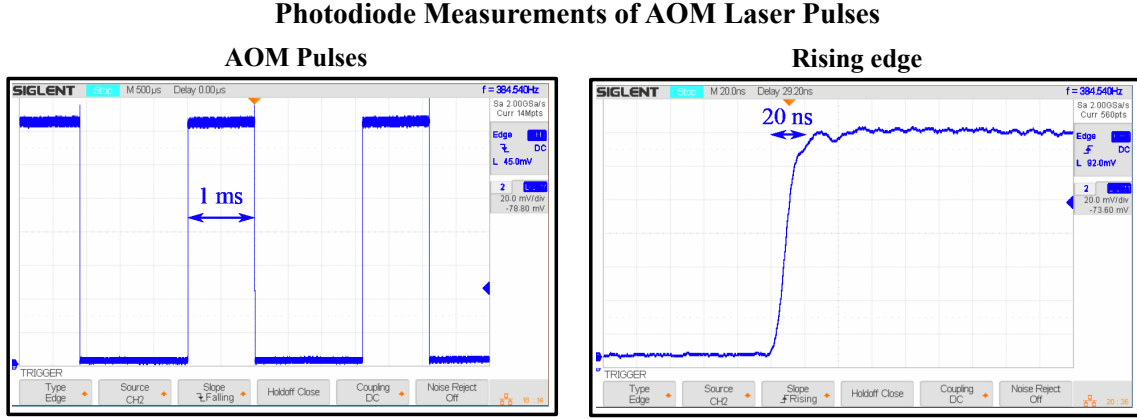


Figure 8: Measurement of the optical laser pulses created by the two AOMs using a 100 MHz photo diode and an oscilloscope. LEFT: 1 ms pulses used for the **Pump** and **Probe** pulses in the spin lattice relaxation measurements. RIGHT: Closeup of the rising edge of a pulse showing a width of less than 20 ns.

3.1.4 Pulse Creation

The creation of pulses is accomplished using two acousto-optical modulators (AOMs) in series to reduce off-state leakage, i.e. increase the suppression, from 40 dB-50 dB of one individual AOM, to 90 dB.

The first AOM (AOM1) is a custom polarization maintaining 1260 nm-1360 nm 160 MHz acousto-optical modulator, with a 10 ns rise time. The 160 MHz RF-signal is supplied by a the corresponding AOM driver and switched by a TTL signal, from a function generator (SGD2042X). It is connected to the phase modulator by a PM fiber.

Light exits AOM1 through a PM fiber and is connected into the SMF28 input of the second AOM (AOM2-Gooch and Housego fiber-Q 80 MHz). AOM2 is specified for a wavelength of 1550 nm, but by supplying the device with 99.5 MHz it works very well for wavelengths at around 1310 nm, with a rise time of less than 10 microseconds. The driving voltage is generated by another tunable MW source (ADF4351 eval board) which is connected to a coaxial high isolation switch (ZASWA-2-50DR+), which is switched by the same TTL signal. The resulting signal is further amplified and supplied to the AOM.

With the combination of the two AOMs it is possible to create very precise (rise time ≈ 20 ns) pulses, which can be seen in fig. 8, and make sure that no leakage could compromise measurements even at very long off-times (~ 1 min) between pulses. After the two AOMs the light passes through a mechanical fiber switch, that can be used to shut off the light going into the experiment.

This mechanical switch is not used in any of the measurements, as it takes several milliseconds to respond, but was very helpful for debugging purposes and it was also controlled by a TTL signal.

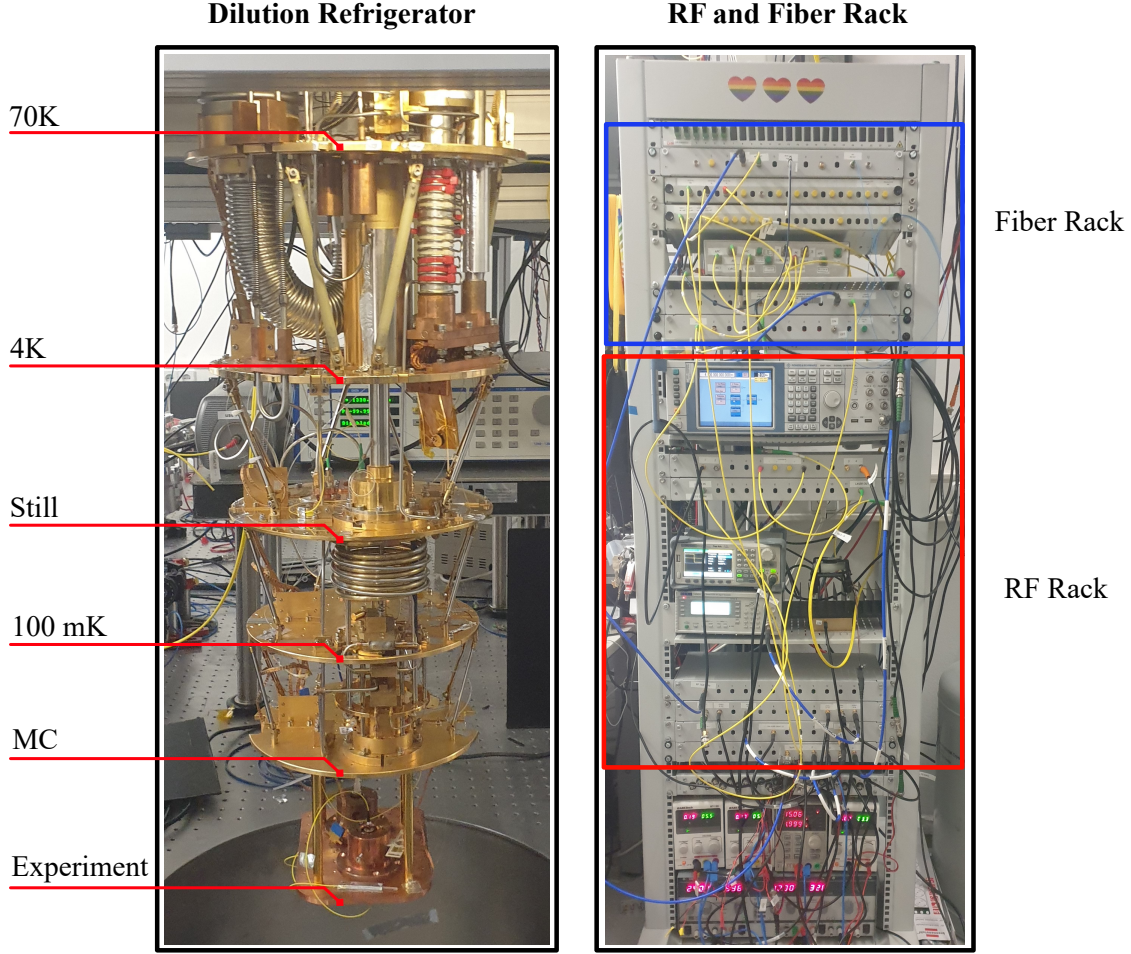


Figure 9: LEFT: Photograph of the open dilution refrigerator used for the experiment. The experiment stage is thermally coupled to the mixing chamber stage, therefore it has the same temperature and they can be considered as one stage. RIGHT: Photograph of the rack that was used for most of the optics (**Fiber Rack**) and the generation of RF signals for the AOMs and phase modulation (**RF Rack**) with laboratory power supplies used by the devices seen in the bottom.

3.1.5 Circulator and Combiner

After the series of switches, the light travels through a single mode fiber to an optical circulator (CIRC), where it is connected to port 1. The device scatters the laser light to port 2, which is then connected to the IR input port of the combiner (COMB). The combiner combines green above band gap illumination with the IR laser light and works as a filter or demultiplexer in the other direction. The combined light then travels through a single mode fiber into the cryostat.

3.1.6 Above Band Gap Illumination

For above band gap illumination of the defects green laser light was used, as the photon energy of ~ 2.4 eV is enough to excite the vanadium electron above the band gap [31].

A 520 nm laser diode in a free space optical setup was coupled into a single mode fiber for the measurements on 4H SiC.

The measurements on the 6H SiC sample were performed with a fiber coupled 520 nm laser diode, eliminating coupling losses.

Both laser diodes are driven by a function generator (SGD2042X) to supply both voltage and current. As the laser diodes have a high modulation capability, there was no need for AOMs in the path of the green laser. The laser light travels through an SMF28 fiber to the combiner, where it is merged with the infrared light from the telecom laser.

3.2 Cryogenic Setup

The defects were placed inside a Vericold dilution refrigerator (100 mK). This section describes the techniques that were used to perform experiments at such low temperatures.

3.2.1 Dilution Refrigerator

Dilution refrigerators achieve temperatures in the millikelvin range by dissolving and evaporating ^3He in and from ^4He . The first experimental implementations in 1964 [35] still relied on a bath of liquid helium, a so called wet system. Meanwhile, the technology has advanced and closed cycle cryostats work without the need of an external liquid helium supply, therefore called dry systems [36]. They instead use a two stage pulse tube cooler. The cryostat used, seen in fig. 9, consist of several stages that are thermally decoupled and shielded, as can be seen in the schematic of the experiment (fig. 6).

At the top is the **300 Kelvin** or room temperature stage, forming the outer vacuum chamber (OVC) together with the OVC shields. The chamber is vacuum-sealed and evacuated, isolating the cryostat from the room temperature atmosphere in the laboratory and has vacuum-tight ports that can be used to feed fibers and wiring into the cryostat.

Second from the top is the **70 K** stage of the cryostat, that also has a shield mounted to it, which is not vacuum tight, but shields the stages below from thermal radiation of the "hot" OVC shields. It is connected to the first stage of the two stage pulse tube (PT) cooler. This cooling system uses acoustic pulses to compress and expand helium inside a tube [37], cooling the stage down to below 70 K in the cryostat used. Connected to it by plastic rods for thermal decoupling is the liquid helium or **4 K** stage, where the cold side of the second stage of the two stage PT cooler is connected. This is critical to liquefy the ^3He ^4He mixture, the coolant that is used in dilution refrigeration, for the following stages. Another heat shield protects the following stages, which are connected to the 4 K stage and each other by stainless steel rods, again from thermal radiation.

The **still** shield is mounted to the 1 K or still stage below the 4K stage. Here the incoming helium mix is cooled to around 1 K and the returning ^3He from the stages below is distilled from the liquefied ^4He .

Below is the **100 mK** stage that is used to exchange heat between the incoming and returning helium and below is the mixing chamber or **MC** stage, where the mixing pot is mounted.

Dilution refrigeration works by condensing the mix into the mixing chamber, where due to the finite solubility of ^3He in ^4He of 6.6%, a phase boundary is created between the ^3He concentrated and the ^3He dilute phase. By pumping helium-3 from the Still pot, new ^3He molecules can dissolve in the ^4He and due to the extraction of entropy of mixing, heat is removed from the mixing chamber stage [38].

Underneath the MC stage a platform for the experiment was mounted that was heat-anchored to the MC stage and equipped with a temperature sensor, adding more space inside the cryostat and achieving temperatures of below 25 mK.

Externally, the helium cycle was driven by a turbo molecular pump that would pump the evaporated ^3He out of the refrigerator, followed by a forepump and a compressor that would either condense the helium mixture into the mixing chamber or pump it to the storage tank.

Temperature Control

The mixing chamber stage is equipped with an ohmic heater to enable temperature control. Usually the experiments were performed at around 100 mK. To observe how the defects behaved at elevated temperatures, the chamber heater was controlled with a PID loop. For temperatures below 1 K the heater was sufficient to create a stable temperature level. For temperatures of above 1 K, the flow of helium in the dilution cycle was reduced by turning off the turbo pump and also removing some of the mix from the cycle by pumping it into the tank. This artificially reduced the cooling power of the cryostat and made measurements at up to 1.9 K at the MC stage possible, while the **still** stage stayed below 1.5 K, critical for the proper functioning of the photon detector (section 3.3.2).

3.2.2 High Current

The most challenging connection was the high-current lead pair that would supply the current for our solenoid that was used for the bias magnetic field. Current was supplied to the cryostat by a 6 A power supply that was controlled by the experiment computer. Two thick copper wires were connected using a feed-through from outside the cryostat onto the 70K stage, where the wire was anchored to a heat post to prevent thermal contact between the 300K shield and the rest of the current line. Thick copper reduces heating of the wires by high currents, but also leads to a high thermal load, which is why very long wires were used. After being cooled to below 70 K, the wires were connected to strips of high temperature superconductor yttrium barium copper oxide (YBCO) strips, which become superconducting at $T < 92\text{ K}$, by clamping them together with copper blocks. The YBCO strips have a thin silver coating and deliver the current down to the 4K stage, where the strips are again heat-anchored with copper blocks. Here a niobium titanium matrix wire with

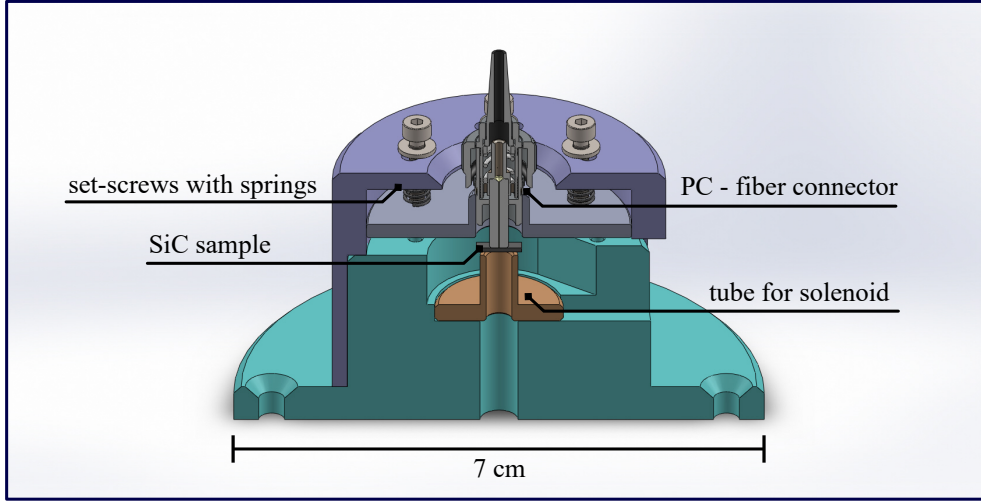
Sample Holder Cross Section

Figure 10: Cross section of the 3D model of the sample holder. Both the lid (purple) and the bottom of the holder (turquoise) were milled out of copper blocks. The copper tube (orange) was used to wind a superconducting solenoid (fig. 11), which created a bias field B_0 . A SiC sample was placed on top of the solenoid and a PC-connectorized fiber was used to illuminate the crystal and collect the emitted photons. The ferrule of the fiber was pressed onto the sample by springs.

a critical temperature of 9.2 K is soldered onto the the strips using lead tin solder, ideally removing all ohmic heating, by keeping all components below the 70 K stage superconducting. The thin wire then goes down to the mixing chamber supplying the solenoid with current.

3.2.3 Sample Holder

A silicon carbide chip was put inside a sample holder on the experiment stage underneath the mixing chamber (see fig. 10). It consists of a copper cylinder which was milled to fit the solenoid with the crystal sample on top. Above the sample holder is a lid that presses a spring loaded PC fiber connector down onto the sample. The springs can be adjusted by set-screws to ensure that the fiber ferrule is perfectly planar with the crystal surface, reducing losses. Another effect of the springs is that they press the sample onto the solenoid and the solenoid into the copper block, improving thermal coupling to the mixing chamber. The single mode fiber inside the ferrule goes outside the cryostat where it is connected to the combiner (COMB). Light coming from the laser line enters the cryostat through this fiber and illuminates the crystal underneath the ferrule. There it enters the sample and can excite the defects' optical transitions, which then decay back into the ground state by emission of photons. Some of the emitted light is then again picked up by the fiber and travels back through the same fiber, where it goes to the combiner and into the return path.

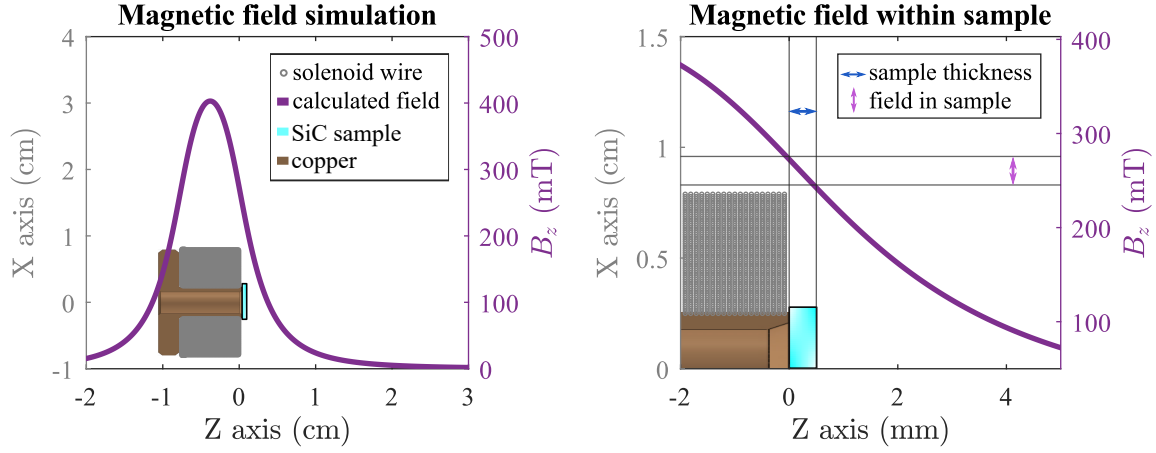


Figure 11: LEFT: Schematic showing the superconducting solenoid that is placed under the sample to create the magnetic bias field. Using the equation for the magnetic field along the z-axis for a single current loop a bias field B_z was simulated for 4000 loops spaced evenly over the solenoid cross section. RIGHT: Close up of the simulated field over the sample thickness, showing the inhomogeneity the crystal experiences over its thickness.

3.2.4 Solenoid

To create a bias magnetic field B_0 , a solenoid was built by winding a superconducting wire (niobium titanium filament in copper matrix), around a copper tube with an outer diameter of 5 mm. In total, 4000 turns of the wire were spun around the tube and glued together using a cryogenic epoxy resin with high heat conductivity (EPO-TEK T7110). This ensures good thermal coupling to the mixing chamber, both of the superconducting wire itself and of the sample that was placed on top. The resulting solenoid had a length of 7.5 mm, an inner diameter of 5 mm and an outer diameter of 16 mm.

For an estimate of the magnetic field experienced by the observed crystal sample volume an approximation was calculated. As the observed part of the sample is roughly the diameter of the fiber core (10 μm -100 μm) over the thickness of the sample (0.5 mm) and the axis of the fiber being the solenoid axis, the bias magnetic field can be estimated as the solenoid magnetic field along the z axis $B_0 \approx B_z(z)$. Using the equation for a magnetic field along the z axis of a current loop and approximating the solenoid as 4000 current loops evenly spaced within the solenoid cross section, with each loop having a radius $2.5 \text{ mm} \leq r_n \leq 8 \text{ mm}$ and position along the z axis $-7.5 \text{ mm} \leq Z_n \leq 0 \text{ mm}$ gives the following equation:

$$B_0(z) \approx B_{\text{loops}}(z) \approx \sum_{n=1}^{4000} \frac{\mu_0}{4\pi} \frac{2\pi r_n^2 I}{((Z_n + z)^2 + r_n^2)^{3/2}}. \quad (18)$$

Here, μ_0 is the magnetic vacuum permeability and I the current in the solenoid, and by summing over all the individual loops with their relative positions, the magnetic field was calculated for a current of 1 A. The simulation of the field can be seen in fig. 11 and gives a value of 276 mT/A at the bottom of the sample, decreasing to

240 mT/A at the surface of the 0.5 mm sample, where the fiber touches the crystal. Due to the approximations used, since the coil was wound manually, resulting in disordered winding ("wild winding"), and a model that expects ideal current loops, the field is probably slightly lower. Combined with the nonzero sample thickness, an uncertainty of about 10% is quite reasonable. As most of the photons collected come from the surface of the sample, closest to the optical fiber, the surface value was used in this thesis.

The maximal current that was applied to the solenoid was 2 A, corresponding to a range from 0 to 480 mT. Higher fields might be possible, but were not realized, as testing the limits of such a superconducting system, would likely be a destructive measurement.

3.2.5 Samples

All measurements were performed with commercially available semi-insulating single crystal SiC material, purchased from the company II-VI. Square 5 mm x 5 mm chips were cut from wafers with a diameter of 100 mm and an average thickness of 496 μm . In our experiments we use the polytypes 4H- and 6H-SiC with a vanadium concentration of $1 \cdot 10^{17} \text{cm}^{-3}$. Other impurities like boron and nitrogen have a concentration of $1 \cdot 10^{16} \text{cm}^{-3}$ and aluminium below $1 \cdot 10^{16} \text{cm}^{-3}$. This gives an average distance between two vanadium atoms in the lattice of $\approx 215 \text{ \AA}$ and roughly 200 $\text{\AA}$ between any two dopants in the lattice. The c-axis is orthogonal to the chip square surface and parallel to the fiber core used for measurement and the applied bias field B_0 . A visualization of how the samples were illuminated can be seen in fig. 12.

3.3 Return Path

The light coming out of the cryostat from the sample contains mainly reflected excitation laser light. To detect the luminescence of the actual defects, this reflected light needs to be filtered out. This section describes the path of photons that were emitted from the crystal sample, collected by the fiber, and guided to the photon detectors.

3.3.1 Filter Wheel

The light emitted from the defects is collected with the same fiber as the excitation light. It then travels back to the combiner, where the green light is filtered out, and then goes back into port 2 of the circulator, exiting through port 3. Port 3 is connected to a free space filter. It consists of two fiber couplers, two mirrors and a filter wheel along the path. To enable remote capability, the filter wheel was modified to be rotated by a servo motor operated by a micro controller. The filter wheel can be used to block all light, pass through everything (for debugging and coupling) and also had two types of edgepass (**CUTON**) filters, of which two each were placed in series to improve extinction of the reflected laser light. The filters had cut on wavelengths of 1300 nm and 1350 nm, to cut off the resonant excitation light of either the α - or β -defects, so that only the photons from the phonon sideband

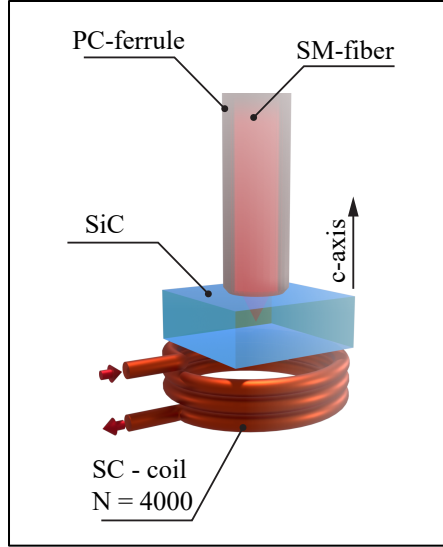


Figure 12: Illustration of the photoluminescence excitation and collection method.

A fiber ferule with a single mode fiber core is pressed onto the silicon carbide sample investigated. Laser light exits the fiber and excites defects in the crystal. Some of photons emitted, as the defects decay, are collected by the same fiber and are lead to the fiber rack for analysis. Underneath the crystal is a superconducting solenoid (SC-coil) with 4000 turns, that creates the magnetic bias field B_0 .

would pass through. After the filter the light is coupled back into a fiber, guiding it back into the cryostat to the single photon detectors.

3.3.2 SNSPD

For the counting of photons superconducting nano wire single photon detectors (SNSPD) from SingleQuantum were used. Instead of using a complete closed cycle system (consisting of a cryostat with integrated detectors with optical and readout ports and the SNSPD-driver) only individual detectors and the SNSPD-driver were purchased. The detector was mounted on the 1 Kelvin Stage (**still**) of the dilution refrigerator and the fiber transporting the photons collected from the sample was connected to it. The electrical bias and voltage readout port was connected to the driver, outside of the cryostat.

Such a detector works by applying a bias current, close to the critical current, to a superconducting strip in a meander shape. When photons exit from the fiber, they have a high probability of hitting the nanostrip or nanowire, thus breaking superconductivity. The strip turns normal conductive, which creates a voltage spike across the wire [39].

This voltage can be measured, creating a "click" signaling a recorded photon, that is then read out, either by using the TTL output and connecting it to the counter on the NI data acquisition card or the internal counter on the SNSPD driver. During all measurements the detector stayed in a temperature range between 800 mK and 1.2 K, which is far below the specified operating temperature of 2 K. To maintain a

low dark count rate (50 Hz-500 Hz), the bias current was adjusted in a range between 14 μ A and 18.5 μ A, as the detector is sensitive to both temperature and bias current. The detector had a quantum efficiency of 0.5, according to the manufacturer.

3.4 Control and Data Acquisition

As work on this experiment started in early autumn 2020 with the second wave of the CoV-19 pandemic approaching Austria, it was imperative to construct the setup in a manner that would allow for the highest possible degree of remote operation, because of both limited access to laboratories and safety concerns. To achieve this, a local areal network (LAN) was set up that connected all devices, ranging from power supplies to the optical filter wheel, to the central measurement computer that could then be accessed through the safe homeoffice environment. All parameters of a given measurement were controllable in a LabView graphical user interface, which would then pass commands to all the components. For the readout of the collected photons two approaches were utilized. Measurements with low time sensitivity ($\approx$ 10 ms) were conducted by reading out the internal counter of the SNSPD driver box through the network. For the electron spin lifetime measurements, the counts were recorded using the DAQ card of the experiment computer.

3.4.1 DAQ Card

The experiment computer was equipped with a National Instruments data acquisition card (PCIe-6363) allowing the computer to read and write digital and analog voltage signals through the connected breakout box (BNC-2090A). It also has counters with a resolution of up to 10 ns given by the internal 100 MHz clock, as well as a 10 MHz clock that could be used independently.

Analogue ports were used to monitor the laser power on the photo diode. A counter channel was connected to the SNSPD driver TTL output port, hence recording every click on the detector with a resolution of up to 10 ns, acting as a time-tagger. To synchronize the entire experiment for the jitter-sensitive spin lattice relaxation measurements, the 10 MHz clock was used to output one digital TTL signal as a trigger that would start a measurement sequence. This trigger signal was then simultaneously fed directly to the internal 10 ns counter, to start the collection window for incoming photons, and to the AWG array that would perform the given sequence.

3.4.2 AWG Array

For devices that required time sensitive digital communication, three arbitrary waveform generators (Siglent SDG2021X) with a total of 6 programmable output channels with nanosecond resolution were used, also referred to as AWG array. All three were connected to the same measurement trigger as the 10 ns counter, to synchronize each measurement sequence. The sequences could be programmed arbitrarily using the graphic user interface through the network.

Two channels were used in parallel to supply enough current to act as a laser driver for the green laser diode. Changing the voltage of the signal made it possible to control the laser power of the green laser. One channel would switch both acousto optic

modulators (AOM1 and AOM2) and another one was used to control the mechanical fiber switch. Finally, the remaining two ports would turn the two sidebands of the EOM on and off individually to perform different two laser spectroscopy schemes.

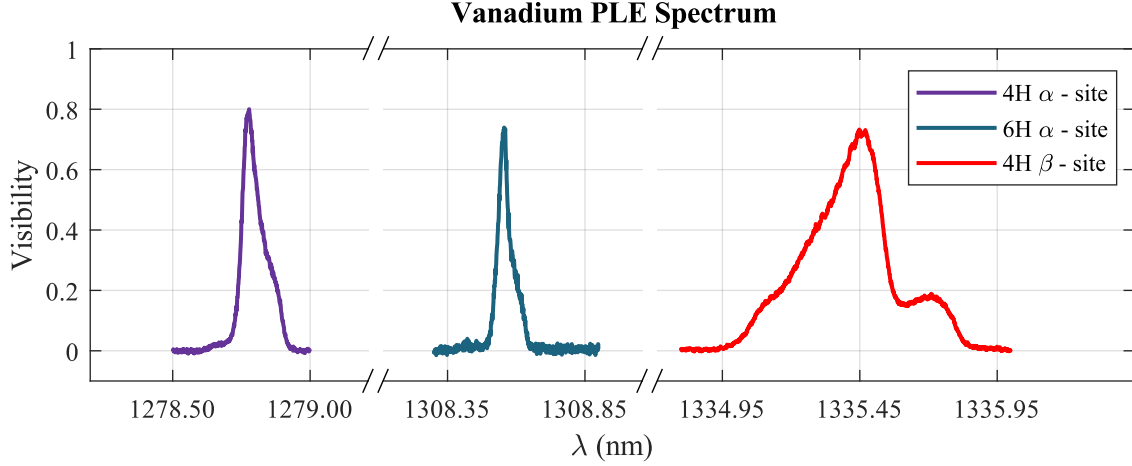


Figure 13: Photoluminescence emission comparison of the three measured defects. The traces show the maximal visibility of the defect in the experiment. The two α sites have a narrow resonance, compared to the β site, with 4H- α being the most prominent defect observed.

4 Measurements

The experiments were performed on three types of vanadium defects in silicon carbide. Two of the defects were measured in a SiC sample of the 4H polytype, called 4H- α and 4H- β , and one in a 6H SiC sample, namely the 6H- α defect. Both samples had a high vanadium concentration, hence all measurements are ensemble measurements. Four different types of measurement were conducted on each defect allowing for comparison of different potential vanadium qubits, all at ultra cold temperatures (~ 100 mK).

First, resonant photoluminescence emission spectroscopy (PLE) was performed under different conditions. Next, the charge stability was examined by means of resonant bleaching. The third type of measurement investigated the spin lattice relaxation or electron spin lifetime of the defect, by means of pump probe recovery measurements. Finally, two-laser spectroscopy was performed on the defects, making it possible to retrieve detailed information about the electron level structure.

All measurements were taken with both illumination and collection of light parallel to the crystal c-axis. In measurements for which a magnetic field was applied, it was also along the crystal axis.

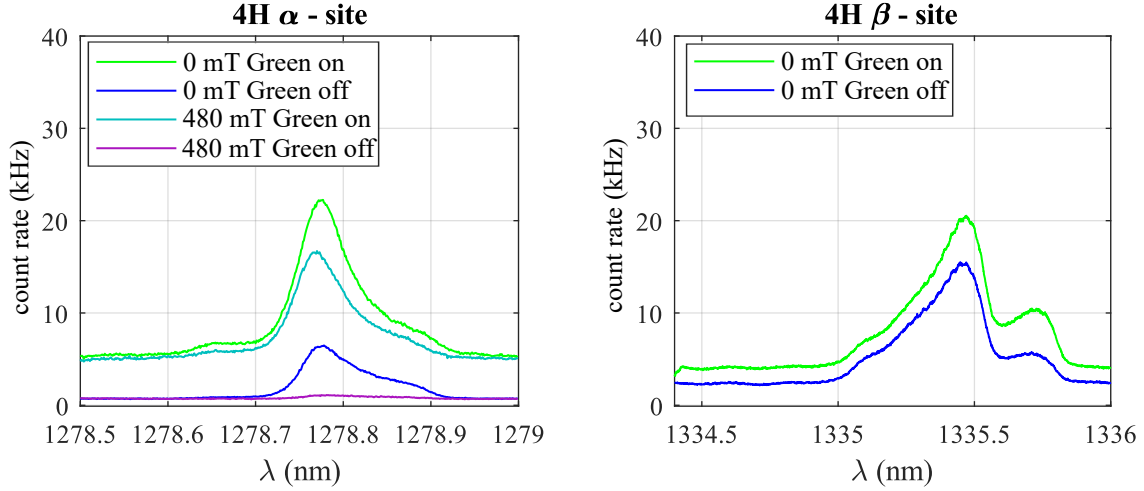


Figure 14: LEFT: The recorded spectrum of the 4H- α defect GS1-ES1 transition is shown. The resonance signal is recorded at zero magnetic field and at 480 mT, each with above band gap green laser illumination added and without. RIGHT: The spectrum of the GS1-ES1 and the GS2-ES1 transitions in the 4H- β defect are displayed.

4.1 Resonant Photoluminescence Spectroscopy

To identify the optical transitions of the defects, resonant photoluminescence emission (PLE) spectroscopy was performed by sweeping the laser over the zero phonon line, while collecting the photons emitted in the phonon sideband. The measured wavelengths can have an offset of several tens of picometers, as the excitation laser had a drift in its accuracy but would maintain high precision over several sweeps. In fig. 13 the PLE of the three defects is shown. As the absolute count rate depends strongly on the filter wavelength and other external factors, the visibility V of the resonance is displayed, allowing for a better comparison:

$$V = \frac{S - B}{S} . \quad (19)$$

This value compares the resonance signal S , to the off resonant background B .

4.1.1 4H Alpha

The 4H- α defect's GS1-ES1 zero phonon line is at the lowest wavelength of the defects with a peak at 1278.78 nm, as can be seen in fig. 14. Without magnetic field, a small shoulder can be seen at the lower wavelength end that is attributed to ^{13}C isotope shifts, but the main signal spans from 1278.3 nm to 1278.9 nm, with a prominent shoulder towards longer wavelengths, which is thought to be due to silicon isotope shifts [18]. Adding above band gap continuous wave (CW) illumination to the sweep strongly enhances the absolute signal in the resonant PLE, but also raises the background significantly. The enhancement might be due to charges that are being promoted to the neutral state vanadium V^0 for short times in unstable sites during the sweep and their contribution to the emission. The raised background

can be attributed to leakage in the optical setup and to the fact that the green light always excites a wide range of defects in the crystal, whereas resonant IR excitation does not.

Next, the spectrum was recorded again without green excitation, but with an applied field of $B_0 \approx 480$ mT, leading to an almost total extinction of the fluorescence. This effect can be attributed to the g -factors of the defects ground and excited states, which separate the optical transitions for each individual defect's electron spin $|\uparrow\rangle$ and $|\downarrow\rangle$ states, as explained in section 2.2, and the stepped sweep mode of the measurement. When the laser sets a new wavelength it polarizes each, now optically separated, electron spin state into the other state, where it is no longer resonant with the laser, thus not contributing to the fluorescence. It is an analogue process to that discussed in more detail in section 4.3, fig. 19, where the pump pulse depleats resonant spin populations. The fact that this darkening effect is so strong hints at a long electron spin lifetime. Using the signal suppression observed between the spectrum with and without magnetic field gives a lower bound for the spin population reduction of $93.5 \pm 0.5\%$. Finally, the measurement was repeated at high field but with added green light. Under this condition, the defect spectrum was visible again, suggesting that above band gap illumination not only promotes unstable charges, but also mixes the spins of the more stable vanadium defects, increasing the spin temperature.

No transitions involving the GS2 or the ES2 could be seen in our measurements. The absence of population in the second ground state GS2 was to be expected from previous measurements, which observed a strong depopulation at around 4 K compared to 15 K [18]. This implies that at around 100 mK the higher energy ground state is completely frozen out. Absence of fluorescence involving the ES2, was also not surprising, as the GS1-ES2 transitions are not expected to absorb or emit light along to the c -axis [40].

4.1.2 4H Beta

The vanadium defect with the lowest optical transition energy measured in this thesis belongs to the beta defect in the 4H Polytype. Its resonance is visibly wider than the 4H- α site with a width of almost 1 nm and stretches from 1335.0 nm to 1335.8 nm. With no magnetic field applied the optical resonance, which can be seen in fig. 14 RIGHT, consist of two wide overlapping transitions: GS1-ES1 and GS2-ES1. When comparing the spectra with and without green above band gap illumination, a clear increase in the GS2 population with the green laser on can be observed. This can be either due to the higher local temperature in the sample, while the green laser is illuminating it, or due to the capturing of promoted charges, that drop into the GS2 and did not decay into the GS1. A boost in total fluorescence was observed with the green laser on, but it was not as significant as in the 4H- α defect. The measurements were taken at around 100 mK and the GS2 was never fully depopulated, which is attributed to the low splitting energy separating GS1-GS2.

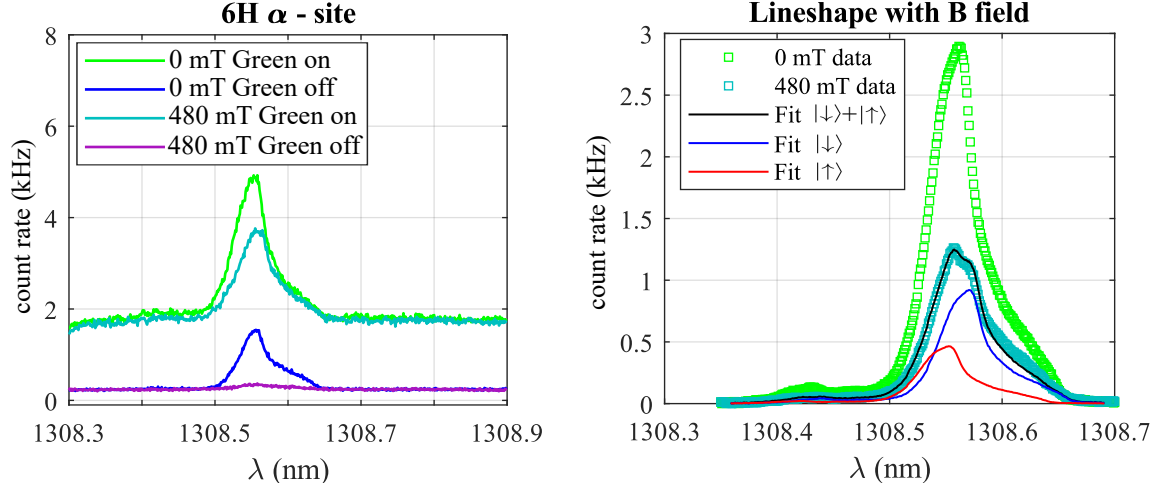


Figure 15: LEFT: Spectroscopy results of the 6H- α GS1-ES1 transition at around 100 mK are shown. The resonance signal is recorded at zero magnetic field and at 480 mT, each with above band gap green laser illumination added and without. RIGHT: Averaged spectroscopy signal of the transitions with the green laser added, integrated for 6-7 hours and tracked with the wavemeter. Using the zero field measurement as the baseline the 480 mT data was fitted with as a sum of the two resulting spin $|\uparrow\rangle$ and $|\downarrow\rangle$ populations contributing to the signal.

4.1.3 6H Alpha

The third defect measured was in a crystal sample of the 6H-SiC polytype, the 6H- α . Its optical resonance (GS1 to ES1) is at 1308.56 nm and it has a very similar appearance as the 4H- α site (see fig. 13). On the lower wavelength side there is again a ^{13}C isotope shoulder at around 1308.4 nm. The brighter part of the resonance starts at 1308.5 nm and goes until roughly 1308.7 nm with a prominent shoulder towards the longer wavelengths. Adding above band gap illumination gives a boost in fluorescence at zero field like in the alpha defect in 4H. Similarly, applying around 480 mT during the measurement makes the defect go dark, if no green laser is added (see fig. 15), which for this defect corresponds to a spin population reduction of $89 \pm 1\%$. With the green laser the fluorescence comes back, also implying that this can be used to mix the electron spins in the defect ensemble.

To see if the optical splitting could be resolved in the optical spectrum at 480 mT, another more precise spectroscopy measurement was performed on this site. First, it was important to reduce the noise in the signal, which is why the signal was integrated over several hours, compared to the several minutes on the other measurements. To account for the drift of the laser of this period, the wavelength was tracked and recorded for each step in the wavelength using the wavemeter. The wavelength vector was then binned in 1 pm steps and the recorded count rate was mapped to the resulting wavelength vector and averaged for each picometer. Using this measurement the spectrum was recorded at zero field and at 480 mT, both with above band gap illumination added. In the resulting signal (see fig. 15) a structure was observed on the peak at 480 mT.

By interpolating the signal at zero field and shifting the resulting function symmetrically with two separate amplitudes, a close match of the observed resonance with the applied magnetic was achieved.

Using the splitting extracted from the fit of about 3.25 GHz and using eq. (6) it was possible to calculate the difference between the ground and excited state g-factors. The resulting fit gives $\Delta g = 0.483 \pm 0.009$, which is slightly lower compared to the literature value [18], but within the margin of error of the fit. Considering the uncertainty of B_0 , the margin of error is estimated at 10%. The amplitudes of the two spin subpopulation ($\frac{N_{|\uparrow\rangle}}{N_{|\downarrow\rangle}} \approx 0.5$) can be used with eq. (7) to get an estimate for the spin temperature during the spectroscopy measurement. This fit would result in a spin temperature $T_S \approx 227$ mK, which is significantly higher than the temperature at which the measurement was recorded ($T_0 \approx 169$ mK), implying that even the low power above band gap illumination used here does raise T_S .

One major factor that is omitted by this analysis is that the field experienced by the probed ensemble is not homogeneous, as described in section 3.2.4, and taking the spacial distribution of spins into account could yield an improved match of the observed spectrum. Unfortunately, this distribution not exactly known and guessing it could lead to flawed results. Conversely, by developing a more sophisticated fitting algorithm it might be possible extract the spacial distribution of the observed defects along the z axis. This approach could prove very powerful for investigations into the optical coupling and collection volume of this, and similar experiments.

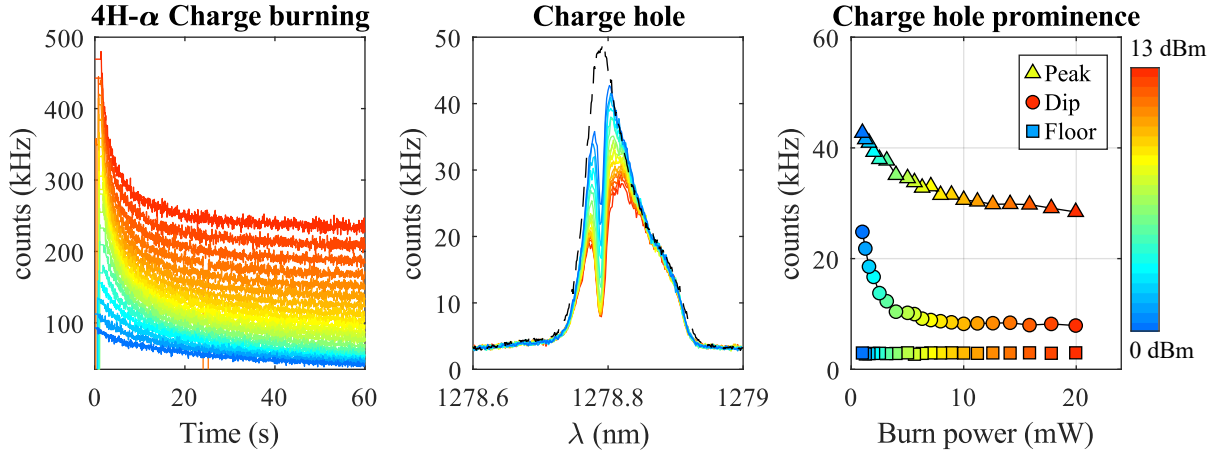


Figure 16: Result of the charge burning measurements on the 4H- α site. The powers are displayed in different colors from blue at 0 dBm to red at 13 dBm laser output power. LEFT: Bleaching of the recorded fluorescence over the 60 seconds of the burn pulse. MIDDLE: PLE of the defect after the burning using a quick sweep with -3 dBm to avoid ionization. The black dashed line shows an unburned spectrum of the defect for comparison. RIGHT: The recorded maximum (peak), minimum (floor), and the value of counts recorded at the wavelength of the charge hole (dip).

4.2 Charge State

As can be seen in the results of the PLE measurements, adding a green laser to stabilize charges gave a significant boost to the fluorescence. Further it was observed that, when the laser would be turned on for a while on the α and β resonances in 4H the signal would bleach over time, while no bleaching was observed with the green laser also on. The next sections will discuss measurements that were taken to further investigate the stability of the charge state of the defects. Measurements consisted of two parts:

First, the infrared laser was locked with the wavemeter to the peak of the defects resonance, then switched on to "burn" the defect for 60 seconds and the resulting decay of the fluorescence was recorded for different laser powers (1 mW-20 mW). This would be used to measure the rate of photobleaching due to ionization of resonant defects. After this burning, a low power (0.5 mW) PLE measurement was performed to see if there was a change to the lineshape. The powers in the following sections are given as a function of the laser output power, not the actual power at the sample. After each measurement the sample was illuminated with the green laser to restore the original charge distribution.

4.2.1 4H Alpha

The result of the charge burn measurements performed on the 4H- α site can be seen in Figure 16. For low laser powers, a steady decay could be observed over the entire 60 seconds of the measurement. As the power is increased, the rate of bleaching increases, where most of the signal decays over the first 10 seconds, reaching almost

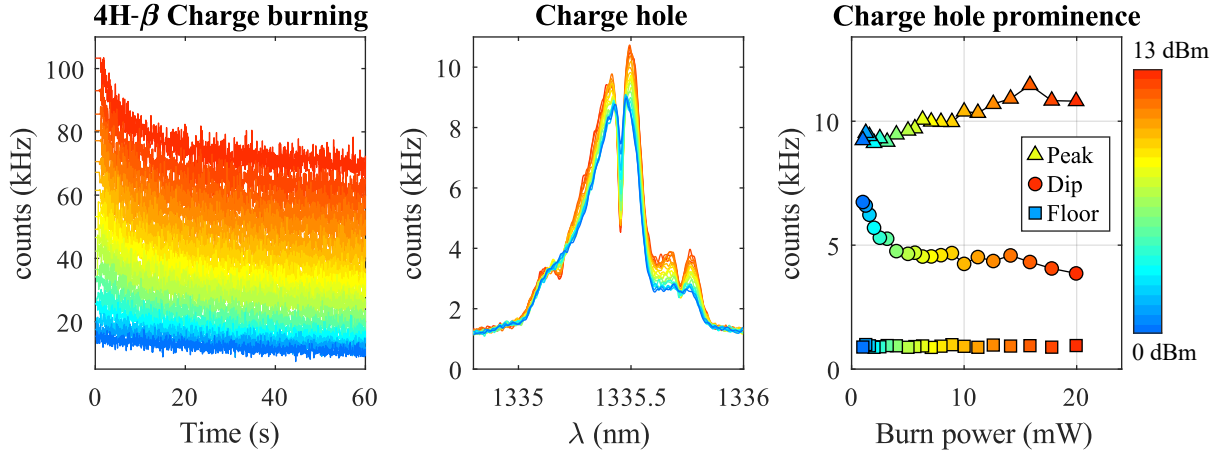


Figure 17: Result of the charge burning measurements on the $4H\text{-}\beta$ site. The powers are displayed in different colors from blue at 0 dBm to red at 13 dBm laser output power. LEFT: Bleaching of the recorded fluorescence over the 60 seconds of the burn pulse. MIDDLE: PLE of the defect after the burning using a quick sweep with -3 dBm to avoid ionization. RIGHT: The recorded maximum (peak), minimum (floor), and the value of counts recorded at the wavelength of the charge hole (dip).

a steady state at the 60 second mark at the end of the burn. Due to the geometry of our optical excitation and collection process, which creates a very inhomogeneous illumination, it is hard to draw conclusions on the individual defect charge behavior from this decay. Looking at the spectrum a clear dip is visible in the PLE at the wavelength, where the defect was burned, with a FWHM of around 2.1 GHz. Increasing the power created a deeper charge hole in the spectrum, saturating at around 10 mW of laser power. With higher powers, the maximum of the signal was also decreased. This may be due to the narrow width of the ensemble resonance and power broadened bleaching, causing defects that are not fully resonant to also be ionized partially. The off resonant background in the different spectroscopy measurement stayed constant. Even though the contrast between the bleached and the unbleached parts of the signal was significant, it was not possible to fully extinguish it and reach the background floor. Using the green laser and illuminating the sample for one second fully restored the spectrum to the "unburned" state, which is displayed in the sections above. To see how the charges would behave over time, a hole was burned again, but this time only with the highest, most efficient, laser power. Now however, the PLE measurement was not performed right after the burning pulse, but with a delay that was varied from 10 seconds up to 2 hours. As can be seen in fig. 18 both the amplitude of the PLE signals' maximum and the depth of the dip in the resonance stayed near constant over the observed period. From this it is possible to conclude that the charge states are stable and do not redistribute at this low temperatures (≈ 100 mK) without external influence, such as resonant or above band gap illumination. This measurement showed that the charge state can be bleached over timescales of less than 20 seconds, but the resulting hole persists over several hours. Such behaviour makes it possible to engineer the lineshape of

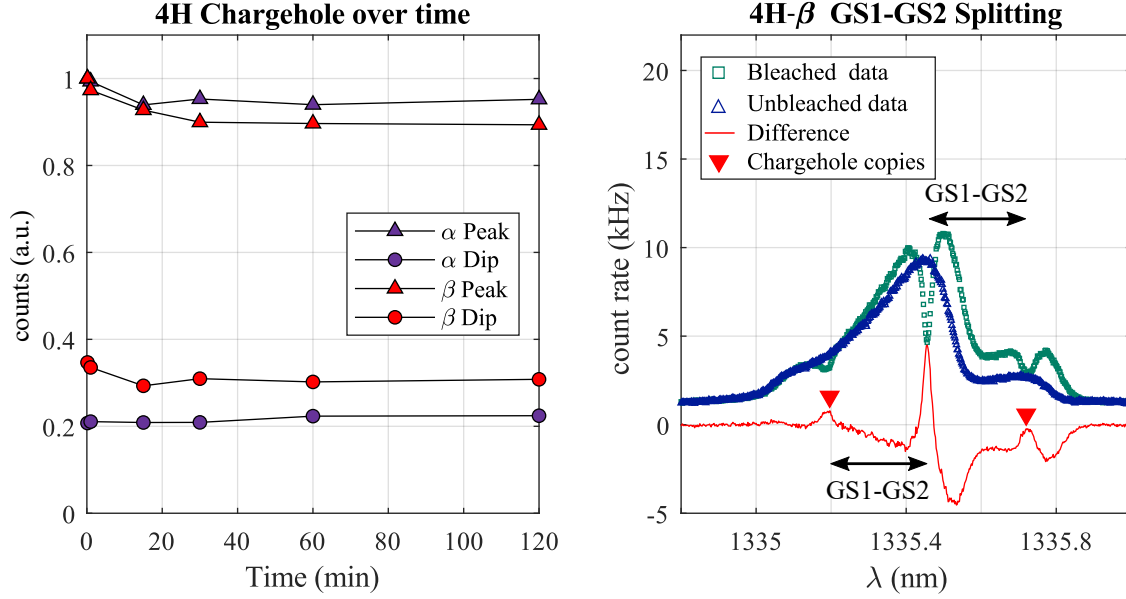


Figure 18: LEFT: Dips and peaks of the chargeburned PLE spectra for the 4H- α and 4H- β defects measured with varied delays from 10 seconds to 2 hours. RIGHT: Comparison of the unbleached to the bleached PLE spectrum of 4H- β at the highest burn power. A clear boost to the GS2 population can be seen. The difference between the hole and the copies is portrayed showing the relative splitting between GS1 and GS2.

the 4H- α defect with long term stability.

4.2.2 4H Beta

4H- β was also susceptible to photobleaching, which can be seen in fig. 17, but with several differences compared to the 4H- α defect. At low powers the bleaching was far less visible, both in the recorded fluorescence during the bleaching process, as well as in the resulting PLE spectrum. Similarly to the other defect, the prominence of the charge hole, as well as the bleaching efficiency, increased with higher power. However it did not reach saturation within 60 seconds of burning at the maximum power the laser could deliver (13 dBm) and also not reaching nearly as much contrast, as the alpha site. Before and after each measurement it was again possible to restore the initial state by applying green laser light for one second. Another interesting effect was an apparent boost to the maximum of fluorescence in the unburned area of the spectrum at high laser powers. This is probably due to the increase in population in the GS2 as the sample is locally heated or due to electrons that decayed into the second ground state. Looking at the shoulder towards the lower wavelengths, there is no change of the fluorescence recorded, but towards longer wavelengths, where the GS2-ES1 transition is located, a strong increase is observed at high laser powers, corresponding to stronger local heating. The increased GS2 population then decays, returning back to the normal levels, as the sample thermalizes after the burn over a couple of minutes. In fig. 18 it can be seen that the holes created do also last over two hours, but generally have significantly less contrast, compared to 4H- α .

Finally, the PLE signal showed, that even though only one charge hole was burned only at the center wavelength of 1335.45 nm, three dips could be observed in the spectrum, shifted symmetrically from the main hole. At the burned wavelength mostly GS1-ES1, but also some GS2-ES1 transitions, are resonant with the laser. By ionizing GS1 electrons, a copy of the charge hole was created in the GS2, and vice versa, implying a very high dynamic between the two ground states. Using this dynamic and resulting changes to the spectrum, as can be seen in fig. 18, it was possible to measure the splitting of the two ground states in the 4H- β defect: GS1-GS2 $\approx$ 44(3) GHz. Within the margin of error, given by half the linewidth of the charge hole copies, this value is in agreement with previous measurements [18].

4.2.3 6H Alpha

In the 6H- α defect, no bleaching was observed. It is not possible to say from these measurements whether the charges are more stable in this site, or if ionization does happen, but there are more electrons available in the crystal that quickly compensate ionization. Another possibility is that other vanadium charge states are also excited by near infrared illumination, which would all lead to a lack of observed bleaching. However, the reduced electron mobility of this SiC polytype, compared to 4H, is likely the cause of the observed behavior, indicating more stable V^0 defects [16]. The observed behaviour might also be dependent on the particular sample investigated, and its dopant composition, as spectral hole burning has previously been observed in 6H SiC vanadium defects [33].

4.3 Electron Spin Lifetime

The main interest of this thesis was to evaluate whether vanadium defects in silicon carbide would make for good qubits. One critical limiting factor of a solid state electron spin qubit is the electron spin lifetime T_1 or spin lattice relaxation rate Γ_1 conversely, which is therefore an important unknown quantity for these defects.

To lift the degeneracy of the spin $|\downarrow\rangle$ and $|\uparrow\rangle$ states in the vanadium $S = \frac{1}{2}$ system, a magnetic field of $B_0 = 480$ mT was applied to the sample. As was demonstrated in the PLE measurements, the difference between the excited and ground state g-factors, now allows for pumping the electron into the opposite spin state, which is then out of resonance with the driving infrared laser (figs. 14 and 15). Another mechanism shown in the previous measurements is that above band gap illumination can create a mixture of the electron spins. Using those two mechanisms the following pump probe recovery scheme was introduced to measure the lifetime of the electron, consisting of 4 steps, which are illustrated in fig. 19.

First, the system was initialized with a pulse of green laser light (**Init**). This creates a mixture of spin states $|\downarrow\rangle$ and $|\uparrow\rangle$ in the ensemble. As the above band gap illumination pumps all the electrons in the sample, this mixing should apply to the entire resonance equally. The green laser pulse used in the experiments was 500 μ s long and the power was tuned to maximize the signal of the following resonant pulse, without going far beyond the saturation, as this would contribute to unnecessary and unwanted heating of the sample.

The next step in the sequence was the resonant infrared **Pump** pulse, pumping the spins into the other state. At the applied magnetic field, even though individual spins should be sufficiently optically separated, the ensemble as a whole is not optically separated and still has a big overlap, as seen in fig. 15. This results in two effective subpopulations of the defect ensemble that contribute to fluorescence from the pump pulse. One population (Pop 1) is resonant with the laser for electrons in the $|\uparrow\rangle$ state, which are driven by the laser into the excited state and can decay into the $|\downarrow\rangle$ electron ground state, where they will be trapped. By pumping for several optical cycles this leads to a polarization of Pop 1 into the $|\downarrow\rangle$ state. The second population (Pop 2) is resonant with the laser at $|\downarrow\rangle$ and will be similarly pumped into the $|\uparrow\rangle$ electron ground state. As illustrated in figure 19 this leads to a decay in recorded fluorescence during the **Pump** pulse as the two populations electrons are being oppositely polarized.

After polarizing the system, a variable delay τ is added after the first infrared pulse with no light added to the systems and no transitions being driven. During this **Wait** period the electrons relax back into the depleted states, until the spin temperature reaches the temperature of the sample ($T_S \rightarrow T_0$). At low temperatures this thermal relaxation itself will lead to a significant polarization of both Pop 1 and Pop 2 into the $|\downarrow\rangle$ ground state level, as the phonon bath does not have enough energy to excite the electrons into the $|\uparrow\rangle$ state, creating a polarization given by equation 8. The resulting state is therefore not necessarily the initial state created by the **Init** pulse. Assuming that the state before pumping was a fully mixed state and the resulting state is polarized equally for both populations, this procedure will lead to the same number of electrons in the bandwidth that is resonant with the IR laser as long as the populations Pop 1 and Pop 2 are equal at that frequency.

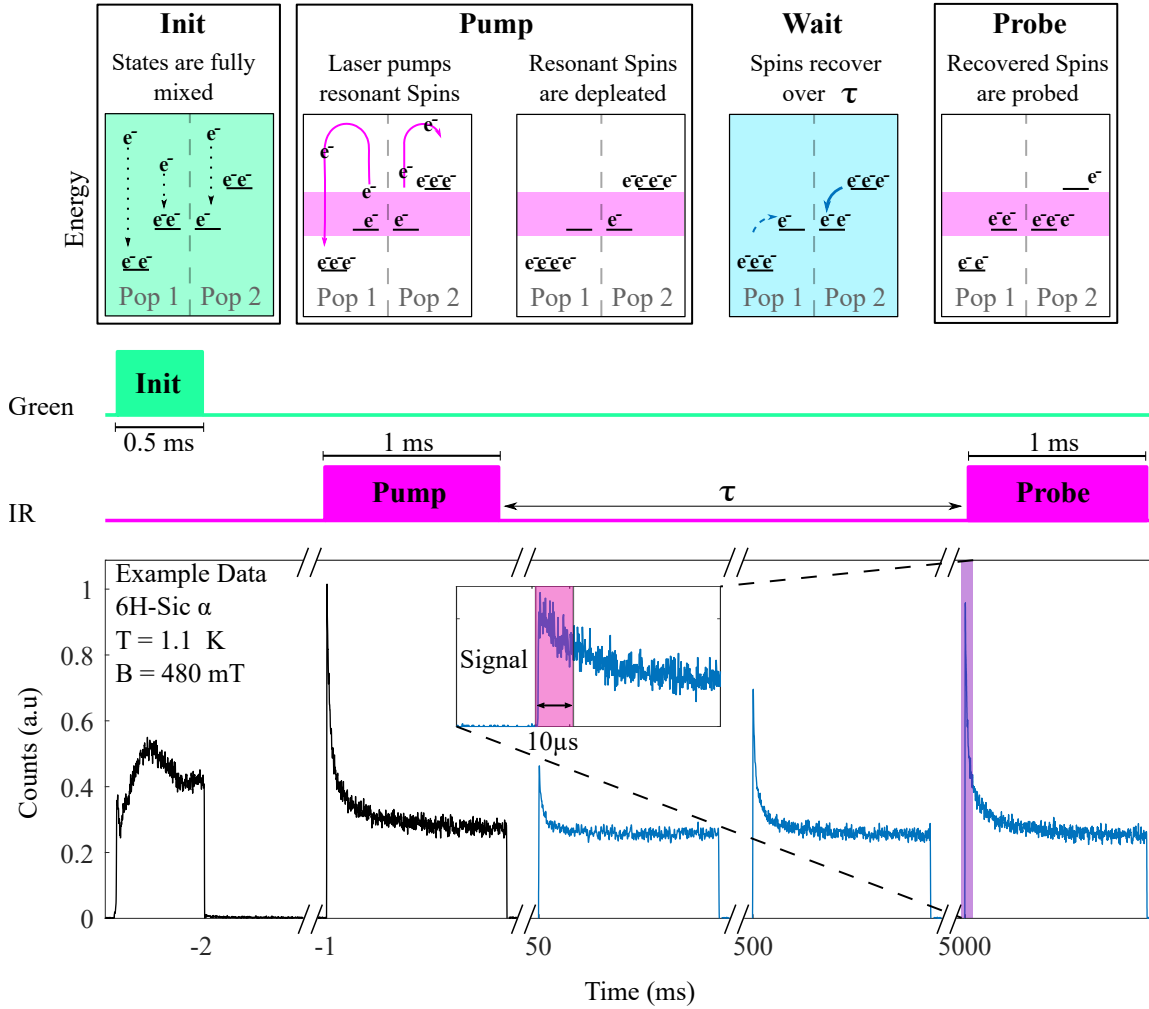


Figure 19: Schematic of the Pump-Probe-Recovery sequence used to measure the ground state electron spin lifetime. In the top an illustration of the electron dynamics is shown corresponding to the pulse sequence in the middle. The ensemble is first initialized into a mixed state by a green laser pulse, then polarized by a pump pulse. Over a variable delay time τ , spins relax and the population that relaxed is then probed to identify the spin lattice relaxation rate. The bottom panel shows an example of the normalized fluorescence signals for three different wait times taken at 1.1 K on a 6H- α defect. In the last pulse displayed, the 10 μ s integration window of the measurements is further shown.

If there were an imbalance between the two populations, this would lead to either more or less electrons in the pumped band as the system reaches equilibrium at low temperatures. To get information on the relaxation rate, it is irrelevant whether a recovery from the up to the down state or vice versa is measured, as both spin flipping transitions should act symmetrically, being dominated by the phonon bath. By varying the delay τ , the number of electrons that relaxed (flipped their spin) in this period is varied, leading to a recovery of population that is resonant with the laser over time.

The final part of the sequence is the **Probe** pulse. It has the same length and power as the **Pump** pulse and performs the same task, exciting the Pop 1 electrons that are in the $|\uparrow\rangle$ state as well as those from Pop 2 that are in the $|\downarrow\rangle$ state. Fluorescence that is recorded at the beginning of the pulse gives a measure of how many electron spins relaxed over a time τ after being polarized by the **Pump** pulse.

This sequence is repeated several thousands of times, and the single photons that are received are integrated for each time τ with 100 ns resolution. To measure the recovery of the population, the first 10 μ s of each **Probe** pulse fluorescence signal are integrated and then normalized by dividing this value by the same integral, but of the corresponding **Pump** pulse. This procedure is used to take drifts of the optical coupling during the run of the experiment, as well as other drifts, into account. Since some of the measurements took several weeks, this normalization was critical. This resulting normalized recovery signal was used to extract the T_1 of the electrons in the defects. For the measurements the IR laser was always on and locked using the wavemeter, to avoid a drift between the **Pump** and **Probe** pulses, which were created using the two AOMs. The following sections will present the results of the measurements conducted using this type of pulse sequence.

4.3.1 4H Beta

This section presents the results of the pump probe recovery T_1 measurement conducted on the 4H- β site at a temperature of around 100 mK. A sequence was performed as explained above with delays τ ranging from 5 μ s to 20 ms. The sequences in this measurement were performed at a constant rate, to ensure a constant sum of energy delivered to the sample for all data points, so to not distort the resulting recovery rate, by different sample temperatures. The recorded fluorescence can be seen in fig. 20 and shows the significant decay of fluorescence during the 1 ms **Pump** pulse and the gradual recovery of the signal as the delay time τ is increased. In this defect, the resulting contrast was less than 25% at 480 mT and a fast recovery was observed with a single exponential behaviour and a decay rate $\Gamma_1 = (1.0 \pm 0.6) \cdot 10^4 \text{ s}^{-1}$, recovering to around 95% of the initial **Pump** signal height. The weak contrast could be due to a very broad single defect optical transition, leading to insufficient separation of the electron $|\uparrow\rangle$ and $|\downarrow\rangle$ ground states. Another factor is the fact that at the measured wavelength $\lambda = 1335.45 \text{ nm}$, which was chosen to be the peak of the resonance, to maximize photon collection, a lot of GS2-ES1 transitions were driven as well. There still is a significant population in the GS2 at those temperatures and the transitions have been shown to overlap.

To verify that the observed contrast during the pumping is not just a dynamic be-

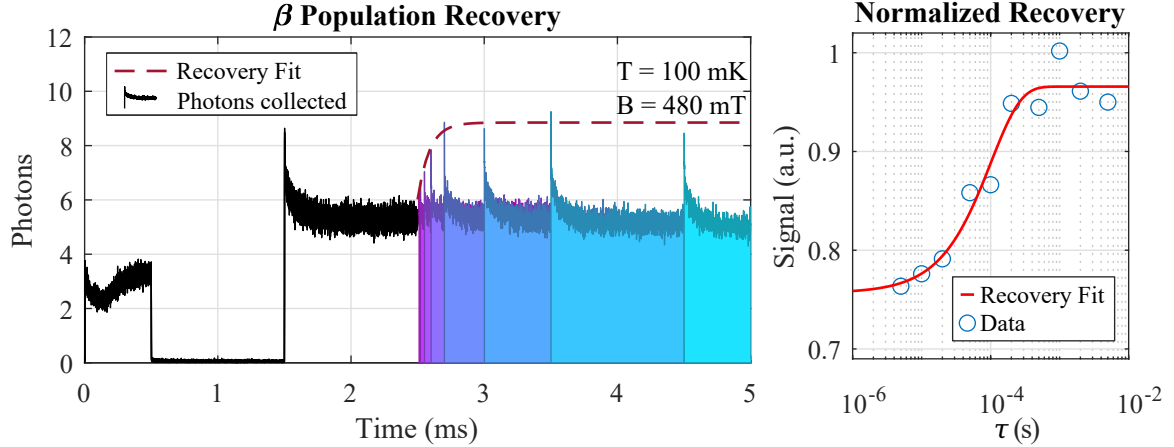


Figure 20: LEFT: Recorded fluorescence of the T_1 measurement on the 4H- β site for all the individual delay times τ overlayed. In black is the part of the sequence which is constant for all data points (**Init** and **Pump**) and the individual **Probe** pulses are plotted in color. The measurement was recorded with a 10 ns resolution, but to improve visibility a 1 μ s moving average was plotted. RIGHT: Resulting recovery signal and fit to extract the spin lifetime $T_1 \approx 100 \mu$ s .

tween the two ground states, the experiment was repeated at zero magnetic field, resulting in no relevant contrast at all. As transitions between the two ground states should not be very susceptible to magnetic fields, the observed contrast and subsequent recovery is believed to be actually spin lattice relaxation between the separated electron spins. However, the presence of an energetically close electronic state could offer additional decay channels for the electron spins to relax through, thus shortening their lifetime. More reference measurements were conducted by varying the delay between the green **Init** and the first **Pump** pulse, and the sequence was repeated with the IR lasers wavelength locked off-resonant to the sites optical transition, both showing no dynamic. From the previous measurements on the charge stability, no significant bleaching is to be expected at the observed timescales and the set laser power of 0 dBm.

The short lifetime ($T_1 \approx 100 \mu$ s) observed even at such low temperatures and the small contrast makes the 4H- β site unsuitable for a telecom qubit, at least under the conditions tested in this thesis. Therefore, no further investigation into the T_1 behaviour was conducted on this defect.

4.3.2 4H Alpha Temperature Dependence

Compared to the 4H- β defect the 4H- α defect turned out to be far more promising. In order to avoid photo bleaching at all costs, the **Pump** and **Probe** pulses were shortened to 200 μ s, while the green laser pulse was kept at 0.5 ms. Performing a measurement at the same constant rate over 20 ms showed very little recovery, indicating that the spin relaxes significantly slower than in the beta site. As each measurement was repeated tens of thousands of times in order to collect enough photons, it would take over a year to collect enough data points for an accurate de-

Alpha site Spin Population Recovery

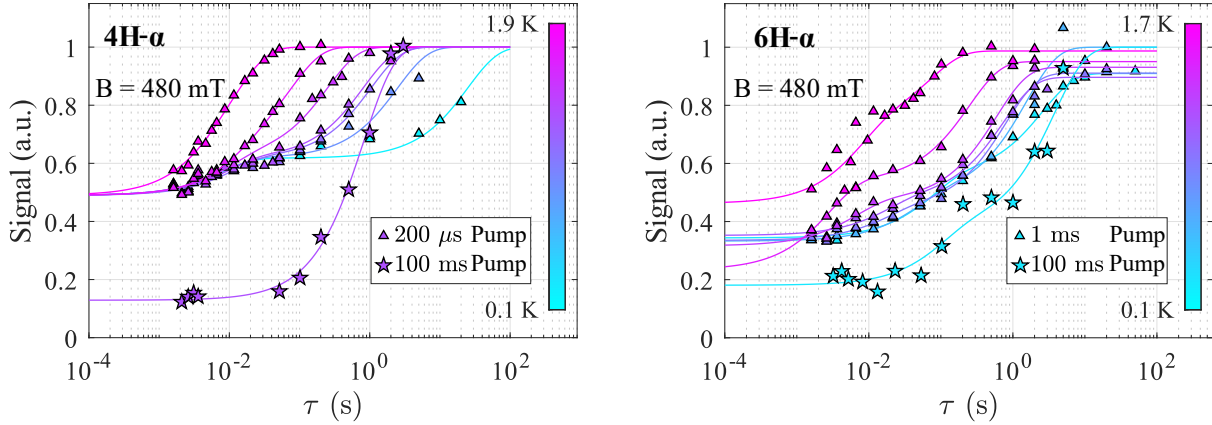


Figure 21: Recorded fluorescence recovery signal of the α defects in 4H (LEFT) and 6H (RIGHT) SiC. The standard measurement data points are displayed as triangles and the long 100 ms **Pump** and **Probe** measurements are displayed as stars. Datapoints are colored corresponding to the temperature there were recorded at, as can be seen in the colorbar next to each plot. In 4H α the "quick" decay disappears after long pumping, but does not in 6H, where the "quick" decay is significantly slower compared to 4H.

scription of the defects' electron relaxation dynamics. For this reason the sequences were played back to back, without additional delay added at the end of the sequence. Even though this method changes the thermal energy delivered to the sample per datapoint, which could also been seen on the cryostat temperature sensors, both the constant rate measurements and the variable rate measurements were performed for the short time scales and showed no significant differences in comparison. Other than that, the measurement was performed in the same general manner as in the section above. First, a cold temperature pump probe recovery measurement was performed at around 100 mK and data points were recorded from 1.6 ms up to 20 seconds.

With the 200 μ s **Pump** pulse it was possible to achieve a contrast of around 50% at 480 mT and steady recovery over time. However, even after 20 seconds, only about 80% of the total fluorescence was recovered, indicating a long T_1 , but also creating a problem of very long measurements, as each sequence was repeated 30 thousand times to collect enough photons. This would mean measurement times exceeding several weeks, to reach a recovery to 95% just for this one defect at this specific temperature, at the collection rate given by the experimental setup, creating a practical problem. To verify if the fluorescence would recover fully, the measurement was then repeated for different temperatures ranging up to 1.9K, thus increasing the spin-lattice relaxation rate Γ_1 in order to be able to measure a saturation in the spin recovery. From the collected recovery data, which can be seen in fig. 21, it was clear that it did not follow a single exponential behaviour, as expected for spin lattice relaxation. One fast decay was observed with a decay rate $\Gamma_0 \approx 100 \text{ s}^{-1}$,

which showed no significant temperature dependence, together with a slower decay Γ_1 that varied strongly with temperature. Also, the relative amplitudes of the recoveries seemed to remain fairly constant, with $\frac{1}{4}$ for the fast and $\frac{3}{4}$ for the slow decay ratio. At higher temperatures full recovery was observed and at lower temperatures, where due to long measurement times the wait time τ was capped before full recovery was seen, the fluorescence did still show a steady rise with no signs of saturation. Combined with the knowledge from the charge burn experiments performed, knowing that the laser was set exactly to the peak of the resonance, makes it reasonable to expect full recovery of the low temperature measurements for $\tau \rightarrow \infty$. This assumption led to the following fit function for the recovery curves, which described the system far better than a single exponential:

$$\text{Signal}(\tau) = (1 - c) \cdot \left(\frac{1}{4} \cdot (1 - e^{(-\tau \cdot \Gamma_0)}) + \frac{3}{4} \cdot (1 - e^{(-\tau \cdot \Gamma_1)}) \right) + c \quad (20)$$

Here $(1 - c)$ is the polarization achieved at $\tau = 0$ and Γ_0 , Γ_1 and c are the free fit parameters for each temperature's results. All the decay rates can be seen in fig. 22, showing that Γ_0 stays constant within the margin of error over the entire temperature regime observed, except for the 1.9 K measurement, where the two decay rates were less distinct, as they were of the same order of magnitude. Assuming that the temperature dependent "fast" Γ_0 decay at this temperature, actually surpassed the "slow" Γ_1 one, effectively flipping the last two 1.9 K recovery rates, the statement of the "slow" decay being constant could still hold. Γ_1 on the other hand does show a strong temperature dependence, as would be expected for a spin relaxation process. This observed behaviour could have several different explanations, one being that in the observed sample there is a discrete amount of "bad" sites, with spins that are not as protected and decay more rapidly. Such rapid decay could be due to a neighboring spin causing a spin flip due to spin-spin interaction, which should be temperature independent, or just local strain or other local lattice effects. Another possible explanation could be that some unknown charge dynamic is induced by the measurement sequence. Just like with the β site reference measurements were taken at zero field, off resonant and by varying the length of the pause after the green laser pulse, with no depletion or recovery dynamics being observed, to make sure the results were not compromised by external factors or other defects in the crystal. A possible electronic shelving state with a lifetime of ~ 10 ms could also lead to the observed behaviour. To get a better understanding of the dynamic, the T_1 measurement was repeated at 1.3 K but this time with a 100 ms **Pump** and **Probe** pulse, which can be seen in fig. 21. A temperature of 1.3 K was chosen because at this temperature, with a "long" life time of about 800 ms, it was reasonably fast to perform the measurements, while still having the possibility of distinguishing two clearly separate decay rates, as seen in the previous measurement.

The measured data shows that a longer pulse polarized the system with a higher contrast, in excess of 80%, and still did not bleach the defects, as full recovery was observed. Also the "fast" decay Γ_0 was no longer visible, as the "slow" decay Γ_1 stayed the same at roughly 800 ms. This demonstrates that the fast decay can be removed by pumping for a longer time, leading to just Γ_1 being the true spin lattice relaxation rate, while the fast decay that was observed behaves more like a

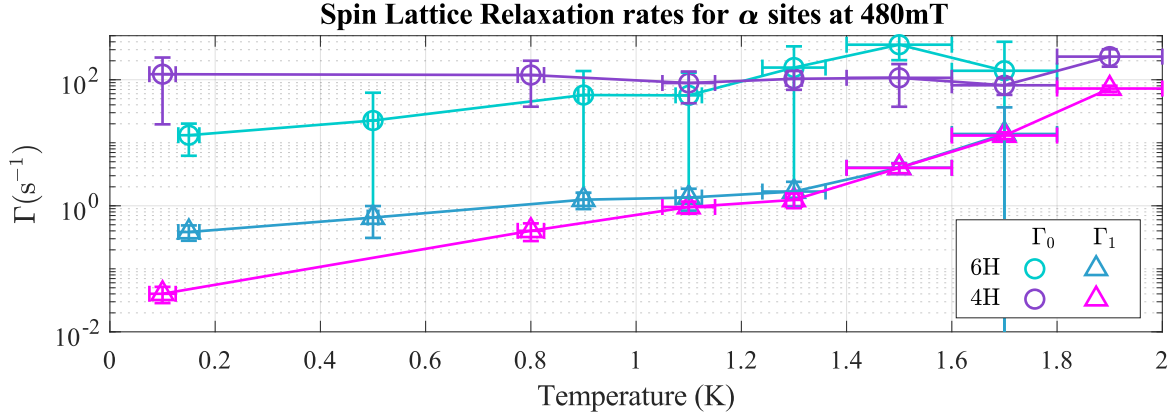


Figure 22: Decay rates Γ for the two α defects, in 4H (turquoise/green) and 6H (pink/purple), for temperatures from 100 mK to 1.9 K. The fast decays Γ_0 are depicted in circles and the slow Γ_1 data points as triangles. The errorbar in the decay rate was extracted from the fit functions, whereas the error in the temperatures were estimated by the jitter in thermal readings from the sensor on the experiment stage in the cryostat. All errors given are symmetrical. 6H Γ_0 errors that would extend into negative values were capped at the corresponding Γ_1 data point in this graphic to improve visibility.

metastable shelving state.

Therefore, the 4H- α defects spin lifetime at around 150 mK is 26(7) s and shows a linear increase with temperature T_0 up until 1.3 K, where it starts to rapidly decrease to less than 10 ms at 1.9 K. Unfortunately, due to strong temperature oscillations of the dilution refrigerator at elevated temperature, giving rise to large error bars with respect to the samples temperature, and the lack of data points in the low temperature regime, it was not possible to create good fit of the decay mechanisms (see section 2.4). However, it is possible to assume a rapid rise of the decay rate above 1.9 K, explaining why no spin polarization and lifetime T_1 was observed in previous research, which was conducted at higher temperatures [18].

4.3.3 6H Alpha Temperature Dependence

The α defect in 6H also seems to be a promising defect with respect to the spin lattice relaxation. A sequence was performed in the same fashion as shown in fig. 19, using 1 ms **Pump** and **Probe** Pulses, as there was no fear of bleaching for this defect (see section 4.2.3). Again the T_1 was measured for several temperatures ranging from 150 mK to 1.7 K and the recorded recovery can be seen in fig. 21.

Initially, the **Init** pulse was kept at the same power level as before, but this turned out to be insufficient to mix the electron spins. The 6H- α site needed significantly more green laser power to initialize the system sufficiently, which is why a new, stronger, fiber coupled 520 nm laser diode was implemented in the experiment, that replaced the previous green laser outside the fiber rack. More laser power leads to more heating, increasing the minimal temperature achieved in the experiment. For

the 4H- α defect the charge hole measurements were used to make sure the laser wavelength for this particular measurement was set to the peak of the resonance, ensuring that Pop1 and Pop2 were roughly the same size. It was therefore not possible to make recovery to 100% a reasonable assumption, as such calibration mechanism was not available for the 6H SiC alpha defect, the measurements were performed for τ long enough such that the recovery would saturate. And indeed at low temperatures the fluorescence did not recover fully, but saturated slightly below 100%, indicating that, at the measured wavelength, the two resonant populations were not of the same size (Pop1 $\neq$ Pop2). In the later performed PLE measurement, which was shown in fig. 15, that tracked the sweep wavelength, it was verified, that the used wavelength ($\lambda = 1308.57$) was slightly off the peak, leading to less visible recovery. As the low temperature relaxation rate turned out to be an order of magnitude faster, than in the 4H Alpha defect, it was possible to this within a reasonable timeline. Similarly to the 4H- α , also two decay constants were visible, but in contrast the quick decay Γ_0 did not seem to always have the same amplitude or value. For this reason a slightly different double exponential fit function was used to retrieve the decay constants for the various temperatures:

$$\text{Signal}(\tau) = a \cdot (1 - e^{(-\tau \cdot \Gamma_0)}) + b \cdot (1 - e^{(-\tau \cdot \Gamma_1)}) + c \quad (21)$$

Here a , b and c are free amplitude fit parameters, Γ_0 is the quick decay and Γ_1 is the slow decay which were extracted from the datapoints in fig. 21. Not only did the relative amplitudes of the two decays vary strongly depending on the temperature, but also Γ_0 itself seems to be temperature dependant. The extracted decay constants are displayed in fig. 22.

To find out whether the quick decay could again be removed by pumping longer, a 100 ms **Pump** and **Probe** pulse measurement was performed at around 150 mK, but although this sequence resulted in a higher contrast after pumping ($\sim 80\%$), both Γ_0 and Γ_1 recoveries were still visible. This finding means that the "quick" decay seems to follow a different mechanism in 6H- α than in 4H- α and can not be removed using longer **Pump** pulses. The resulting low temperature lifetime is therefore a mix of the two Γ 's, and it is not yet possible to say whether they are caused by a discrete population with shorter electron spin lifetime similar to a quantum fluctuator model, a decay channel or shelving state that affects all defects, or some other mechanism. At 150 mK, $\sim 37\%$ of the fluorescence recovers quickly with a lifetime of about 76(40) ms and the rest ($\sim 63\%$) recovers more slowly with a lifetime of 2.6(7) s. Both decays become faster as temperature increases, in a similar manner to 4H- α for temperatures above 1K. At about 1.3 K, the slow decay constant is still $\Gamma_1 = 1.7(7) \text{ s}^{-1}$, with $\sim 75\%$ of the fluorescence recovering at this rate. As temperatures are increased further, the two decays become harder to distinguish, leading to larger error bars in the fit, and a lifetime of roughly 7 ms for the quick decay and roughly 70 ms for the slow decay are found, both with similar amplitudes.

4.3.4 Magnetic Field Dependence

For the two α defects in 4H and 6H SiC, the dependence of the electron spin lifetime was also measured with respect to the applied magnetic field B_0 . These measurements were performed with the same sequence style (**Pump/Probe** = 200 μs for 4H

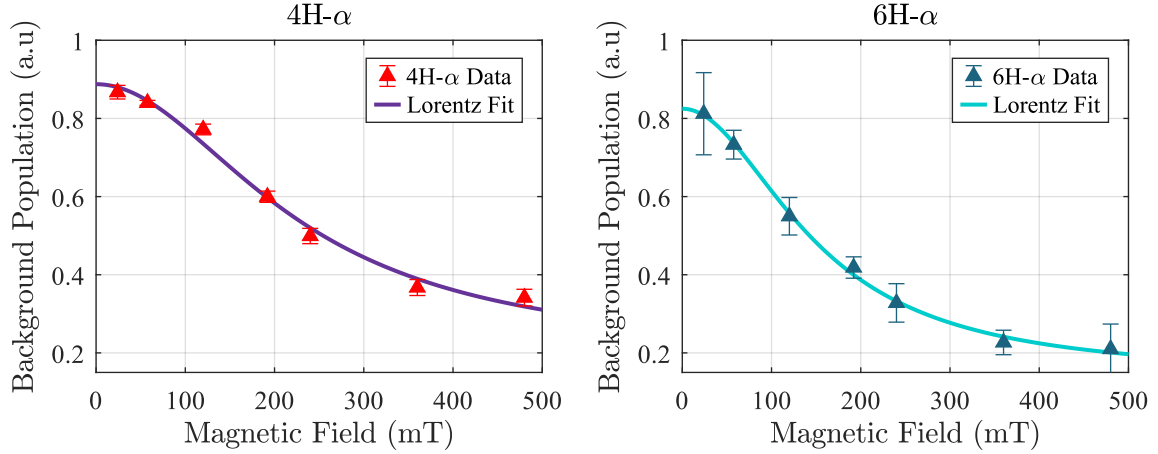


Figure 23: Plots displaying the background fluorescence after optical pumping as a function of the magnetic field for 4H- α and 6H- α . The datapoints were fitted with a lorentzian lineshape to extract information about the optical linewidth. Measurements of the 4H- α were performed at 1.3 mK and for the 6H- α the temperature was 150 mK.

and 1 ms for 6H), as in the previous sections. While it was possible to measure the 6H sample at the lowest temperature of 150 mK, the temperature had to be raised to 1.3 K for the measurement of the 4H polytype. This temperature was chosen because the shortened lifetime allowed reasonably quick data collection, with the cryostat temperature being fairly stable, so systematic temperature errors could be minimized. Pump probe recovery measurements were performed at different field from 24 mT to 480 mT.

Interestingly, the spin lattice relaxation rate showed no significant B_0 dependence for neither of the sites. Both Γ_0 and Γ_1 did not deviate from the previous measurements at maximum field within the margin of error. However, the contrast did decrease rapidly with a decrease in field in both cases. The values for the residual background after pumping c (eqs. (20) and (21)), extracted from the decay rate fits, are displayed as a function of the magnetic field in fig. 23.

Assuming that the observed effect is solely due to the finite optical linewidth of the spin conserving $|\uparrow\rangle$ and $|\downarrow\rangle$ transitions, the data points were fitted with a lorentzian lineshape. Together with the literature values for Δg (0.492 and 0.491 for 4H and 6H [18]) and eq. (6), the resulting fit was used to extract the individual defects' optical linewidth, giving a FWHM of 1.6(5) GHz for 4H- α and 1.0(1) GHz for 6H- α , respectively. Since, in both cases, the pumping pulse applied was too short to create a steady state, this value should rather be considered as an upper value. By longer pumping, as shown in the previous sections (fig. 21), the polarization could still be improved, which would make the resulting Lorentzian more accurate and probably narrower. This flaw can also be seen in the maximum contrast achieved at 480 mT, which appears higher in 6H- α , but is likely caused by the longer optical pumping. From this observation it can be concluded that, in an ensemble at least, quite strong magnetic fields ($B_0 \approx 0.5$ T) are needed to sufficiently split the $|\uparrow\rangle$ and $|\downarrow\rangle$ optical transitions and thereby achieve a high fidelity in this type of initialization scheme.

4.4 Two Laser Spectroscopy

The final type of measurement in this thesis was an investigation into the hyperfine structure of vanadium defects in silicon carbide, as it is critical to understand the level structure of an electron for potential qubit applications. This measurement was performed employing a variant of two-laser spectroscopy, using a single laser and a phase modulator (EOM). As shown in eq. (17), applying an AC-voltage to the EOM creates sidebands with a shift relative to the laser frequency and by changing the AC frequency the relative shift will also be changed. Sweeping through different frequencies and by performing those sweeps for different magnetic fields it is possible to detect resonances within the hyperfine structure of both the ground and the excited state of a defect.

Initially, the idea was to lock the carrier at fixed wavelength and sweep the frequency shift of the sideband across the range of the EOM, which can be seen in fig. 24 TOP. This style of spectroscopy was termed F-2F spectroscopy because transitions can be driven between the carrier and a sideband, but also between the two sidebands themselves, creating a copy of each feature at half the set frequency. It is also susceptible to higher order harmonics of the sidebands, leading to additional artefacts. One advantage of this method is a high contrast in the measurement, especially at high fields, as can be seen in Figure 25, but the copies of each feature make it extremely difficult to extract information about the hyperfine level structure at the origin of the scan and at low field, where the feature density is highest.

For this reason, F-F (F minus F) spectroscopy (fig. 24 BOTTOM) was implemented, which relies on the use of two sidebands. In the F-F method one sideband is positioned at a fixed EOM frequency (4.5 GHz) and the second sideband is used for sweeping. This method makes it possible to scan only for features or transitions that are resonant with the difference between the two sideband frequencies, as the other sidebands and the carrier itself are far off resonance from transitions within the hyperfine structure of the defect. However, this method also comes with some challenges. Apart from overall reduced contrast, it is very sensitive to the positioning of the laser carrier. Choosing the laser wavelength is critical, as the carrier has a much higher intensity than the modulated sidebands. The carrier therefore contributes strongly to the fluorescence, which is why the measured defect must be sufficiently narrow to tune the carrier away from the defect's resonance.

Fortunately, the two α defects had a very steep slope to which the laser could be tuned, so that only one pair of sidebands was actually resonant with the optical transition of the defect. Also, the sweep range had to be chosen carefully, so as to not drive transitions between the carrier and the sweeping sideband, limiting the range of the sweep compared to the F-2F type scheme.

In the experiments, a range from 3.0 to 4.5 GHz was chosen to avoid pumping between the carrier and the sweeping sideband, especially at higher fields. Even though this scheme is less sensitive to higher order sidebands than the F-2F method, the RF-power was always reduced until no higher order harmonics were visible in the Fabry-Perot-interferometer. All of these effects strongly reduce the contrast in the recorded fluorescence, making integration times longer by up to two orders of magnitude, and making it more sensitive to power fluctuations of the sidebands with regards to frequency. As all of the used microwave components, as well as the phase

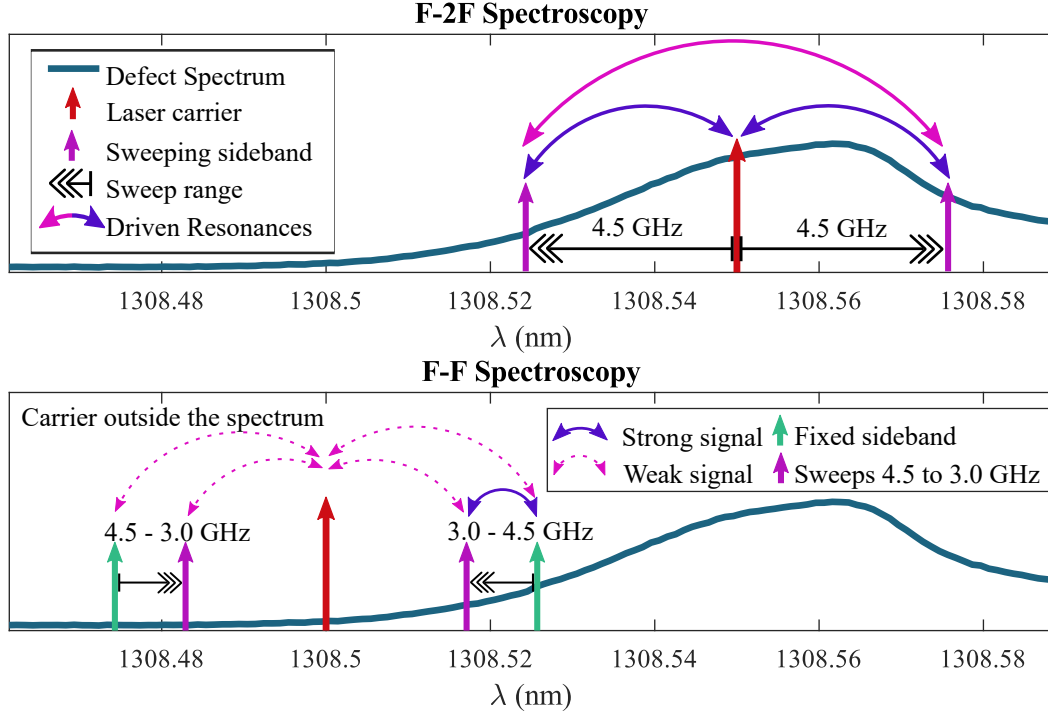


Figure 24: TOP: Schematic showing the F-2F two laser spectroscopy scheme on an example 6H- α resonance. The carrier is positioned at the center of the resonance and the two sidebands sweep the full range of 4.5 GHz symmetrically. Transitions are being driven not only between the carrier and the sidebands, but also between the two sidebands, leading to copies of each feature in the Scan. BOTTOM: Schematic of the F-F spectroscopy method, showing the positions of the carrier and sidebands as they were used in the experiment. The carrier is outside the resonance to contribute less to the collected fluorescence, one sideband is fixed at 4.5 GHz and a second sideband sweeps a reduced range from 4.5 GHz to 3.0 GHz. The transitions driven depend on the difference between the two sidebands and no other transitions should be resonant within that range.

modulator itself, do not behave constant for every applied frequency, the power of the sidebands can strongly fluctuate over the range.

To counteract this, the recorded data was processed by subtracting each data point in the scan by a moving mean in the y-direction to counteract frequency dependent errors and the x direction to eliminate time dependent external effects (e.g. lights in the laboratory, optical decoupling,...). For an improved visual of the scans, the remaining noise was removed using a Wien-filter in all of the two laser spectroscopy scans displayed [41]. Overcoming all those challenges made it possible to resolve the hyperfine structure of the both the α defects ground state (GS1) and excited state (ES1) with unprecedented quality. All measurements were performed at very low temperatures of around 100 mK.

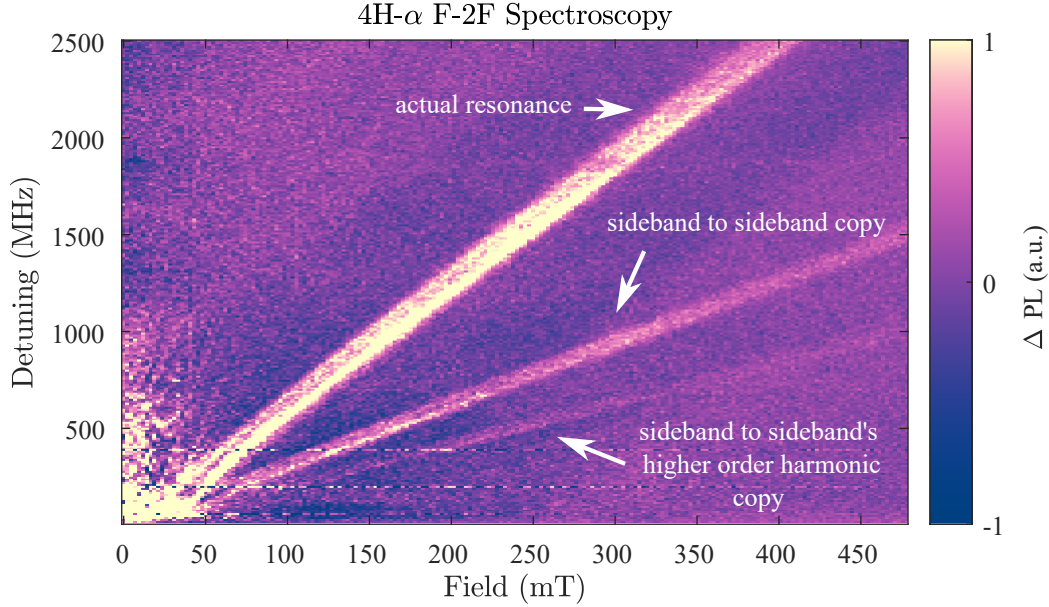


Figure 25: Two laser spectroscopy scan using the F-2F method, with only one pair of sidebands, of the $4H-\alpha$ site GS1-ES1 transition. Most visible is the actual resonance between the carrier and the sidebands and the copies that are caused by pumping between the two sidebands and their higher order harmonics. No structure can be identified at the origin of the plot, due to the signal copies obscuring the actual signal. The measurement was taken at around 100 mK, raw data was subtracted by a running mean in the x and y direction and noise was removed using a Wien filter.

4.4.1 4H Beta

This section presents the results from the two laser spectroscopy that was performed on the $4H-\beta$ defect. Due to the enormous width of the resonance, a F-F scheme was not possible and a F-2F spectroscopy was performed instead. In this measurement the sideband swept in a range from 10 MHz to 1990 MHz in 10 MHz steps. The magnetic field B_0 was ramped from 0 to 96 mT in steps of 0.24 mT.

Even at high fields no strong resonance features were observed. However, slight dark patches starting at around 400 MHz at zero field and moving outward, eventually fading, as the magnetic field increases, are visible. In fig. 26 a subsection of the scan in the range from 0 to 59 mT is displayed, showing those dark features. Unfortunately no further structures could be identified during this measurement, giving no improved understanding of the defect.

This lack of signal could be due to the very short electron spin lifetime of the defect, making it impossible to properly polarize the electrons into states, that could have been recovered by the pumping of the second laser. Another factor that might obscure the results is the big overlap and the strong dynamic within the ground state GS1-GS2 manifold. Even though an F-2F scheme should deliver more contrast, the consequent pumping between the different sidebands and higher order harmonics could have also contributed to a smearing of possible features.

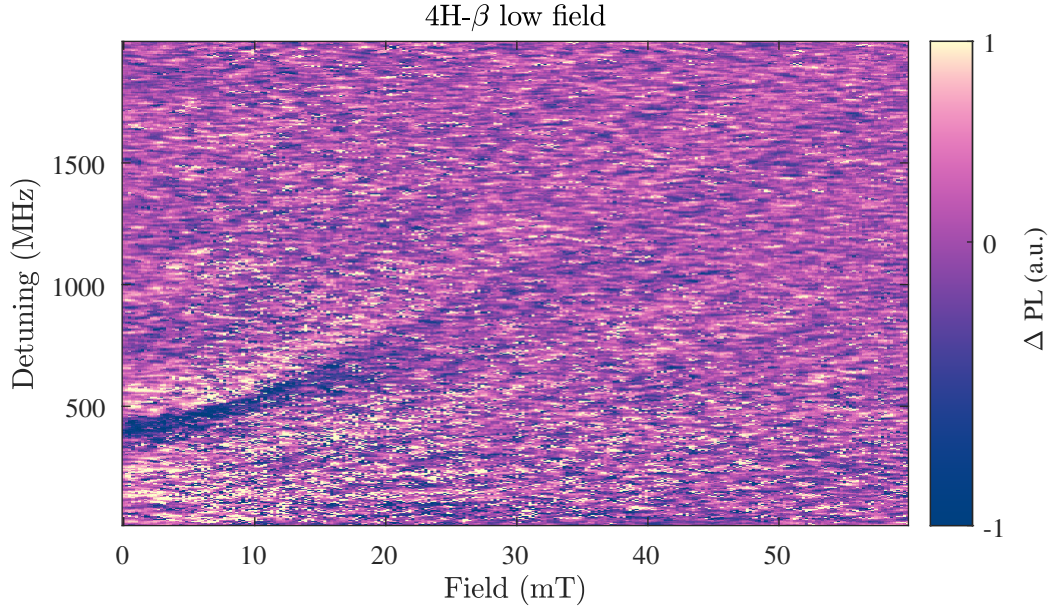


Figure 26: Two laser spectroscopy scan using the F-2F method, with only one pair of sidebands, of the $4\text{H-}\beta$ site GS1-ES1 transition. Dark areas were observed but no concrete level structure could be identified. The measurement was taken at around 100 mK, raw data was subtracted by a running mean in the x and y direction and noise was removed using a Wien filter.

4.4.2 4H Alpha

This section presents the results from the two laser spectroscopy that was performed on the $4\text{H-}\alpha$ defect's GS1-ES1 resonance. The first two laser spectroscopy measurement performed on this site was an F-2F type measurement that can be seen in fig. 25. As explained above, this approach yielded strong signals but also artefacts that obscured the structure, which is why the F minus F (F-F) scanning scheme was implemented.

First a high resolution measurement was performed. The magnetic field B_0 was ramped from 0 to 57 mT in steps of 0.24 mT and the sideband scanned a range from 0 to 1.5 GHz with steps of 1 MHz.

In fig. 27 TOP the result of this measurement is plotted, showing a signal that is rich in features, a big improvement to the previous scans. Both dark and bright lines can be seen starting at zero magnetic field, spreading out as the field is increased. Dark features seem to go in straight lines with origins distributed from 0 MHz to around 800 MHz with a spacing of a little over 100 MHz. The lines split as the field increases crossing each other and forming a checked pattern. Also at zero field bright lines can be seen originating all around 800 MHz and spreading out initially. As field increases they all bend towards the higher frequencies, parallelizing and eventually fading. At fields of around 10 mT very clear bright lines are visible going towards the x-axis and being "reflected". Those lines seem to converge as the bias field increases and start to move towards higher frequencies, becoming the dominating feature at 57 mT. Many more fainter both dark and bright features can be seen in the plot. Some of the fainter bright lines cross, forming bright spots, three of which are very

distinct. One is at $B_0 = 29$ mT and 330 MHz, the second can be seen at $B_0 = 38$ mT and 240 MHz and the last at $B_0 = 44$ mT and 590 MHz. All those features together with the crossing point markers, can act as a hyperfine fingerprint of the defect, making it possible to fit and improve theoretical models of the α -sites Hamiltonian.

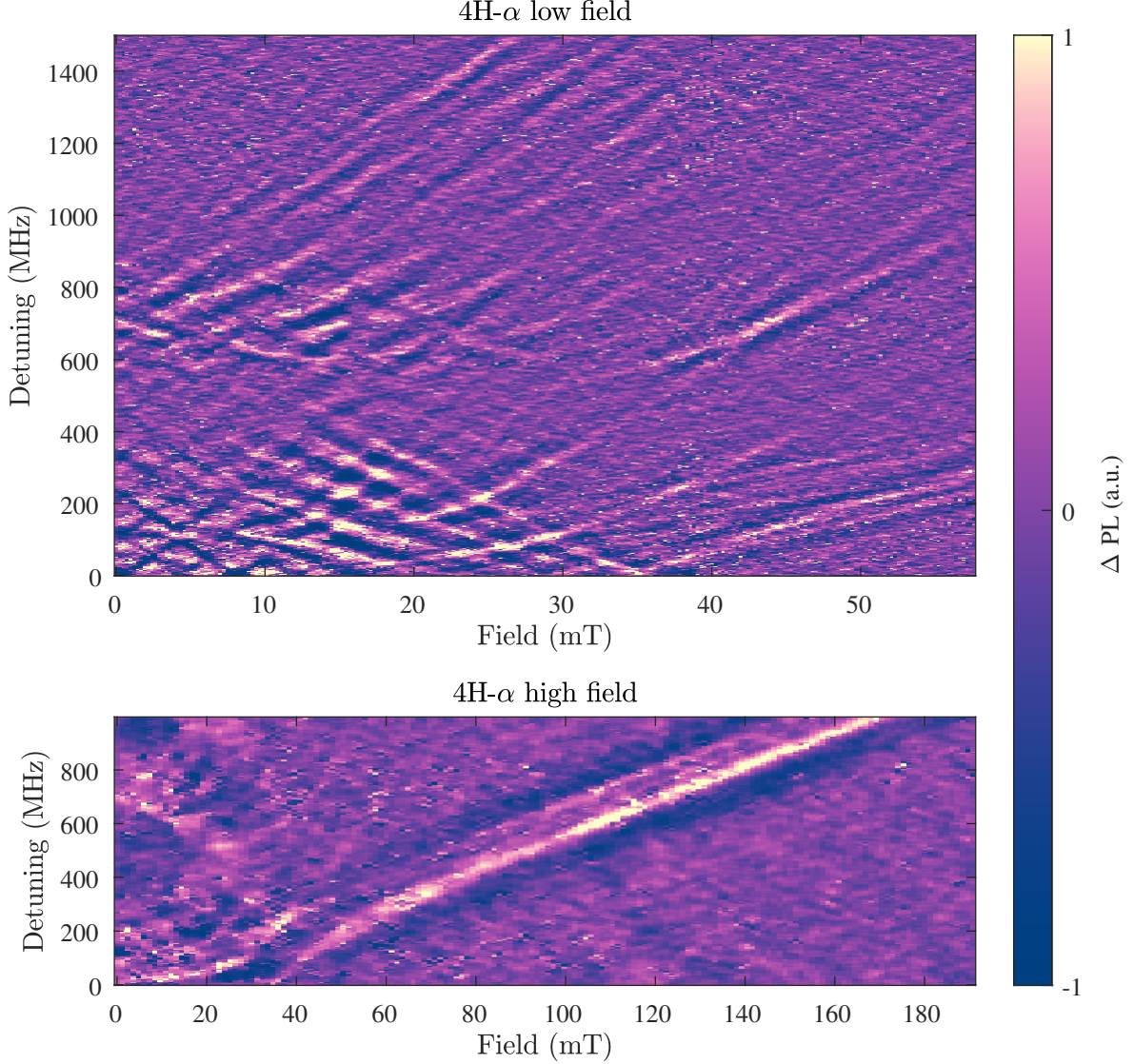


Figure 27: Two laser spectroscopy scans using the F-F method, with two pairs of sidebands, one fixed and one sweeping, of the 4H- α site GS1-ES1 transition. The measurements were taken at around 100 mK, raw data was subtracted by a running mean in the x and y direction and noise was removed using a Wien filter. TOP: Low field scan with high resolution of 1 MHz and 0.24 mT. Many bright and dark features can be seen as well as distinct crossing points of individual lines. BOTTOM: High field scan with a resolution of 5 MHz and 1.2 mT. At high field the dominating transitions start to parallelize and move outward at a constant slope.

For a better understanding of the behaviour at higher magnetic fields, a coarser scan was performed, with B_0 going from 0 to 191 mT in steps of 1.2 mT. To increase measurement speed, the frequency range was reduced to 995 MHz and steps of 5 MHz.

The resulting scan can be seen in fig. 27 BOTTOM. Here, the fine lines at low fields are not visible, only bright and dark patches, hinting at the structure seen before. However it can be seen that the lines that dominated at the high end of the low field scan, start to parallelize and move outward from the x axis in a straight line as the magnetic field increases. The lines do not seem to be significantly broadened as the field increases, showing that the field experienced by the observed defects is rather homogeneous.

Assuming that the slope of this line is given by the optical splitting of the spin conserving spin $|\uparrow\rangle$ and $|\downarrow\rangle$ transitions as given in eq. (6), the difference in the ground and excited states g-factors can be calculated. This fit results in a $\Delta_g = 0.460 \pm 0.001$, which deviates significantly from the literature value of 0.492 [18], but is probably due to the uncertainty in B_0 .

4.4.3 6H Alpha

Finally a two level spectroscopy measurement was performed on the alpha defect in the 6H polytype, again on the GS1-ES1 transition. As the resonance also was quite narrow a F-F type measurement was possible. First a high resolution low field scan was performed with the sideband sweeping from 0 to 1.5 GHz in 1 MHz steps and B_0 going from 0 to 57 mT, with a 0.24 mT resolution.

Even though this measurement was integrated for a significantly longer period of time, the features seen are a lot less sharp compared its 4H counterpart. This is probably due to the lower fluorescence emitted by the defect and could be improved by even longer integration times, but could also have intrinsic causes. The resulting scan can be seen in fig. 28 TOP.

Nonetheless the signal is still very rich in features that look very similar to 4H- α , showing both dark and bright lines. A dark checked pattern can be seen again and also the bright lines that emerge at zero magnetic field at around 800 MHz and spread out as field increases. Many bright lines seem to fade as the magnetic field increases except for the ones that move towards the x-axis and then cross it. They converge as they move outward at the high field end of the scan. There are also crossing points, that are less distinct, but still visible.

A measurements for high fields was also performed with in a detuning range of 0 to 1495 MHz with 5 MHz steps. The magnetic field was ramped up to 238 mT with steps of 1.2 mT in the scan, which can be seen in fig. 28. Here again a strong resonance line can be seen that emerges at around 30 mT and moves outward as the field increases. Using the slope and eq. (6), this results in $\Delta_g = 0.460 \pm 0.001$, again deviating from the literature value 0.491 [18] and the previous measurement presented in section 4.1.3.

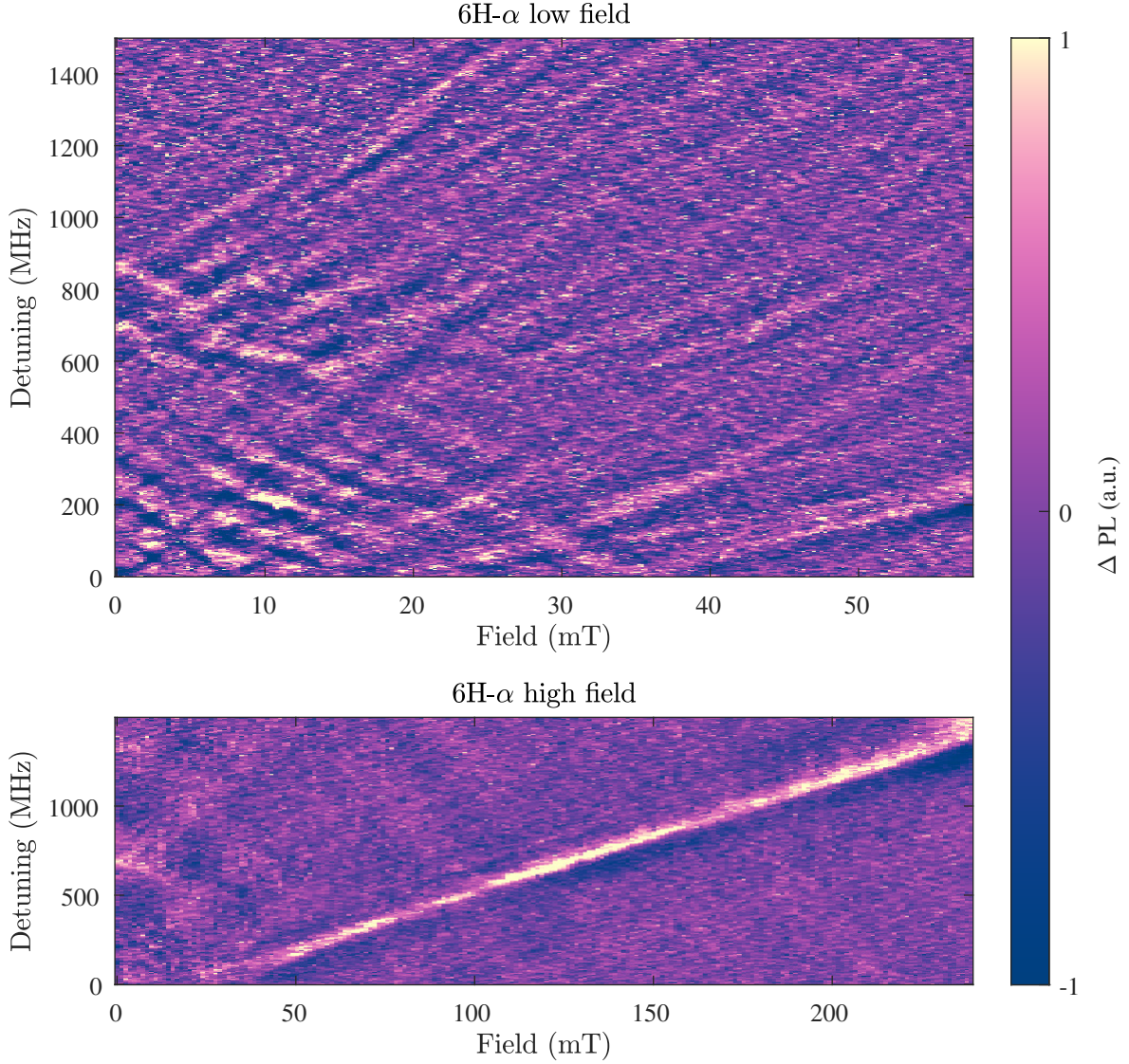


Figure 28: Two laser spectroscopy scans using the F-F method, with two pairs of sidebands, one fixed and one sweeping, of the 6H- α site GS1-ES1 transition. The measurements were taken at around 100 mK, raw data was subtracted by a running mean in the x and y direction and noise was removed using a Wien filter. TOP: Low field scan with high resolution of 1 MHz and 0.24 mT. Many bright and dark features can be seen as well as distinct crossing points of individual lines, even though less clear than in the 4H α site. BOTTOM: High field scan with a resolution of 5 MHz and 1.2 mT. At high field the dominating transitions parallelize and move outward at a constant slope.

Assuming the literature value is correct, this can be used to measure the magnetic field created by the solenoid. This would mean the B_0 experienced by the defects is lower by 6.3 % and actually only 225 mT/A are created by the solenoid, resulting in a maximum field of around 450 mT, instead of the 480 mT, which was used for this thesis. It is, however, within the margin of error expected for the field. Compared

Two Color Optical Transition Schemes

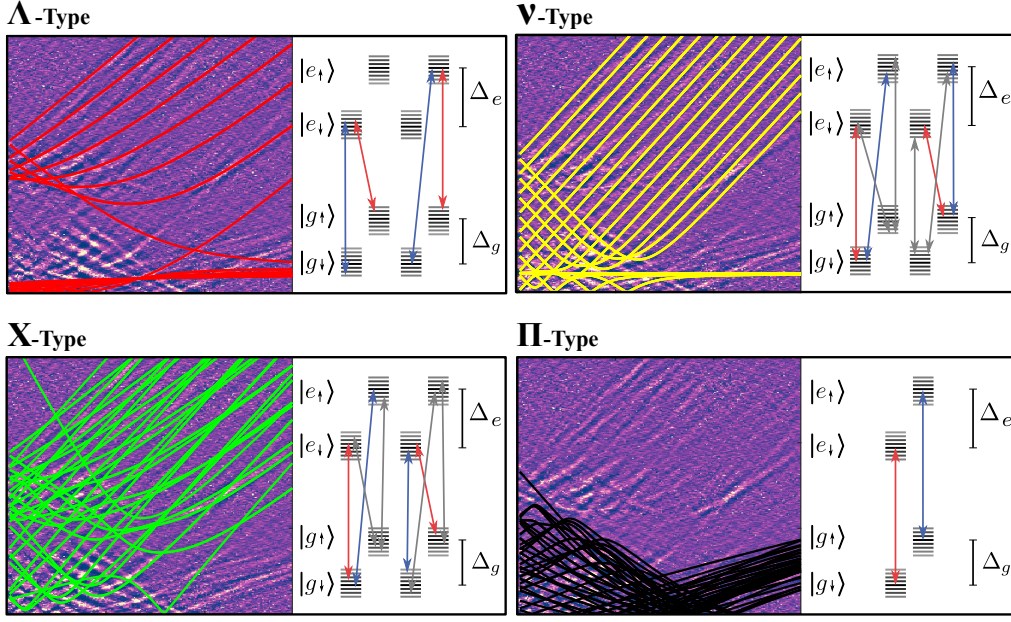


Figure 29: Visualization of the different two color resonance types. The different kinds of transition lines are plotted for each of the four types on top of the data that can be seen fully in fig. 27. Next to each plot a schematic of the pumping mechanism. The spin up and down manifolds are drawn in separated groups (left and right) for both the excited and the ground state. Red and blue (and grey) arrows represent the two different driving laser frequencies. Their splitting due to the Zeeman effect is denoted by Δ_g and Δ_e . TOP LEFT: Λ -type transition lines plotted in red. TOP RIGHT: ν -type transition lines are plotted in yellow. BOTTOM LEFT: X-type lines are plotted in green. BOTTOM RIGHT: Π -type lines are plotted in black.

to the previous PLE measurement of the Δ_g , fitting the slope seems like a more dependable measurement, since the PLE approach ignores a lot of possible optical transitions, while here only the transitions governed by Δ_g are used, as will be explained in the following section.

4.4.4 Transition Schemes

Using the two laser spectroscopy data from this thesis it was possible to get a better understanding of the level structure and Hamiltonian of the system. Michael Trupke in collaboration with Benedikt Tissot calculated the possible two color optical transitions of the system and they were able assign them to the features observed. In this section the results of the spectrum of the 4H- α site will be discussed and used for illustration, as this site showed the clearest features. However, the alpha site in 6H seems to work in a similar fashion, and can be treated analogous. Out of the immense amount of resonances a system of 16 ground and 16 excited states allows for,

four subgroups could be identified. Those four sub-groups dominate the observed spectrum, implying that the involved transitions have a higher probability.

A schematic of the different transition schemes can be seen in fig. 29 and they will be called Λ -, ν -, X - and Π -type resonances.

Starting with the bright lines that at around 800 MHz mentioned above that spread out and move outward. Those belong to the group of Λ -type schemes. The brightness originates from the two involved lasers pumping two different ground states into the same excited state. This leads to a continuous cycling of electrons into the excited state, which decay into the second involved ground state, from which they are being pumped back again. Thus an electron, that would be otherwise shelved in an off resonant hyperfine ground state, is recovered and contributes to fluorescence. Independent of the energy of the common excited state the resonance is just dependent on the difference between the two ground states. Λ -type resonances therefore purely give information of the ground state hyperfine structure, without needing to correct for the excited state and are displayed in fig. 29 TOP LEFT. To this group of transitions belongs the bright spot at $B_0 = 38$ mT and 240 MHz mentioned above (see fig. 27).

Next the checked pattern of dark lines was identified to correspond a set of ν -type resonances. Here the opposite effect takes place, involving only one ground state and two excited states. The two lasers pump the electron from the common ground state into two different excited states. From those excited states there is a possibility for the electron to decay into a different non resonant ground state, from which there are no longer addressed by any of the two lasers. This leads to the electron no longer contributing to the fluorescence, resulting in a darkening of the point in the scan. Similarly to before, the energy of the common ground state is irrelevant for the observed structure, as it is only dependant on the excited state hyperfine structure. The calculated structure can be seen in the TOP RIGHT of fig. 29.

Another type of transitions are the X -type resonances. Here four different levels are involved, two in the ground state and two in the excited state. The two lasers drive two transition, that seem to be independent, exiting the electron to different excited states. However, those two excited states have a high probability of decaying into the other laser's transition's ground state and vice versa, which can lead to an increase in brightness. As the transition driven by the two lasers share no common state, the resulting structure comes from a combination of both the ground and the excited states hyperfine structure. The lines can seem quite chaotic, as can be seen in fig. 29, but some of the afore mentioned distinct crossing points can be attributed this scheme. Notable examples are where X -type lines cross with Λ -type lines at the aforementioned bright features at $B_0 = 29$ mT and 330 MHz, and $B_0 = 44$ mT and 590 MHz. These were very useful for matching the theoretical model to the data, as there are many parameters involved.

Finally, Π -type resonances are shown in figure 29. These account for the bright lines that first move towards the x-axis and cross it. They converge as the field increases and become parallel at high fields. It is those that dominate the fluorescence in the long range scans (see fig. 27 BOTTOM and fig. 28 BOTTOM). Here the two lasers drive electron spin conserving transitions, one laser for spin $|\uparrow\rangle$ and one for spin $|\downarrow\rangle$. As an electron is excited there is always a probability of a spin flipping optical decay. The more the field increases the further away the different electron spin state's transition is. If only a single laser pumps a system, this leads to a depopulation of a specific electron spin state. In the case that a second laser is detuned in a way so it drives the other states transition, the electron can now decay back into the previous state. This leads to a pumping between the two electron spin ground states back and forth, resulting in a brighter fluorescence when hitting the resonance. At high fields (> 50 mT) the slope is given by the difference between the excited and ground state's g -factor Δg and is described well by eq. (6).

5 Conclusion and Outlook

In this thesis, a thorough examination into three sites of the neutral vanadium defect V^0 in silicon carbide was performed. Several new findings were enabled by performing a variety of optical spectroscopy measurements at far lower temperatures than all previous explorations of this center. The sites investigated were α and β in 4H SiC and α in 6H SiC, with all the measurements being performed on ensembles. On all the defects resonant PLE, photobleaching experiments, pump probe spectroscopy and two laser spectroscopy was performed. It was possible to measure a spin lattice relaxation rate for the first time and to get a good picture of the hyperfine energy levels.

This research made it possible to identify two possible candidates for qubits, namely the α sites in 4H and 6H, while the investigated β site does not appear viable for quantum applications, due to its very short lived electron spin state.

When it comes to the alpha sites, it is not yet clear which of the two could be more viable, as both show great promise, but differ in some aspects. One advantage that the 6H defect has is not a fundamental one, but rather due to the practicality of the wavelength (1308 nm) of its zero phonon line. As it is situated close to 1311 nm, which is very common in telecommunications, and there is a wide supply of devices that are optimized for this range that can be bought of the shelf cheaply. The ability to engineer the lineshape with long term stability, by bleaching the 4H- α site, and reviving the states with green laser light, could prove to be a very powerful tool, especially when dealing with broadened resonances in ensembles. With an anticipated switch to single defects for quantum devices, this would be less useful and could turn into more of a nuisance.

Undoubtedly, the biggest question mark hanging over vanadium defects as qubits was the spin lattice relaxations rate. Here, it was shown that both defects perform well, with the 4H specimen showing a lifetime of almost 30 seconds and over 1 second for 6H at very cold temperatures (~ 100 mK). Even more promising is the fact that at temperatures of around 1 K both defects' electron spins remain polarized for several hundreds of milliseconds, which makes them viable without having to resort to a dilution refrigerator system.

There are, however, open questions to the observed two decay constants and the different behaviour of the two sites. While in 4H the fast decay seems to be constant at around 10 ms, and can be removed by initializing the system with longer laser pulses, in 6H it seemed to change with temperature and it has so far not been possible to suppress it. From the measurements performed, it appears that, even though both defects can be optically polarized in the presence of a magnetic field, it is more efficient in 4H, showing a better maximum fidelity.

The two laser spectroscopy measurements that were performed revealed a wealth of information about the hyperfine structure, which is similar for both of the alphas. It further proved that optical pumping between the different spin and nuclear spin levels of the system is possible.

For a direct comparison between the sites, an overview of key results can be seen in table 2.

	4H- α	6H- α	4H- β
GS1-ES1 ZPL (nm)	1278.8	1308.6	1335.3
Spin contrast measured (%)	93.5 ± 0.5	89 ± 1	~ 30
T_1 max (s)	26 ± 7	2.6 ± 0.7	$(1.0 \pm 0.5) \cdot 10^{-4}$
T_1 at 1.3 K (s)	0.8 ± 0.2	0.6 ± 0.3	-
Optical resolution and adressability of hyperfine structure	Yes	Yes	No
Individual electron spin optical linewidth FWHM (GHz)	1.6 ± 0.5	1.0 ± 0.1	-
Charge switching	Yes	No	Yes
Thermal polarization in GS1	High	High	Low

Table 2: Overview of key results for the three investigated defects in this thesis. Electron spin lifetime values for 4H- α and 6H- α displayed are given by the "slow" decay constant: $T_1 = \frac{1}{\Gamma_1}$ (see sections 4.3.2 and 4.3.3).

Even though much further research is needed, both alpha sites look promising so far. They fulfill many of the key requirements needed to realize a quantum node, as described in section 1, as they have strong ZPL transitions in the telecom band, can be optically spin polarized and exhibit very long spin lifetimes, that should not put a critical limit on the coherence times. To realize a qubit, one major challenge will be to show that it is possible to polarize not only the electron spin, but also the nuclear spin. Currently, theories are being developed for an all optical polarization method, inspired by the presented two laser spectroscopy measurements, as well as methods that combine optical driving with added oscillatory magnetic fields.

This leads to the next objective, which is magnetic driving of the spins. Even though previous research shows that it is possible [18], it has never been implemented at temperatures below 2 Kelvin for the V^0 defect in SiC. The need for cold temperatures creates several technical challenges when trying to achieve high Rabi frequencies through magnetic driving, as a high field requires high currents, resulting in heating.

The next step would be a switch from ensemble to single emitters, which entails multiple challenges as well. First, it would be important to grow isotopically purified silicon carbide and implant it with vanadium. This is not a straightforward process, as the right balance of dopants, that act as charge donors and acceptors to create an environment that could stabilize the desired vanadium charge state, is still a mystery.

Another problem of single emitters is the low count rate, compared to ensembles. One way to overcome this problem could be solid immersion lenses or packing the defect into a microcavity that could increase the number of collected photons significantly [42].

Once these challenges are overcome, the vanadium defect in silicon carbide could become a very useful tool in the emerging field of quantum technologies. It combines an optical interface at a very practical wavelength with a long lived electron spin and may thus find application as a qubit or quantum memory within photonic quantum nodes in the years to come.

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