Development of measurement protocols for quantum magnetometry

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Acknowledgments

In December 2014 my journey as a PhD student in Denmark began. In the Quantum Physics and Information Technology group, or short QPIT, headed by Prof. Ulrik L. Andersen, I had the great possibility to start my PhD studies. And the following three years promised to be a very exiting time in my life.

The PhD project offered me an opportunity to do my studies at two different places with an almost equal duration at each stay. The other place was the quantum optics group at the University of Ulm, headed by Prof. Fedor Jelezko. Together with Associate Prof. Alexander Huck, also from the QPIT group, I got offered a unique and professional supervision team.

Although, it was not always easy to balance and organize the stays (and especially to manage the differences between the German and the Danish system), I could always rely on the help and guidance of Ulrik, Alex and Fedor. I have learned a lot from my supervisors, more than I can express in a few words, but maybe I can try to mention at least one important contribution they attached to my work and my personal development.

Ulrik showed me the wisdom of looking at the things from a different perspective. Even though, if they do not work out in the first place, there is still a lesson to learn and you have to take the chance to do it. Alex taught me most of all, that there are actually no problems - just challenges and there is always a way how to approach them (this might be true also for the real life). And Fedor showed me the necessity of having a firm determination, and prudence, which are truly qualities each scientist should pursue. These (and many more) are the reasons why I would like to express at first my sincere gratitude to my supervisor, Ulrik, Alex and Fedor, without whom this thesis would have never been possible.

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Haitham El-Ella bear a lot of discussions with me, about theoretical ideas, setup related matters, experimental realizations, and so much more. He has helped me to shape clearly
my ideas and concentrate on the important parts of the work. Nati Aharon gave me a lot of valuable insights in how to understand and to formulate theoretical concepts. His great patience in explaining and discussing, provided me with a deeper understanding of the world, which I try to explore by experiment. I had the pleasure to work during my stay in Ulm with Thomas Unden and Daniel Louzon. They supported me with a combination of great theoretical and experimental expertises and were always interested in a deeper understanding and exploration of the current findings. Also Nikolas Tomek has committed many hours to help me fixing issues with the FPGA and to discuss further concepts, either software or setup related. And there were of course, besides Haitham, my other lab fellows, Adam Wojciechowski and Sepehr Ahmadi, who made working in the lab a very pleasant and instructive experience.

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Abstract

The work presented in this thesis revolves around quantum enhanced magnetic field sensing. The core of the study is the nitrogen-vacancy (NV) center, a point defect in the diamond lattice with atomic like properties and magnetic field susceptibility. The central task was to develop control schemes and protocols to enhance the lifetime and the coherences of the NV center with the overall goal of enhancing the capabilities of this sensor in the field of magnetometry.

In order to realize complex protocols, a sophisticated software control of the measurement setup is required. A general software framework, termed Qudi, was designed and developed from scratch to improve drastically the measurement abilities and to facilitate the transfer of knowledge between the measurement protocols and the underlying physical idea. The transparent character of the core code, fully open to the scientific community, is serving as rigorous framework to reduce the complexity of the setup configuration by a fundamental separation of tasks. As a consequence, the general idea of this framework is not limited to experiments with color centers in diamond, but can find application in any laboratory environment.

The measurement of magnetic fields in the high-frequency GHz regimes is challenging. In this thesis, a continuous dynamical decoupling protocol is developed and implemented, which extends the capabilities of the NV sensor to probe GHz signals with a narrow bandwidth. Moreover, the protocol protects the system from noise, attributed to the drive, and from external magnetic noise thereby prolonging the coherence time and reaching the lifetime limitation of the quantum states. We measured a coherence time of 1.43 ms at room temperature in a diamond crystal with a natural abundance of $^{13}$C atoms, resulting in a smallest detectable magnetic field strength of 4 nT at 1.6 GHz.

Creating a protected qubit from a three-level system requires multiple drive fields addressing different transition frequencies. Continuous dynamical decoupling can protect the system from external magnetic noise, but typically introduces large drive noise to the system. The combination of on-resonant and off-resonant driving fields serves an approach to circumvent the challenges associated with drive noise. While on-resonant drive maintains a large energy gap in order to protect the sensor from external magnetic noise, the off-resonant drive creates AC-Stark shifted energy levels, thereby significantly reducing the amount of drive noise on the three-level system. We experimentally demonstrate an improvement in coherence time, which is only limited by the second order contributions of the magnetic field noise while suppressing entirely the drive noise contributions. Further improvement can be achieved by a mere increase of the drive field as its noise is efficiently decoupled from the sensor.
Dansk resumé

Arbejdet i denne afhandling er udført inden for emnet kvanteforstærkede magnetfeltsmålinger. Studiet fokuserer på nitrogen-vakancecentret, en punktdefekt i diamantgitteret med atomlignende egenskaber og modtagelighed over for magnetfelter. Opgaven er at udvikle måleprotokoller for at forbedre denne sensors kapacitet inden for magnetometri.

For at realisere komplekse protokoller kræves en sofistikeret softwarekontrol af måleopsætningen. En generel softwareramme, kaldet Qudi, blev designet og udviklet fra bunden for at forbedre måleevnen drastisk og lette overførsel af viden mellem måleprotokollerne og den underliggende fysiske ide. Den gennemgåelige karaktér af kernekoden, der er fuldt åben for det videnskabelige samfund, tjener som streng ramme for at reducere kompleksiteten af opsætningskonfigurationen ved en grundlæggende adskillelse af opgaver. Som en følge heraf er den generelle ide om denne ramme ikke begrænset til eksperimenter med farvecentre i diamant, men kan finde anvendelse i ethvert eksperimentelt fysiklaboratorium.

Målning af højfrekvensfelter i GHz-regimet er udfordrende. Her præsenteres en implementering af en kontinuerlig dynamisk afkoblingsprotokol, som udvider NV sensorers evne til at probe højfrekvente signaler med en smal båndbredde. Desuden beskytter protokollen systemet mod støj, der tilskrives det kontinuerlige drev (not sure) og fra eksterne magnetiske støjfelter, og derved forlænges kohærenstiden, der når levetidsbegrensningen på kvantetilstandene. Vi kan med den naturlige forekomst af $^{13}$C demonstrere en opnået kohærenstid på 1.43 ms ved stuetemperatur, hvilket resulterer i den mindste detekterbare magnetfeltstyrke på 4 nT ved 1.6 GHz.

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Chapter 1

Introduction

Life is a concept, like “universe”, that expands as soon as we reach what we think is its edge.

— Komand Kojouri
Quantum physics has significantly enriched and shaped technological evolution in the past years. As a consequence, our perception of the world has changed.

Quantum-based technology revealed new insights into a microscopic world which was alien to us. The development process spans wide, from small-sized inventions, enabling smartphones, computers and Lasers, up to the establishment of huge facilities like CERN, ITER or LIGO, serving primarily as hotbeds for further research in fundamental science. The results from all these inventions are constantly improving our understanding of the world. The knowledge is condensed into technology, such as measurement devices and sensors, that are equipped with new intricate abilities originating from the quantum world.

The start of the process was laid by the founding fathers of quantum physics in the beginning of the 20th century. Together, they developed a theory which allows us to understand the intrinsic nature of motion of small atomic size particles and their interactions. They proposed and verified concepts like the Uncertainty Principle [1], Quantum Superposition [2], Tunneling [3], Entanglement [4] or Decoherence [5] - all phenomena which cannot be described by classical Newtonian dynamics. This led to the evolution of Quantum Mechanics.

A domain in quantum physics, which actively pushes the current development to its limits, is the field of quantum optics. It is a theory which describes how the quantized field of light interacts with classical and quantum objects [6, 7]. Looking at our world from the perspective of optical photons (the quantum particle of light), it appears that it is a very cold one ($hν ≫ k_BT$). This viewpoint reflects the statement that the thermal occupation of the light field at optical frequencies represents the lowest energy state, making the photon interactions with matter very weak, except at certain resonant conditions.

Another active research branch pursuing the study of physical properties of solids is known as Condensed Matter Physics. It focuses on a variety of quantum objects like electrons, polarons, phonons, excitons, plasmons, or magnons [8, 9]. A subfield of that is Solid State Physics which investigates crystalline materials consisting of many atoms. More recently, research on single defects in all kinds of materials was guided by the premise of having individual atomic like structures. Eventually, it lays the basis for the ability to scale isolated systems and to design their mutual interactions. This development process has started from classical transistors and continues into the world of quantum objects [10, 11].

At the interface of quantum optics and solid state physics, the nitrogen-vacancy (NV) center has emerged [12, 13]. This system interacts with light while possessing atomic like properties, which originate from the configuration within its crystalline host material, diamond [14]. Its unique characteristics lends itself to a variety of potential applications ranging from quantum metrology [15–22] to quantum communication [23–25], as well as simulation and computation [11, 26–31].
In particular, employing the nitrogen-vacancy center as magnetic field sensors has received much attention as magnetic interactions are imprinted on its quantum state [32, 33]. Sensitivities of the order of nT/√Hz can be achieved with a single defect while maintaining a spatial resolution in the nanometer scale [15, 16].

The intrinsic quantum state of the NV center can be initialized and read out via the interaction with photons. The immediate access to quantum information and the ability to manipulate the system with magnetic fields create a fascinating quantum system for experimental physicists operating at room temperature.

This thesis deals with the investigation of the NV center use in magnetometry and the exploration of the intrinsic interaction of control fields with the sensor itself. The paramagnetic character of the NV center will be exploited to utilize the defect as a highly sensitive, nanometer-sized sensor for single spin magnetometry. The interaction with external magnetic fields lay the basis for the development of sophisticated protocols for which the susceptibility of the sensor to external magnetic signals can be shaped and controlled.

1.1 Structure of the thesis

The present work is structured as follows.

The first part, Chapter 2, is intended to give a concise overview of some basic properties of diamond, followed by an introduction to the NV center, its fundamental properties, and resulting applications.

Chapter 3 introduces briefly the setup configuration and highlight some important measurement techniques, which were used in this work.

The main part of the thesis consists of the three chapters, Chapter 4, Chapter 5 and Chapter 6. In the beginning of each chapter a motivation and summary of the investigated topic is given, followed by the research article attributed to this study. Within each paper the context of the study will be described, followed by the main findings, and concluded by a discussion and an outlook.

A final conclusion and outlook is given in Chapter 7.
Theoretical background for the nitrogen-vacancy center in diamond

There is pleasure in recognising old things from a new viewpoint.

— Richard Feynman
At the heart of each quantum system stands the desire to prepare, manipulate, and design its quantum state to test and verify the laws of quantum physics. With such systems we gain the opportunity to explore deeper the world of quantum mechanics, to identify the limitations, and capabilities of the given systems and to assuage the scientific thirst for curiosity.

For this reason the present chapter will try to elucidate the basic properties of the nitrogen-vacancy center and its host material. It will show why this quantum system has gained so much attention in the scientific community and why it has become a popular tool with a large potential for applications in the fields of quantum information processing [25, 34, 35], nanoscale imaging [17, 19, 21], electric field detection [20], thermometry [36], magnetometry [15, 16, 33] or biological sensing [22, 37, 38].

2.1 The host material

In 1978 J.H.N. Loubser and J.A. Wyk posed the question of studying diamond and its properties with regards to the presence of various defect centers [39]. In these early days diamond was already known as an extraordinary material. The lattice structure of diamond can offer explanations for the origin of its unique properties.

The crystal structure of diamond can be described by a face-centered cubic lattice arrangement of tetrahedrally bonded carbon atoms. Each carbon atom is connected to 4 neighboring carbon atoms by covalent sp\(^3\) hybridized orbital bonds, formed by two sp\(^3\) bands, each with the possibility to host 4 electrons. Starting from the carbon electron configuration, \((1s^2, 2s^2, 2p^2)\), the 2s and 2p electrons fill entirely the lower sp\(^3\) band forming the valance band [40, p. 67]. Thus, a completely unpopulated sp\(^3\) (conduction) band remains, which will be, due to the lattice constant of \(a_0 = 0.357\, \text{nm}\), apart by 5.47\,\text{eV} from the valance band. As a consequence, it would require light with a wavelength of 225\,\text{nm} to excite electrons from the valance band to the conduction band, constituting diamond as insulator. Therefore, diamond is optically transparent until the far infrared of 2700\,\text{nm} [41, p. 2.55 Table 21] (the visible spectrum ranges from 1.65\,\text{eV} to 3.26\,\text{eV}, or equivalently from 750\,\text{nm} to 380\,\text{nm}).

The strong covalent sp\(^3\) bands are also responsible for the high thermal conductivity (2200\,\text{Wm}^{-1}\text{K}^{-1} at room temperature [42]) as they allow an efficient energy transport of elastic vibrational modes in the crystal, known as phonons. The breakdown field of 10\,\text{MV cm}^{-1} [42] is one of the highest values for electrical field resistance making it a superior candidate over silicon to be used for very compact, high frequency field-effect transistors (FET). As a comparison, 20\,\mu\text{m} thick diamond can withstand 10\,\text{kV} bias voltage, whereas 1000\,\mu\text{m} thick silicon would be needed to perform equally.

Among other extreme parameters the high refractive index of around 2.380 [41, p. 2.55 Table 21] presents challenges on optical applications with diamond.
2.1 The host material

Table 2.1: Condensed classification of diamond in four types [43].

<table>
<thead>
<tr>
<th>Type</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ia</td>
<td>Aggregated nitrogen impurities ($\leq 0.3%$).</td>
</tr>
<tr>
<td>Ib</td>
<td>Isolated single nitrogen impurities ($\leq 0.05%$).</td>
</tr>
<tr>
<td>IIa</td>
<td>Nearly no nitrogen or boron impurities ($\leq 10^{18} \text{cm}^{-3}$).</td>
</tr>
<tr>
<td>IIb</td>
<td>Mainly boron impurities ($\leq 10^{20} \text{cm}^{-3}$).</td>
</tr>
</tbody>
</table>

The foundation of the type classification system (cf. Fig. 2.1) for diamond is mainly based on the present or absence of nitrogen (N), which is, besides boron, the major impurity color center in diamond [44, 45]. Type I diamonds are defined to have a measurable amount of nitrogen impurities ($> 5 \text{ppm}$) in the infrared absorption spectrum, whereas in type II diamonds, nitrogen ($< 5 \text{ppm}$) is not detectable by infrared spectrometers. These two general types are subdivided in categories based on the nature of impurities that are present. Table 2.1 gives a summarized version of the diamond characterization.

The creation electronic grade diamonds (with impurity concentration $< 50 \text{ppb}$) was considered to be one of the main challenges [39] and have developed massively in the past years [42]. Nowadays, diamond can be synthesized with two well-controlled methods, either with the high-pressure, high-temperature (HPHT) synthesis [46] or with the

![Fig. 2.1: Illustration of various appearing colors in diamond and their categorization.](image)

The color is strongly influenced by the impurities/defects in the diamond lattice. Figure adapted and taken from Ref. [45].
growth by chemical vapor deposition (CVD) [47]. This opened possibilities for the investigation of artificially incorporated impurities [44], which can change drastically the characteristics of diamond host. One major change is related to the color of diamond, which is determined by the leading impurity concentration [45]. Hence, the impurity concentration is a criterion for the characterization of diamond (cf. Fig. 2.1).

The large energy gap in diamond can serve as a host for various kind of isolated, atomic-like level structures, which can be addressed and manipulated optically. Those additional energy structures within the diamond lattice are introduced by impurities. Hence, this is the very reason why impurities are also termed “color centers” as they literally give diamond its color by absorbing specific quanta from the incoming visible light spectrum.
2.2 A defect among others and yet almost unique

A well known impurity is the nitrogen-vacancy (NV) center, which gives pink diamond its hue [12–14, 17, 38]. Such a system is created if a nitrogen and a vacancy replace the respective carbon atoms as shown in the crystallographic unit cell in Fig. 2.2 exhibiting a $C_{3v}$ symmetry. The combined entity 5 unbound electrons (3 from adjacent carbon atoms, 2 from the nitrogen) form the neutrally charged NV center ($NV^0$). An additional electron can be trapped from an electron donor in the lattice resulting in the stable negatively charged state ($NV^-$). The process of trapping can be enhanced by an optical excitation of the $NV^0$ state [48]. The conversion from $NV^-$ to $NV^0$ can occur by two-photon excitation at a suitable wavelength followed by an Auger process, which lifts one electron into the conduction band and removes it from the system.

The present work is based on the $NV^-$ center, hence, for the sake of brevity, the minus sign is dropped and the notation “$NV$” or “NV center” is used instead.

The absorption and emission spectrum at room temperature for the NV center is depicted in Fig. 2.3. The zero-phonon line (ZPL) of $NV^-$, which represents the direct transition from the excited to the ground state is a less prominent feature in both, the absorption and fluorescence spectra. Only 3-5% of the emission is attributed to the ZPL at room temperature [49]. Instead, a large phonon site band (PSB) contribution dominates the

**Fig. 2.2:** Crystallographic unit cell in diamond and the electronic configuration of an NV center. (a) Two carbon atoms (grey spheres) are replaced by a nitrogen (blue) and a vacancy (blurry orange) forming the nitrogen-vacancy defect in diamond. Electronic configuration of the defect determining the (b) ground state of $NV^0$, (c) its excited state, (d) the ground state of $NV^-$ and (e) its excited state.
Theoretical background for the nitrogen-vacancy center in diamond

Fig. 2.3: Room temperature optical absorption (red) and emission (black) spectra for high-pressure high-temperature (HPHT) diamond with NV$^-$ $\approx$ 15 ppm. The NV$^-$ exhibits the most absorption around 560nm. By exciting the NV center with a 532nm laser, an emission/fluorescence spectrum can be obtained. The peak feature in the absorption and in the emission spectrum corresponds to the zero phonon line (ZPL) of the NV$^-$ state. The ZPL of the NV$^0$ state is barely visible. The large band structure right from the ZPL in the emission, and left from the ZPL in the absorption spectrum, corresponds to the phonon sideband (PSB). Plot modified from Ref. [50]

spectrum. The sharp ZPL and the well defined vibronic bands imply that the optical transitions occur between discrete states deep within the diamond band gap [14].

The absorption spectrum reveals that photons in the range of 560nm excite efficiently the NV$^-$ state [14] yielding the strongest fluorescence response. A laser at wavelength of 532nm will excite off-resonantly into a vibrational mode of the excited state, as displayed in Fig. 2.4. The phonon sideband of the fluorescence spectrum highlights the difficulties in using NV centers for fast optical quantum information processing as phonon interaction will disturb a well defined optical transition. Excitation and subsequent decay to vibrational modes, results in distinguishable photons [51]. Phonon coupling, however, leads to dephasing and only photons from the ZPL are anti-bunched and applicable for quantum information purposes.

There are ongoing efforts in the scientific community to utilize cavities in order to shape the emission spectrum of the NV center [52, 53]. Enforcing the decay into the ZPL,
2.2 A defect among others and yet almost unique

Fig. 2.4: Energy level scheme of the NV$^-$ center. The discrete states of the NV center are situated between the valance and conduction band of the diamond. The ground state $|g\rangle$ and the excited state $|e\rangle$ exhibit both a zero-field splitting (ZFS). Two different decay paths are depicted, if excited with a 532 nm laser (green arrow). One direct spin conserving decay yields photons around 637 nm (red arrow) and an indirect spin non-conserving decay path over the metastable state $|s\rangle$ produces photons in the infrared region (dark red arrow). As the $m_s = \pm 1$ state (labeled with a moon) shows less fluorescence than the $m_s = 0$ (bright state, labeled with a sun), it is referred as the dark state.

which coincides with the optical mode of the cavity [54], would greatly enhance abilities of the NV center for the application as a quantum information processing unit [25] and single photon source [55] under ambient conditions.

The present understanding of the energy level scheme of the NV center within the diamond band gap is illustrated in Fig. 2.4. The level scheme can be considered in simplified terms as a ground state, $|g\rangle$, an excited state, $|e\rangle$, and a metastable state, $|s\rangle$. These states are analogous to the discrete levels of an atomic level structure. The fine structure can be obtained by considering the remaining unpaired electrons in Fig. 2.2 (d), (e). The energy level configuration is a result of the electronic configuration, the orbital symmetry and the surrounding environment. This can be explained by the powerful group theoretical approach, which starts from basic symmetry considerations and transfer the gained knowledge to the spin-spin and spin-orbit interaction model of the NV center.
Thus, the effective spin angular momentum with \( S = 1 \) of the NV\(^-\) constitutes to be a consequence of all these factors. The two remaining electrons (cf. Fig. 2.2 d) form a permanent magnetic dipole moment resulting into a zero-field splitting of \( D = 2.87 \) GHz [17] and lift the degeneration between the \( m_s = 0 \) and \( m_s = \pm 1 \) spin states. A similar situation occurs in the excited state, \(|e\rangle\), which exhibit a ZFS parameter of \( D = 1.42 \) GHz [57]. The other notations \(^3A_2, ^3E, ^1A_1\), and \(^3E\) stem from the group theory approach and describe the electronic configuration connected to the underlying orbital symmetry of the states [56, 58].

The ZPL of the NV center denotes the direct transition between the states \(|g\rangle\) and \(|e\rangle\). The off-resonant excitation with a 532nm laser will populate states in the vibronic band of the excited state, displayed as a fading gray band in Fig. 2.4, which have a lifetime of about 13ns [59, 60]. Depending on the spin state of the system different transition probabilities occur. In particular, if the ground state, \(|g\rangle\), is initially in the \( m_s = 0 \) spin state, it will almost always decay back to the same spin state upon excitation. On the contrary, having a spin in the \( m_s = \pm 1 \) state will yield in the excited state two main possibilities. In about 70% of the case, it will decay back to the ground state \(|g\rangle\) not changing its spin state, but in the other approx. 30%, the excited state will decay mainly to the intermediate state, \(|s\rangle\) via a non-spin-conserving, non-radiative decay path [61, 62]. After a short lifetime of about 1 ns [63], an infrared photon (at about 1042nm) will be emitted from the \(^1A_1 \rightarrow ^1E\) state [64]. The last decay step has an exceptional long lifetime of about 300 ns [63] and will almost always decay via a non-radiative, non-spin-conserving transitions to the ground state, \(|g\rangle\), with \( m_s = 0 \).

The described transitions are a consequence of selection rules. Not allowed transitions do exist, but occur with a small finite probability and are not mentioned here. A rigorous analysis can be found in Refs [61, 65].

To summarize, the decay mechanism is pointing out two main characteristics. The first is the spin dependent fluorescence intensity of the NV center [66]. Due to the additional, long-living, non-radiative decay path through the metastable state \(|s\rangle\) for the \( m_s = \pm 1 \) spin states, less photons per unit time are produced in comparison to the \( m_s = 0 \) spin state. Hence, the \( m_s = 0 \) state is denoted as the bright state and \( m_s = \pm 1 \) appear to be darker, thus, they are identified as the dark state(s) (marked with a sun or a noon, respectively, in Fig. 2.4). The second, almost unique feature is the ability to prepare the NV center with off-resonant excitation in the \( m_s = 0 \) state (with \( \leq 75\% \) fidelity, limited to ionization processes [62]) by cycling the decay path several times. This will optically “pump” the NV center in the \( m_s = 0 \) spin state, hence, yielding a very simple method for spin initialization. The two key properties will be highlighted in a subsequent section (see Sec. 2.4).

Apart from the NV center, there exists about 500 optically active defect centers in diamond [43, 67], which slowly started to gain attraction. The driving force besides the scientific curiosity is the hope to find a replacement for the NV center to overcomes its drawbacks [68]. The silicon-vacancy (SiV) center, for instance, shows superior optical
properties such as narrow ZPL of 0.7 nm at 738 nm with 70% emission into ZPL and a weak phonon sideband at room temperature [69–71]. But it lacks in good electronic spin properties, which is reflected by a small spin dephasing time $T^*_2 = 115 \pm 9$ ns and spin-lattice relaxation time $T_1 = 350 \pm 11$ ns, only achievable so far at 3.6 K and without an aligned static magnetic field [72]. The importance of these parameters for magnetometry will be highlighted in Sec. 2.5.

The germanium-vacancy (GeV) center is another candidate, fluorescing strongly with a about 40% in the ZPL at 602 nm [73–75]. However, the spin coherence time $T^*_2 = 19 \pm 1$ ns and the spin-lattice relaxation $T_1 = 25 \pm 5$ µs [75] are not reaching so far the achievable numbers of the NV center (in room temperature: $T^*_2$ in the order of µs, $T_1$ in the order of ms, ).

### 2.3 Theoretical model of the NV center

The nitrogen vacancy center in diamond can be modeled as a central spin system, which is coupled directly to its environment via dipole-dipole interactions [76, 77] Depending on the orientation and the species involved in the coupling, the interactions can be simplified to commonly known terms. The Hamiltonian formalism introduced here is focused mainly on the electron spin description and its various interactions. The description is kept as general as possible, and will be expanded upon when needed in the following chapters.

The static spin Hamiltonian, $\mathcal{H}$, under which the NV center is evolving, can be divided into an external part, $\mathcal{H}_{\text{ext}}$, and an internal, intrinsic part, $\mathcal{H}_{\text{int}}$.

$$\mathcal{H} = \mathcal{H}_{\text{int}} + \mathcal{H}_{\text{ext}} .$$

The internal part incorporates the ZFS contribution, $\mathcal{H}_{\text{ZFS}}$, the hyperfine interaction of the NV electron spin, $S$, with the nitrogen nuclear spin, $I_N$, and the higher order quadrupole interaction of the nitrogen

$$\mathcal{H}_{\text{int}} = \mathcal{H}_{\text{ZFS}} + \mathcal{H}_N + (\mathcal{H}_{\text{Q}}) = S^\dagger \mathcal{D} S + S^\dagger \mathcal{A}_N I_N + (I_N^\dagger \mathcal{Q}_N I_N) ,$$

where $\mathcal{D}$ represents the zero-field splitting tensor, $\mathcal{A}_N$ the hyperfine interaction tensor and $\mathcal{Q}_N$ the quadrupole interaction tensor [78, p.26]. The last term of eq. (2.2) is intended to be in brackets, since the quadrupole interaction only appears for a spin system greater than 1/2. Nitrogen has two naturally existing isotopes, the $^{14}$N as a spin $I_N = 1$ system, with natural occurrence of 99.63%, and the $^{15}$N as a spin $I_N = 1/2$ system. If the considered nitrogen of the NV center is $^{15}$N, than the quadrupole contribution has to be omitted.

$\mathcal{H}_{\text{ZFS}}$ results from the dipole-dipole interactions between the two unpaired electrons (cf. Fig. 2.3b), where $\mathcal{D}$ becomes a symmetric tensor. Diagonalizing $\mathcal{D}$ with respect to the
principle axis representation (the symmetry axis connecting the nitrogen atom and the vacancy, cf. Fig. 2.2) will yield

$$H_{\text{ZFS}} = D_x S_x^2 + D_y S_y^2 + D_z S_z^2 = D \left[ S_z^2 - \frac{1}{3} S(S + 1) \right] + E(S_x^2 - S_y^2), \quad (2.3)$$

with the parameters $D = 3D_z/2$ and $E = (D_x - D_y)/2$, and the relations $S_x^2 + S_y^2 + S_z^2 = S(S + 1)$, and $D_x/2 + D_y/2 = -D_z/2$ As the tensor $D$ is traceless,[39, p. 1208] the last part in eq. (2.3) after the equation sign is a valid expression. If a perfect C$_{3V}$ symmetry is assumed for the NV center with $S = 1$[39], then the impact of strain, $E$, in the diamond lattice can be neglected and the ZFS Hamiltonian reduces to

$$H_{\text{ZFS}} = D \left[ S_z^2 - \frac{2}{3} \right]. \quad (2.4)$$

The matrix notation of the spin matrices are given by

$$S_x = \frac{\hbar}{\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \quad S_y = i \frac{\hbar}{\sqrt{2}} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & -1 \\ 0 & 1 & 0 \end{pmatrix}, \quad S_z = \hbar \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}, \quad (2.5)$$

with the known relations of $S_+ = S_x + iS_y$, $S_- = S_x - iS_y$ and $S_x^2 + S_y^2 + S_z^2 = S(S + 1)$. The hyperfine interaction $H_{\text{HF}}$ between the electron spin $S$ and the nitrogen nuclear spin $I_N$ are in general connected through the hyperfine tensor $A_N$. As the NV center exhibits an axial symmetry, the hyperfine tensor $A_N = A_{N, \text{iso}} + A_{N, \text{aniso}}$ can be displayed in a diagonalized form in the principal axis representation of the NV:

$$A_N = \begin{pmatrix} A_\perp & 0 & 0 \\ 0 & A_\perp & 0 \\ 0 & 0 & A_\parallel \end{pmatrix} \quad (2.6)$$

The eigenvalues $A_\perp$ and $A_\parallel$ are related to the isotropic and anisotropic part [14, p. 24] and for a $^{14}$N nucleus take the values $A_\perp \approx -2.7$ MHz and $A_\parallel \approx -2.14$ MHz [79]. As a result, the hyperfine interaction $H_{\text{HF}}$ becomes:

$$H_{\text{HF}} = S^\dagger A_N I_N = A_\perp (S_x I_x + S_y I_y) + A_\parallel S_z I_z = A_\perp \left( S_+ I_- + S_- I_+ \right) + A_\parallel S_z I_z \quad (2.7)$$

where for the last part the relations $I_+ = I_x + iI_y$ and $I_- = I_x - iI_y$ has been exploited. The quadrupole moment stemming from the nitrogen $^{14}$N can be expressed in a similar way as the ZFS tensor in eq. (2.3). The quadrupole tensor in eq. (2.3) can be rewritten in the principal axes system to

$$Q_N = \begin{pmatrix} Q_{xx} & 0 & 0 \\ 0 & Q_{yy} & 0 \\ 0 & 0 & Q_{zz} \end{pmatrix}, \quad (2.8)$$
This Hamiltonian can be recast in the same manner as $D$, yielding the quadrupole Hamiltonian

$$H_{NQ} = I_N^2 Q_{2N} I_N = Q \left[ I_z^2 - \frac{1}{3} I(I+1) \right] + \eta E \left( I_x^2 - I_y^2 \right)$$

(2.9)

which exhibits a quadrupole moment $Q$ and an asymmetric part, related to the electric field gradient $\eta E$ [39, p. 1210]. For the case of the NV center $\eta E = 0$ and $Q = -5.01 \text{ MHz}$ [79], denoting the quadrupole coupling constant.

To summarize, the intrinsic Hamiltonian, $H_{\text{int}}$, can be expressed as

$$H_{\text{int}} = D \left[ S^2 - \frac{2}{3} \right] + \frac{A}{2} (S_+ I_- + S_- I_+) + A || S_z I_z + Q \left[ I_z^2 - \frac{2}{3} \right].$$

(2.10)

The external Hamiltonian, $H_{\text{ext}}$, describes the interactions with the surrounding environment of the NV center, as well as the external influences of applied magnetic fields. In this consideration, two main external contributions will be included, which are used within this work. They are formulated to

$$H_{\text{ext}} = H_{\text{HF,ext}} + H_{\text{mag}}$$

(2.11)

being the hyperfine interaction to external nuclear spins, $H_{\text{HF,ext}}$, and the interaction with an external magnetic field, $H_{\text{mag}}$.

The general representation of an external hyperfine interaction of the NV electron spin with adjacent nuclear spins writes

$$H_{\text{HF,ext}} = \sum_{i=1}^{N} S_i^\dagger A_{HF,i} I_i,$$

(2.12)

and is by construction identical to the hyperfine interaction in eq. (2.2). The major difference appears in the representation of the hyperfine interaction tensor

$$A_{HF} = \begin{pmatrix}
A_{xx} & A_{xy} & A_{xz} \\
A_{yx} & A_{yy} & A_{yz} \\
A_{zx} & A_{zy} & A_{zz}
\end{pmatrix},$$

(2.13)

which is usually not diagonalizable, making the description of the dynamics more complicated to conceive and to calculate. Hence, the external hyperfine interaction used to be adapted by approximating the interaction. A more detailed picture of the external interactions of the NV, including the fine and hyperfine structure, is given in Ref. [80].

The interaction with a magnetic field

$$H_{\text{mag}} = \mu_{B} g_e S^\dagger B = 2 \pi \gamma_{NV} (S_x B_x + S_y B_y + S_z B_z) = H_{\text{stat}}(B) + H_{\text{drive}}(B(t))$$

(2.14)

splits in two contributions. Here, the gyromagnetic ratio, $\gamma_{NV} = \mu_{B} g_e = 2.8 \text{ MHz G}^{-1} = 28 \text{ MHz mT}^{-1}$, of the NV electron was introduced. If the $z$-axis is identified as the
principle axis connecting the vacancy and the nitrogen, then the component $B_z$ will alter the separation of the eigenstates of $S_z$. This is often realized by a static magnetic field leading to

$$\mathcal{H}_{\text{stat}}(B) = \mathcal{H}_{\text{Zeeman}} = 2\pi \gamma_{\text{NV}} S_z B_z,$$

(2.15)

which describes a Zeeman interaction between the electron spin and the magnetic field [17]. Static magnetic field components, $B_x$ and $B_y$, alter the eigenstates of the $\mathcal{H}_{\text{Zeeman}}$ and introduce in general state mixing, which should be avoided [33, 81, 82]. Sec. 3.2 is going to elaborate that statement.

An oscillating magnetic field $B(t)$ can alter the population of the NV center (by the components $B_x(t)$ or $B_y(t)$), if the frequency of the magnetic field will correspond to the energy separation, $\nu_1$ and $\nu_2$, of the eigenstates. Due to the large value of $D$, only components of $B$, which are orthogonal to the NV symmetry axis, have an effect on the populations of the eigenstates (and the effect on $S_z$ through $B_z(t)$ averages out).

A exemplary field, which changes the eigenstate population is denoted as a drive and can have, the following appearance

$$\mathcal{H}_{\text{drive}} = 2\pi \gamma_{\text{NV}} \left( S_x |B_x| \cos (2\pi \nu_1 t + \varphi) \right).$$

(2.16)

but any other form could also be conceivable.

In summary, the main external contribution to the NV center can be expressed as

$$\mathcal{H}_{\text{ext}} = \sum_{i=1}^{N} S_i^\dagger A_{HF,i} I_i + \mathcal{H}_{\text{Zeeman}} + \mathcal{H}_{\text{drive}}$$

(2.17)

### 2.4 Readout and initialization process

The ability to prepare the NV center in an initial state is a major key for reproducible quantum state manipulations. The combination of a non-radiative decay path over the $|s\rangle$ state (cf. Fig. 2.4) and especially the long lifetime, $T_{|s\rangle}$, in the $^1E$ state enables to determine the current state of the NV electron spin by just counting the fluorescence response.

The state with $m_s = 0$ can be achieved, by cycling the decay paths in Fig. 2.4 with a laser. After the laser has been turned off for a time $t \gg T_{|s\rangle} \approx 300 \text{ ns}$, the NV center can be found initialized to $m_s = 0$ in its ground state $|g\rangle$. The lifetime of the eigenstates of the NV (described by the quantum number $m_s$) is then primarily determined by spin-lattice relaxation time, $T_1$.

It is important to note, that the determination of the spin state requires several thousand runs of summing up the photon counts (since one laser pulse of an exemplary length of...
Fig. 2.5: Accumulated count trace of the laser pulse for two different initial spin states. The spin state can be determined, by illuminating the NV center with a laser for a given time \( t \) and recording the fluorescent photons. For \( m_s = 0 \) more photons are detected in the first 250 ns, since the decay to the metastable state \( |s\rangle \) has a very low probability. After 1000 ns of illumination a steady state configuration is reached, where the NV is polarized into \( m_s = 0 \) state. This can be used to normalized the obtained curve to extract a normalized contrast, which determines essentially the spin state.

3000 ns would approximately yield \( 3000 \text{ ns} / T_s = 10 \). The result after several measurement runs would look similar to the orange curve in Fig. 2.5. The same measurement can be performed by preparing the system in a state \( m_s = \pm 1 \) (a procedure to achieve this will be presented in Sec. 2.5.2). This yields the blue curve in Fig. 2.5. The difference between the two areas under the curves determines the maximum obtainable contrast (magenta curve in Fig. 2.5).

By comparing both time traces, it becomes apparent, that they reveal different shapes. Fitting to those curves a simplified three level system, \( |g\rangle, |e\rangle \) and \( |s\rangle \) with decay rates \( \Gamma_{|e\rangle \rightarrow |g\rangle} = 1/T_{|e\rangle \rightarrow |g\rangle}, \Gamma_{|e\rangle \rightarrow |s\rangle} = 1/T_{|e\rangle \rightarrow |s\rangle} \) and \( \Gamma_{|s\rangle \rightarrow |g\rangle} = 1/T_{|s\rangle \rightarrow |g\rangle} \), will yield an initial estimation, that the fluorescence difference correspond to a contrast of about 30% [83].

The signal of the orange curve can be for instance \( \propto \left[(1 - \exp(-t/T_{\text{Laser}})) + \exp(-t/T_{|e\rangle})\right] \). For the blue curve, the additional decay path over the \( |s\rangle \) has to be included.

All in all, the information about the state is encoded in the first 1000 ns of the signal. The signal can be normalized by forming the ratio with the back part of the pulse and compute the area under the gained curves. The largest value corresponds to the case, where all population is in the \( m_s = 0 \) state, whereas the smallest value represents all population in the \( m_s = \pm 1 \) state. Intermediate values are achieved for mixed population states. With this technique it is possible to read out optically the state (populations) of the NV center while in the end a preparation in the initialized state \( m_s = 0 \) is the result.
2 Theoretical background for the nitrogen-vacancy center in diamond

2.4.1 Bloch sphere representation of the NV center states

To explain the dynamics of the NV center, a bloch sphere representation can be utilized. Here, the description of the NV is reduced to a two-level system (TLS). The concept of the Bloch sphere shall give a pictorial idea, how the state evolution of a TLS could be imagined.

Without loss of generality, the states with $|m_s = 1\rangle \equiv |1\rangle$ and $|m_s = 0\rangle \equiv |0\rangle$ are chosen to be the considered TLS. Then any quantum mechanical state in that system can be represented as

$$|\Psi\rangle = \alpha|0\rangle + \beta|1\rangle = \cos \frac{\theta}{2}|0\rangle + e^{i\phi}\sin \frac{\theta}{2}|1\rangle. \tag{2.18}$$

The state $|\Psi\rangle$ has to be normalized, fulfilling the condition $|\langle \Psi | \Psi \rangle|^2 = |\alpha|^2 + |\beta|^2 = 1$, which is validated by selecting $\alpha = \cos(\theta/2)$ and $\beta = \exp(i\phi)\sin(\theta/2)$. The special choice of $\alpha$ and $\beta$ becomes apparent in the definition of the Bloch Vector

$$R = \langle \sigma | \sigma \rangle = \langle \Psi | \sigma | \Psi \rangle = \begin{pmatrix} \sin \theta \cos \phi \\ \cos \theta \sin \phi \\ \cos \theta \end{pmatrix}, \tag{2.19}$$

which represents a point on the Bloch sphere in spherical coordinates, where $\theta$ is the azimuthal and $\phi$ the polar angle. $\sigma = (\sigma_x, \sigma_y, \sigma_z)^T$ denotes the vector containing the Pauli matrices as components.

Secs (2.18) and (2.19) imply that two parameters are needed to describe the state $|\Psi\rangle$. Hence, $\Psi$ is called a pure state and fulfills only in that configuration the purity condition $\text{Tr}\{\rho^2\} = 1$. Or stating it differently, if the vector $R$ does not lie on the Bloch sphere (due to dephasing processes of the state $\Psi$), 3 parameters are required to describe the vector$^1$. Consequently, such a vector will not fulfill the purity condition and would eventually be a mixed state. Additionally, if $|\Psi\rangle$ is a pure state, then $|0\rangle$ and $|1\rangle$ are eigenstates of the system.

An illustration of the Bloch sphere is given in Fig. 2.6. The positive z direction is defined as the $|m_s = 0\rangle$ state, whereas the negative direction is denoting one of the remaining spin states $|m_s = \pm 1\rangle$. The spin vector in Fig. 2.6 (red arrow) is chosen to be the initialized state of the NV center.

An important aspect of the Bloch sphere is the fact that the projection of the Bloch vector onto the z-axis represents the populations in the current spin state. The projected part is the normalized contrast different, which will be read out by the application of a laser (cf. Sec. 2.4). On the contrary, the position in the xy-plane of the Bloch vector is related to coherences. Thus, obtaining the coherence of the system requires a $(90^\circ)$ rotating of the Bloch vector towards the z-axis. Otherwise the projection on the z-axis could not be distinguished from a mixed state situation.

$^1$The third component was determined to be 1, by purity condition
2.5 Basic measurements

The basic measurements, performed in this subsection, emphasize some mayor implications for the NV center, and show for which approaches this system can be utilized.

2.5.1 Optically detected magnetic resonance.

A common technique to determine the Zeeman interaction or the zero-field splitting for the NV center is the optically detected magnetic resonance (ODMR), which is based on the principles of conventional electron spin resonance (ESR), but incorporates the optical readout character of the NV center. In an ODMR measurement, the fluorescence difference between the spin states are exploited (as demonstrated in Sec. 2.4) to detect a transition between the spin states.

If an externally applied microwave field is hitting the energy difference between the eigenstates, an effective population transfer can occur between the bright and the dark state (shown as a dip in Fig. 2.7 b). The contrast of the measurement is taken relative to the bright spin state, $m_s = 0$, and can be at maximum about 30%, reflecting the

---

2 The figures of the Bloch sphere, here and in the following, are based on the template from the Master thesis of Jochen Scheuer (2013). The Bloch sphere has been modified and adapted for particular purpose.
different spin dependent decay paths \[14\]. Fig. 2.7 a) depicts the impact of a static magnetic field with strength $|B| = B_z$ on the eigenstates of the system and Fig. 2.7 b) shows the measured fluorescence response at certain magnetic field values.

Moreover, Fig. 2.7 a) highlights some special features of the NV center which happen at the excited state level anti-crossing (ESLAC) at about 51.2 mT, and at the ground state level anti-crossing at 102.4 mT. At these points a state mixing between the nuclear and electron spins occur, inducing spin flip-flop processes. The transverse part of the nuclear hyperfine interaction leads to the exchange of electron and nuclear spin polarization, while the spin selective non-radiative decay of the electron spin is responsible for the maintenance of the electron spin polarization \[84, 85\]. Thus, the nuclear spin can be effectively polarized up to $> 98\%$ at the ESLAC \[84\] and $> 90\%$ at the GSLAC \[86\].

![Diagram](image.png)

**Fig. 2.7:** The impact of a static magnetic field and its measurement via ODMR. (a) Change of the energy eigenstates of the NV center under an increasing static magnetic field, aligned along the principal axis of the NV, lifting the degeneracy of the $m_s = \pm 1$ spin sublevels. The intersystem crossing points, denoted as ESLAC and GSLAC, can be utilized for efficient polarization of the hyperfine coupled nitrogen nuclear spin. (b) Recorded ODMR spectra at different magnetic field strength. While shining a laser onto the system an externally applied microwave source swept through frequencies in GHz regime and leads to a detection of transitions between the spin sub-levels by a change in contrast.
2.5.2 Rabi oscillations

The idea of the Bloch sphere can be used to describe the manipulation of the spin states with pulsed microwave fields. The manipulation of the NV center at its spin transition frequencies constitutes an electron spin resonance (ESR) measurement.

One basic sequence in pulsed ESR is the Rabi measurement. Here, on an initialized spin state, a microwave field for an increasing times $\tau$ is applied. The read out of the current spin state is a projection of the Bloch vector onto the z-axis of the system revealing the population in the spin state (cf. Sec. 2.4). It has to be stressed, that in any pulse sequence for the NV, the applied laser pulse is acting in the first 1000 ns as a readout followed by a polarization of the spin state (initialization to $m_s = 0$). If the applied microwave frequency $\nu_{\text{ODMR}}$ fits to the spin state transition frequency, then the resulting signal reveals periodic nutations, indicating the transfer or populations between the spin states, which depend on the time $\tau$ of the applied microwave and its amplitude.

In Fig. 2.8a) a general Rabi measurement is illustrated in a block sphere representation. By using such a scheme, Rabi oscillations between two spin states, as displayed in Fig. 2.8b), can be observed.

This basic measurement highlights the ability for the coherent manipulation of the NV

![Diagram of the Bloch sphere and Rabi oscillations](image)

Fig. 2.8: Coherent manipulation of the electron spin by observing Rabi oscillations. (a) Bloch sphere representation for the Rabi measurement with a simplified pulse scheme. On the initialized spin state $m_s = 0$ a microwave drive with a certain strength $\Omega$ is applied over a time $\tau$. Afterwards, the remaining population of the $m_s = 0$ state is read out by probing the fluorescence response with a laser. (b) Recorded Rabi measurement at 1.866 GHz illustrating the population transfer from the bright state (larger norm. fluorescence signal) to the superposition state ($\pi/2$-pulse) and finally to the dark state (corresponding to a $\pi$-pulse). After $2\pi$ the population is recovered again.
center spin state, which is a crucial ingredient for any type of quantum information processing and sensing application.

2.5.3 DC magnetometry

The spin state of the NV center has the ability to interact with magnetic fields. In a free induction decay measurement (FID), this interaction becomes most effective, if the spin state is prepared in the superposition state \( |\Psi\rangle = (|0\rangle + e^{i\phi_{\text{FID}}(\tau)}|1\rangle)/\sqrt{2} \), which is the xy-plane of the Bloch sphere. The coupling to (internal and external) magnetic fields, \( B(t) \), will alter the phase evolution of the spin eigenstates \( m_s = \pm 1 \) by a phase

\[
\phi_{\text{FID}}(\tau) = g\mu_B \int_0^\tau B(t) dt
\]

(2.20)
denoted as \( e^{i\phi_{\text{FID}}} |\pm 1\rangle \) in the superposition term. A perfect homogeneous and static magnetic field \( (B(t) = B_{\text{const}}) \) will rotate the spin state \( |\Psi\rangle \) with a constant angular frequency, depending on the B-field strength. As a result, a homogeneous phase accumulation by eq. (2.20) takes place, which will differentiate the initial state from the final one. With such a technique, an impact from a magnetic field on the sensor can be detected (see Fig. 2.9).

If the magnetic field, \( B(t) \), contains random noise fluctuation, \( \delta B(t) \), the accumulation of phase will also happen randomly, leading to a dephasing of the spin rotation and finally

\[
\text{Fig. 2.9: Bloch sphere representation of a Ramsey type measurement with a simplified pulse scheme. The initially prepared spin state (red arrow, first Bloch sphere) is rotated by a } \pi/2 \text{-pulse around the x-axis (second Bloch sphere) and evolves freely for a time } \tau \text{ in which the superposition state capture information about the environment in the phase } \phi, \text{ here in the } m_s = 1 \text{ state (third Bloch sphere). The second } \pi/2 \text{-pulse transfers coherence phase information to populations, which are read out optically. The corresponding pulse sequence is shown at the bottom.} \]
to a decrease of coherence. The intrinsic and extrinsic magnetic noise, spread over the whole frequency spectrum, will be measured by the Ramsey measurement scheme shown in Fig. 2.9.

In the Ramsey measurement, an initialized spin state $m_s = 0$ will be brought through a $\pi/2$-pulse in a superposition state. The evolution time, $\tau$, of the phase in that state is varied and finally another $\pi/2$-pulse converts coherences to populations in order to read out the state with a laser. In the analysis, each time value, $\tau$, is plotted against the read out normalized fluorescence signal. Such a measurement is depicted in Fig. 2.10.

The signal of the Ramsey measurement decays during the measurement $\propto \exp(-\frac{\tau}{T_2^*})^2$, if Gaussian noise is assumed. Thus, $T_2^*$ is denoted as the free dephasing time in a Ramsey, or free induction decay (FID) measurement.

Moreover, as the measurement incorporates intrinsic and extrinsic noise, information about the noise spectrum of static (and oscillating) magnetic fields in the range of $T_2^*$ can be obtained. Consequently, the graph in Fig. 2.10 highlights the potential of the NV center by showing its susceptibility to magnetic fields in a simple Ramsey measurement [87, 88].

Fig. 2.10: Free induction decay or Ramsey measurement of the hyperfine interaction to the $^{14}$N. (a) The ODMR transition between $m_s = 0$ and $m_s = 1$ state was detuned by 4MHz to reveal the energy levels of the hyperfine coupled $^{14}$N. The different energy levels induce a rate of change in population connected to their detuning of the drive. The orange envelope of the oscillation is a Gaussian fit revealing a free dephasing time of $T_2^* = (12 \pm 1)$ ps. (b) Fourier transform of the time signal in (a) shows the energy differences, $A_\parallel$, between the nuclear spin states of the nitrogen $^{14}$N (cf. eq. (2.6)).
2 Theoretical background for the nitrogen-vacancy center in diamond

2.5.4 AC magnetometry

Static magnetic field contributions impose a limitation on the coherent evolution of the NV spin states and limit the sensitivity of the sensor by the free dephasing time, $T_2^*$. At room temperature and low magnetic fields, the spin bath is not polarized, and all outer (and inner) contributions disturb the captured phase of the NV. Eventually, this leads to inhomogeneous broadening.

Inhomogeneous broadening can be compensated by effectively decoupling the NV center from slow noise contributions. This can be done by a technique proposed by Erdwin Hahn in 1950 [89]. The scheme applied on the NV center is illustrated in Fig. 2.11.

The rotation direction of the superposition state of the NV center (in the xy-plane of the Bloch sphere) does not change, if B is static. Hence, a phase of $\phi_{\text{FID}}$ is gained after a time $\tau/2$. If the Bloch vector is rotated by 180° (or a $\pi$-pulse) around the x-axis, then the phase evolution is “reverted”. Consequently, after another time $\tau/2$ an echo signal is observed indicating a refocus of the coherence.

It becomes obvious, that this kind of refocusing scheme can only revert the action of magnetic fields, $B_{\text{slow}}$, which are barely changing on the timescale of $\tau$. Magnetic field inhomogeneities and noise, $B_{\text{fast}}$, acting on shorter timescales than $\tau$ will still affect the coherence terms, which can be measured in the population difference of the final state.

Fig. 2.11: Bloch sphere representation of the Hahn Echo scheme with a simplified pulse scheme. An initially prepared state $|0\rangle$ (1. Bloch sphere) is brought into superposition state by an external $\pi/2$ microwave pulse (2. Bloch sphere) and evolves for a time $\tau/2$ (3. Bloch sphere). The $\pi$-pulse (4. Bloch sphere) refocuses the impact of slow magnetic fields (on the scale of $\tau$, 5. Bloch sphere) and reveals effects of fast magnetic field by a non-vanishing phase $\phi$. With the second $\pi/2$-pulse (6. Bloch sphere) the gained phase is transferred to populations which are read out optically. At the bottom a corresponding Hahn Echo pulse sequence is shown.
This idea can be expressed in a captured phase by:

\[
\phi_{\text{Hahn}}(\tau) = g\mu_B \int_0^{\tau/2} [B_{\text{slow}}(t) + B_{\text{fast}}(t)] \, dt - g\mu_B \int_{\tau/2}^{\tau} [B_{\text{slow}}(t) + B_{\text{fast}}(t)] \, dt
\]

\[
B_{\text{slow}}(t) \approx B_{\text{const}} = g\mu_B \left( \int_0^{\tau/2} B_{\text{fast}}(t) \, dt - \int_{\tau/2}^{\tau} B_{\text{fast}}(t) \, dt \right),
\]

where the part with \( B_{\text{slow}}(t) \) is suppressed.

A typical Hahn Echo measurement is shown in Fig. 2.12, where a decay with the time constant \( T_2 \) was fitted to the measured data. Therefore, \( T_2 \) denotes the typical timescale of the Hahn Echo measurement, which is, compared to \( T_2^* \) in Fig. 2.10, usually three order of magnitudes larger. This shows, that the refocusing \( \pi \)-pulse suppresses slow fluctuations in the noise spectrum and extends thereby the coherence time.

![Hahn Echo at \( \nu_1 = (1864.807 \pm 0.020) \text{ MHz} \)](image)

**Fig. 2.12:** Hahn-Echo measurement of a NV. Both traces represent the decoherence process described by \( T_2 \). The difference between the traces constitutes in the last \( \pi/2 \) pulse of Fig. 2.11. For the lower magenta measurement it was replaced my a \( 3\pi/2 \) pulse (cf. Fig. 2.8b) yielding a projecting to the dark state. This way of representation is beneficial in order to identify the normalized decoherence level (here about 0.85) to which both states are decaying to. Both fits to the data have utilized the fact that the signal is \( \propto \exp\left(-\frac{(\tau/T_2)^2}{2}\right) \), where uncorrelated Gaussian noise was assumed.
Hence, the Hahn Echo sequence illustrates one of the simplest measurement techniques to decouple the NV center from slow noise and to measure (weak) fast oscillating signals.

The coherent driving of the NV center, combined with its interactions to magnetic fields, opens the doors for a large series of pulsed [90–97] and continuous microwave drive protocols [98–101]. Those protocols aim at shaping the interaction with the NV center and its environment, and try to prolong simultaneously the coherence time of the sensor by decoupling it from certain noise sources [102–105]. These possibilities make the NV center a valuable measurement device to sense all kind of magnetic fields, ranging from static DC to high frequency signals.

2.6 Relaxation mechanisms - the spin bath

In general, it is not easy to distinguish the impact of surrounding magnetic fields on the NV center in terms of interaction strength, spacial origin, direction, or type of interaction process. However, a possible way for a classification might be a separation in characteristic timescales detected in a certain state configuration of the sensor. I.e. the focus is on the components of the Hamiltonian (coherences or populations) which are affected in the considered timescale in a given measurement basis.

Relaxation (affecting the populations of a quantum state) is the phenomenon, which describes the modification of a system towards its (thermal) equilibrium state. A dephasing process on the other hand relates to incoherent coupling of the quantum state of the NV to its surroundings (therefore affecting coherences of the state). In both cases a transfer of energy (or alternatively entropy) occurs and the coupling to the environment is the essential feature enabling this process. The way how a system relaxes or dephases reveals additional information about the interaction effects of the environment on the NV center.

The decay mechanism can be generally understood as an exponential decrease of the (normalized) fluorescence signal [76, 106] of the NV center

\[
\text{Signal}(t) \propto \exp \left( -\sum_i^N \left( \frac{t}{T_i} \right)^{\alpha_i} \hat{e}_i \right).
\]  

(2.22)

The various types of decay mechanisms are labeled with \(N\). For a particular type of decay mechanism, \(i\), a characteristic time, \(T_i\), with the corresponding decay constant, \(\alpha_i\), acting in the basis, \(\hat{e}_i\), can be found. Here it becomes apparent that it matters in which measurement basis, \(\hat{e}_i\), the fluorescence is measured as only populations are read out by the laser (otherwise, coherences have to be transfered to populations).

In this thesis, eq. (2.22) is simplified in four decay mechanisms

\[
\approx \exp \left( -\hat{e}_{T_1} \left( \frac{t}{T_1} \right)^{\alpha_{T_1}} - \hat{e}_{T_2} \left( \frac{t}{T_2} \right)^{\alpha_{T_2}} - \hat{e}_{T_3} \left( \frac{t}{T_3} \right)^{\alpha_{T_3}} - \hat{e}_{T_4} \left( \frac{t}{T_4} \right)^{\alpha_{T_4}} \right),
\]  

(2.23)
where each of them is acting on a different time scale $T_1, T_2^*, T_2$ and $T_\Omega^2$ and in a different basis configurations. Two of the decay mechanisms were already presented in Sec. 2.5.3 and Sec. 2.5.4, namely $T_2^*$ and $T_2$. Two remain to be explained.

The relaxation process with the characteristic time $T_1$ is referred to as the spin-lattice relaxation time denoting the life time of a quantum state. To obtain $T_1$, the NV center is simply polarized in one of its eigenstates, e.g. in $m_s = 0$. After a time $\tau$ the remaining population of the state is read out with a laser and compared to the initial state. During the time $\tau$ the spin state starts to relax to its unpolarized state. The relaxation process is mainly determined by the coupling to phonons in the diamond lattice.

The exponential coefficient for the $T_1$ decay process can be chosen by $\alpha_{T_1} = 1$, as it can be related to a spontaneous decay principle. Or in another way, this reflects the assumption for a Lorentzian noise spectrum with fast bath dynamics [76] (note, the Fourier transform of an exponential function is a Lorentzian which describes in this case the noise spectrum). Due to the presence of a small phonon contribution at room temperature (originating from the high Debye temperature of the diamond [41, Tab. 19, p. 2.50]), a low spin-phonon interaction occurs, resulting in life times on the order of ms.

The explanation for the presence of predominantly fast dynamics in the noise spectrum for $T_1$ noise can be deduced from the large ZFS parameter ($D = 2.87 \text{GHz}$). The spin states $m_s = 0$ and $m_s = \pm 1$ are essentially protected against noise components smaller than the energy gap, $D$. Hence, $T_1$ covers on its characteristic timescale the effect of high frequency noise and the $T_1$ relaxometry measurement is a method to probe high frequency noise components of the environment with a $T_2^*$ limited bandwidth [107–109]. A considerable improvement in bandwidth, towards $T_2$, will be presented in Chapter 5.

The last decay process to mention is $T_\Omega^2$, which describes for instance the coherence time in a Rabi measurement. In essence, this timescale expresses how long Rabi oscillations are maintained under an applied drive field, $\Omega$. Exemplary measurements of $T_\Omega^2$ can be seen in Chapter 5. In this case the value of $\alpha_{T_\Omega^2}$ will be entirely determined by the noise spectrum of the signal generator producing $\Omega$.

All in all, the choice of $\alpha_i$ is not always intuitive and the characteristic times, $T_i$, depend drastically on the environment of the NV center and its measurement basis. One hint about the noise distribution can be obtained by measuring the noise spectral density, $S(\omega)$, which links the amount of noise gained at a particular frequency, $\omega$. It has to be stressed, that different types of measurements (Rabi, FID, Hahn-Echo,...) emphasize different parts of the noise spectral density. Therefore, without the knowledge of $S(\omega)$, it is hard to fix $\alpha_i$ for a characteristic time, $T_i$. 


Measurement realization

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An experiment which is not performed is not an experiment.

— John Archibald Wheeler
3 Measurement realization

3.1 Setup

The core approach in imaging and identifying single NV centers is based on a home-build confocal microscope setup. This gives the ability to address optically individual NV centers and collect their fluorescence response after being manipulated by various external fields. Fig. 3.1 illustrates the schematics of the setup, with its various stages. In the following, the individual parts of the setup will be introduced.

![Fig. 3.1: Schematics of a standard room-temperature confocal setup including signal sources and data acquisition devices for the NV magnetometry. The different parts in the illustrations are grouped in boxes to separate their functionality and to structure their explanation in the main text. Arrows denote the predominant information flow direction in a connection. Abbreviations used in this figure and their meaning: Spec. A. = spectrum analyzer to record the transmitted signal, AOM = acousto-optical modulator, P1 and P2 = pinholes, F = low pass filter for red light, Struc. = structure for RF (radio-frequency) and MW (microwave) fields, Signal Gen. = set of various signal generators, AMP = set of various amplifiers.]

3.1.1 Laser chopping and AOM, part I

The excitation and illumination for the setup was provided by a 532nm Nd:YAG frequency doubled laser. A lens focuses the beam into an acousto-optic modulator (AOM), which serves as a fast light shutter. The AOM consists basically of an transparent medium, usually a quartz crystal, creating a Bragg cell at which the incident light it diffracted. A piezo-electric transducer, attached to the crystal, converts a 110MHz electrical signal into an acoustic wave which propagates through the crystal. This wave
causes a periodic change in density and refractive index, creating a Bragg diffraction grating. The amount of diffracted light depends on the intensity of the sound wave, and eventually on the driving signal, and can be controlled with that. The frequency of the electro-magnetic signal determines the distance between the Bragg-diffraction planes, thus, changing the angle of diffraction.

The diffraction efficiency and the switching speed for the AOM depend strongly on the beam waist diameter. Increasing the beam waist diameter by choosing a lens with longer focal length will increase the diffraction efficiency [110]. However, an increased beam diameter will decrease the switching speed of the AOM, as the supersonic wave requires more time to intercept a larger diameter than a smaller one. Hence, the focal length needs to be chosen carefully to balance diffraction efficiency and switching speed.

The second lens collimates the laser beam again and the iris is used to filter the desired diffraction order.
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3.1.2 Beam cleaning, part II

The laser beam will be distorted after the AOM part and will not represent anymore a clean TEM\(_{00}\) mode, but will also contain higher order modes. To ensure a uniform TEM\(_{00}\) mode, which represents an ideal Gaussian beam, all the higher order modes causing an imperfect plane wave have to be suppressed. This can be done by several approaches, but the underlying physical idea is the same and describes a spatial filter.

![Fig. 3.3: Schematics for an alternative approach to clean and widen the transverse light mode with a fiber. The laser light is coupled into a fiber via an objective situated on a xyz stage. The out-coupling objective creates a beam with larger diameter, where a high-pass filter suppresses the red fluorescence of the fiber. The tunable \(\frac{\lambda}{2}\) plate after the objective will be used (like in Fig. 3.1) to adjust the polarization of the beam to the dipole field of the NV. FC = fiber coupling objective.](image)

Putting a pinhole at the focal plane of a lens, which focuses the beam, will act as a spatial filter by cutting away all higher order contributions, which are situated further away from the focal spot. The pinhole with diameter of about 50\(\mu\)m could be replaced by a fiber with an equivalent fiber core diameter, as shown in Fig. 3.3.

The advantage of using a fiber is the gained flexibility, i.e. altering the alignment in the AOM part would not require to adjust the part following the fiber out-coupler. However, the fiber coupling approach can also have its disadvantages as the coupling efficiency into the fiber is highly dependent on the quality of the fiber surface, the objective used for the in-coupling and the correctly adjusted polarization (accounted by the \(\frac{\lambda}{2}\) plate before the fiber in-coupling objective). Moreover, a fiber can add red fluorescence contributions to the green light, which requires a red filter at the fiber coupling output.

To ensure a good signal to noise ratio in the detected fluorescence signal, it will be important to overlap the excitation modes of the incident green beam on the NV center with the collection mode of the red fluorescence response from it. This can be done by guaranteeing a full and central illumination of the focusing objective. Hence, the beam has to be widened, which is done in Fig. 3.1 by a inverse telescope configuration or in Fig. 3.3 by adjusting the out-coupling objective accordingly.

A tunable \(\frac{\lambda}{2}\) plate placed somewhere before the (main) objective will help to adjust the polarization of the incident beam to fit it best to the dipolar character of the excitation.
mode of the NV. The diamond lattice provides 4 possible orientation for the NV center, thus, adjusting the polarization will eventually maximize the fluorescence signal.

### 3.1.3 Sample illumination and detection, part III

The last part in the optical path will perform the actual confocal measurement. A dichroic mirror will guide the laser beam into the objective, which is attached (together with the dichroic mirror) to a three dimensional piezo scanner. The laser beam will illuminate a diffraction limited area in the diamond and the red fluorescent response is collected together with the reflected green light by the same objective. This should produce ideally a collimated beam facing towards the dichroic mirror, where most of the green light will be reflected and only red light will transmit.

The detection path aims to resolve the spatial dependent intensity profile of the red light by suppressing all light beams which are not originating from the same focal spot. The spatial filter in this configuration is again a pinhole, placed at the focal point of the lens which separates the spatial components of the light beam in its focal plane. Since the pinhole selects light components that originate from the same focal plane this method is termed confocal (= with the same focus) imaging.

The second lens in the detection box will solely image the pinhole onto the avalanche photo-diode (APD), which transforms the collected amount of photons into an electric signal recorded by a counter. The green filter in the detection box is very crucial as the red light intensity is orders of magnitude lower than the green one, and the APD will not differentiate between the photon frequency. Hence, it has to be ensured that just the red light will arrive at the detector and all parasitic light is removed by placing the detection path in an enclosed box.

### 3.1.4 Sample holder, part IV

The diamond sample is glued with the top side facing a glass slide, on which a gold structure of strip lines is lithographically deposited. To ensure a smooth transition in refractive index from diamond to glass, a small drop of immersion oil is deposited between the glass slide and the diamond. A small amount of super glue is also added to the edges of the diamond to attach it to the glass to avoid polluting the index matching immersion oil. Fig. 3.4 show the glass slide without (a) and with the diamond sample (b).

The glass slide with the 4 strip lines is glued onto a larger printed circuit board (PSB) microwave structure hosting the SMA connectors, where the cables carrying the microwaves are attached. The connection between the glass slide structure and the outer PCB is established through soldering. The PCB board is mounted on a home-build micrometer xyz-stage like depicted in Fig. 3.5, which is used for a coarse positioning of the diamond sample (with micrometer precision).
3 Measurement realization

**Fig. 3.4:** Glass slide with the deposited gold structure and the attached diamond. (a) The bare glass slide shows a tapered microwave structure forming at its center straight strip lines with a distance of about 20 µm. (b) Onto the strip lines a diamond sample is mounted to that. The very close proximity of the strip lines to the diamond surface (and eventually to the NVs) ensures a strong coupling of an applied microwave field to the NV center. The diamond (40 µm 99.8% $^{12}$C low-N grade layer on 98.9% $^{12}$C electronic grade substrate) shown here was mostly used for the measurements in this work. The surface contains laser engraved numbers as landmarks.

A piezo stage with nanometer precision, holding and manipulating the objective, is mounted at the back frame of the micrometer stage (which can be seen in Fig. 3.2 b). The recorded fluorescence intensity of the APD will be mapped to a xyz position of the piezo stage enabling the scanning confocal imaging. A confocal scan is illustrated in Fig. 3.6, where individual NV spots are marked by circles. The strip line structure, which is shown in the photograph in Fig. 3.4 can be also identified on the confocal xy scan.

In Fig. 3.6 two triangle markers, one on each axis, represent the position of one selected NV in the xy scan. At this position a depth scan in xz direction was performed, which is shown in Fig. 3.7 a). Another closer depth scan around one of the NV centers was performed in Fig. 3.7 b), where the point spread function (PSF) of the NV center can be clearly seen. It becomes evident, that the PSF appears to be slightly tilted in the presented picture identifying a small misalignment in the pinhole.

3.1.5 Signal sources and data acquisition, part V

The last stage consists of the data acquisition and signal generation part and represents the brain of the experiment. All the control is realized by a PC, which interacts directly with various devices dedicated for specific operations. The coordinated and sequential control of the measurement procedure and the live processing of the data is an integral
Fig. 3.5: Image of the home-build xyz micrometer state holding on its front side the PCD board with the diamond sample illuminated by the green laser. A magnet contained in a white 3D-printed holder, mounted on a movable xyz phi stage is used to accurately control and alter the static magnetic field applied on the NV center. The microwave field is transported from the left side via the blue (SMA) cables. The transmitted microwave field after the sample can be monitored with the right SMA connectors.

and important part of the setup configuration. For this purpose, a very general software suite was written, enabling all the required tasks. A detailed presentation of the core ideas of this framework and the large potential behind the approach is given in Chapter 4.
Fig. 3.6: Large range confocal xy scan of the diamond sample in Fig. 3.5, 1 µm below the diamond surface at about 40 mT. With a piezo scanner (shown as a black aperture, mounted at the back of the micrometer stage in Fig. 3.2), the objective was moved resulting in a diffraction limited confocal scan of the diamond at a certain depth. Some investigated NV centers with the same orientation (showing ODMR at similar frequencies) are marked with cyan circles, other orientations (without ODMR signal) are marked with green circles.
3.1 Setup

Fig. 3.7: Confocal depth scans in the xz plane. (a) Larger xz scan at about \( y = 2 \, \mu m \) to identify the different layers. The utilized diamond contains low amount of impurities and therefore it is almost non-fluorescing. The laser light becomes scattered at the oil molecules thus appearing much brighter in the scan. The glass substrate in the button contains just a few point-defect like structures where light is scattered at. It has to be noted that the x and z axes have different scale. (b) Detailed depth scan (xz) of one arbitrary chosen NV to illustrate its emission pattern resembling a point spread function.
3 Measurement realization

3.2 Magnetic field alignment by fluorescence

The application of a static magnetic field will alter the (Zeeman) separation of eigenstates of the spin Hamiltonian given in eq. (2.15) and result in specific transitions measured in the ODMR (cf. Fig. 2.7). However, the magnetic field lines of an external magnet have to be aligned to the principle axis of the NV. Missing this requirement would create different eigenstates, where \( m_s \) is no longer a good quantum number. Thus, the states become mixed. As a consequence, the optically-Induces spin-polarization and spin-dependent fluorescence of the NV center decrease drastically with a larger off-axial magnetic field [33, 81, 82].

To ensure a high ODMR contrast and a well defined fluorescence signal, the magnet has to be aligned to the NV axis. A typical measurement outcome of such an alignment is depicted in Fig. 3.8, where the optimal magnet position is found at maximum fluorescence.

The magnetic field alignment constitutes to be a preliminary calibration for any spin and

![Image](image.png)

**Fig. 3.8:** Record the fluorescence signal as a function of magnet position. The magnet, shown in Fig. 3.6, was moved parallel to the sample holder and at each magnet position the focus of the confocal scan was at first optimized for the NV position to account for small drifts followed by the actual fluorescence measurement. Fitting a two dimensional Gaussian function to the picture yields the optimal magnet position aligned to the principle axis of the NV resulting in a maximal fluorescence signal.
fluorescence dependent measurement. The magnet from the setup schematics in Fig. 3.1 is shown enclosed in a white magnet holder in Fig. 3.5. Moreover, Fig. 3.2 b) gives a view on the front part of the magnet. The magnet holder is attached to a rotation stage, which in turn is fixed to a long-distance traveling xyz stage with micrometer precision. This allows a very flexible positioning of the magnet and enables automatized scans to determine the optimal position.

A misalignment of the static magnetic field near the excited state level anti crossing [84] has a much stronger effect on the fluorescence of the NV center as the spin mixing dynamics in the excited state affects the optically-Induces spin-polarization to a much larger extend. Therefore, encoding the magnet position with the fluorescence signal of the NV center will result in fast alignment procedure.

3.3 Sampling as a powerful measurement tool

The sampling theorem, implied by Harry Nyquist in 1928 [111] and finally proven by Claude E. Shannon in 1949 [112], is a well known criterion to measure (reconstruct or [over]sample) a frequency, \( \nu \), unambiguously, without invoking any prior information. To ensure this, a signal must be sampled at a rate, \( \nu_{\text{sample}} \), greater than twice its maximum frequency component, \( \nu_{\text{max}} \). Or more generally,

\[
\nu_{\text{sample}} > 2 \cdot BW,
\]

where \( BW = \nu_{\text{max}} - \nu_{\text{min}} \) denotes the bandwidth of the present signal. If the full spectrum of the signal is of interest, \( \nu_{\text{min}} \) becomes zero.

Violating the Nyquist-Shannon criterion will result in the appearance of artifacts in the sampled data, which are known as aliasing. The name of the term originates from the Latin word \textit{alias}, being the feminine accusative plural of \textit{alius}, meaning “other”, “also” or “else”. Hence, the measured spectrum will contain “other” spectral components not belonging to the real spectrum, but rather appearing as low-frequency images, \( \nu_{\text{image}} \), of the actual frequencies, \( \nu_{\text{actual}} \). Those image frequencies can only be smaller than the sample frequency, \( \nu_{\text{sample}} \), (otherwise eq. (3.1) would hold) and become eventually indistinguishable to real low frequency components in the spectrum. They occur at

\[
\nu_{\text{image}} = \nu_{\text{actual}} - n \cdot \nu_{\text{sample}} \quad n \in \mathbb{N},
\]

where \( n \) can be associated as the number of harmonics for different sample frequencies, \( \nu_{\text{sample}} \), producing the same image frequency, \( \nu_{\text{image}} \). Again, eq. (3.2) holds only if the eq. (3.1) is not fulfilled. In this context, the actual frequency \( \nu_{\text{actual}} \) is said to be undersampled (and in the opposite case oversampled).

Although, undersampling produces artifacts in the spectrum, it can also be exploited as a powerful technique to significantly reduce the amount of measurement time. In
the present work (for Chapter 5 and Chapter 6), undersampling was mainly used to identify the persistence of one or two oscillating signal, i.e. to determine their respective coherence times. Since the coherence time can be easily several magnitudes larger than the oscillating period of the frequency, it would take great efforts to resolve both parameters in one measurement. Instead, separate measurements of the frequency (by oversampling) and the coherence time (by undersampling) would require way less resources yielding eventually the same information content.

Given eq. (3.2) a simple measurement algorithm can be constructed to undersample a frequency at a calculated sample rate, $\nu_{\text{sample}}$, to obtain a desired image frequency, $\nu_{\text{image}}$, which is sampled with $N_p$ data points. 

This algorithm was intensively used throughout the present work and is therefore noteworthy. Apart from that, basic limitation for undersampling will be reveal. 

The three input parameters, the actual/real frequency, $\nu_{\text{actual}}$, a desired image frequency, $\nu_{\text{image}}$, and the number of points per period of the image frequency, $N_p$, should yield a sample frequency, $\nu_{\text{sample}}$, which satisfy the input parameters. However, it will be shown, that the input parameters are not completely independent of each other and relations will be established to constraint the choices of the input parameters. These constrains are crucial to know for the implementation of a general undersampling algorithm. 

At first, an initial sample frequency is obtained by

$$\nu_{\text{sample, init}} = \nu_{\text{image}} \cdot N_p, \quad (3.3)$$

which does not have to be the final sample frequency, since the correct harmonic number $n$ has to be chosen first. From eq. (3.2) the correct number of harmonics is inferred as

$$n = \text{Integer} \left( \frac{\nu_{\text{actual}} - \nu_{\text{image}}}{\nu_{\text{sample, init}}} \right). \quad (3.4)$$

If the inner term in eq. (3.4) results to be smaller than 1, then $n$ has to be forced to be at least $n = 1$. Apart from that, the integer operation will always round down the inner value, which is different to the usual rounding technique, which is later shown to be beneficial. Afterwards, the correct sample rate can be calculated by

$$\nu_{\text{sample}} = \frac{\nu_{\text{actual}} - \nu_{\text{image}}}{n}, \quad (3.5)$$

which has to be used to reproduce the desired image frequency, $\nu_{\text{image}}$, in the sampled data, where the image frequency contains about $N_p$ points per period.

It has to be noted, that the resulting number per period will not be exactly the desired $N_p$, since $n$ has to be an integer value and the mismatch has to be compensated, e.g. by $N_p$. By inserting the corrected sample rate from eq. (3.5) into eq. (3.3), the new number

$$N_{p,\text{real}} = \frac{\nu_{\text{actual}} - \nu_{\text{image}}}{n \cdot \nu_{\text{image}}} \quad \forall N_{p,\text{real}} \in (2, N_{p,\text{max}}], \quad (3.6)$$
3.3 Sampling as a powerful measurement tool

will usually be close to its anticipated value, if the inner term in eq. (3.4) will be ≥ 1. Otherwise \( N_{p,\text{real}} \) will differ significantly from \( N_p \). The round bracket in eq. (3.6) indicate that 2 is excluded from the interval, whereas the square bracket includes the number \( N_{p,\text{max}} \) in the interval.

Hence, by exploiting different harmonics by the number, \( n \), which reproduce the same image frequency, it becomes feasible to fulfill simultaneously eq. (3.2) and to the best possible extend the value for \( N_{p,\text{real}} \). Moreover, the rounding down operation will always select a smaller \( n \) before a larger one leading in eq. (3.6) to an equal or larger \( N_{p,\text{real}} \) making the spectral resolution of \( \nu_{\text{image}} \) rather better than worse.

Before discussing the bounds of eq. (3.6), it has to be mentioned that it is also possible to account for the mismatch of \( n \) by keeping \( N_p \) at the desired value and instead, adjusting the image frequency to

\[
\nu_{\text{image}, \text{real}} = \frac{\nu_{\text{actual}}}{N_p \cdot n + 1} \quad \forall \nu_{\text{image}, \text{real}} \in [\nu_{\text{image}, \text{min}}, \nu_{\text{image}, \text{max}}].
\] (3.7)

But this will also result in a different sampling rate, calculated in eq. (3.5)

From these basic calculations, it becomes obvious, that the parameter \( n \) imposes a certain constrain resulting either in the dependency \( N_{p,\text{real}}(\nu_{\text{image}}) \) or \( \nu_{\text{image}, \text{real}}(N_p) \). \( N_{p,\text{real}} \) has to lie within the denoted interval \((2, N_{p,\text{max}}]\), where it can take in principle any rational number. The interpretation of having \( N_{p,\text{real}} = 2.5 \), for instance, means that 5 sample points are distributed equally on two periods of an image frequency. Or more general speaking, the image frequency, \( \nu_{\text{image}} \), is sampled at a rate

\[
\nu_{\text{image}, \text{sampling}} = N_{p,\text{real}} \cdot \nu_{\text{image}} .
\] (3.8)

Consequently, the value 2 in the interval bound for \( N_{p,\text{real}} \) in eq. (3.6) simply implies, that more than 2 points per image frequency period, \( \lambda_{\text{image}} = 1/\nu_{\text{image}} \), has to be guaranteed to sample reliably the image frequency. That is again the very same requirement formulated in eq. (3.1), but this time applied on the correct sampling of the image frequency, \( \nu_{\text{image}} \).

As a further implication, the lower bound in eq. (3.6) dictates the largest image frequency obtainable by undersampling. By starting from \( N_{p,\text{real}} > 2 \), eq. (3.6) recasts into

\[
\nu_{\text{actual}} > (2n + 1)\nu_{\text{image}} ,
\] (3.9)

where the smallest possible value for \( n = 1 \) requests \( 3 \cdot \nu_{\text{image}} > \nu_{\text{actual}} \). An image frequency larger than this value cannot be reliably obtained by the sampling theorem in general.

From these cross-relations in eqs (3.6), (3.7) and (3.9), the fixed value of the actual frequency, \( \nu_{\text{actual}} \), and the algorithm described by eqs (3.3) to (3.5), two scenarios can be deduced:
3 Measurement realization

- In the first case, the desired number of points per period, \( N_p \), is chosen with the limitation of \( N_p > 2 \) which will require the image frequency to be \( \nu_{\text{image}, \text{real}}(N_p) \).

Then, according to eq. (3.6) the image frequency has to be

\[
\nu_{\text{image, real}} < \frac{\nu_{\text{actual}}}{3} = \nu_{\text{image, max}},
\]

(3.10)

and it is guaranteed that with eq. (3.10), eq. (3.9) will always hold. This can be verified by replacing \( n \) in eq. (3.9) by eq. (3.6). In principle, there exists no limitation for the lower bound of the image frequency, but since a signal will decay eventually, the lifetime (or coherence time) will limit \( \nu_{\text{image, min}} \).

- In the second case, \( \nu_{\text{image}} \) is chosen first under the constraint that the image frequency can be at maximum \( \nu_{\text{image}} < \nu_{\text{actual}}/3 \). This will lead to an \( N_{p,\text{real}}(\nu_{\text{image}}) \) and the possible number of points per period can be within the range

\[
N_{p,\text{min}} = 2 < N_{p,\text{real}} \leq \left( \frac{\nu_{\text{actual}}}{\nu_{\text{image}}} - 1 \right) = N_{p,\text{max}}
\]

(3.11)

Otherwise, \( n \) will need to take a value smaller than 1 to fulfill the inequality above, which is forbidden by eq. (3.2).

Since \( N_{p,\text{real}}(\nu_{\text{image, real}}) \) has a definite lower (upper) bound, valid undersampling of the resulting image frequency can always be achieved. The choice which case to use, depends on the requirements on the parameters, \( N_p \) and \( \nu_{\text{image}} \). In the present work, the second case was preferred, since the expected image frequency served as additional confirmation that the routine works as expected and that \( \nu_{\text{actual}} \) was measured correctly. By choosing \( \nu_{\text{image}} \ll \nu_{\text{actual}} \) a decent number of point per image period, \( N_p \), could be ensured.

Prior to any long undersampled coherence time measurement a short oversampled measurement will be performed to obtain the value for \( \nu_{\text{actual}} \). Consequently, the mismatch of the expected and actual measured image frequency will give additional insight on how well \( \nu_{\text{actual}} \) was determined and how large the associated error bar for the measurement of \( \nu_{\text{actual}} \) should be.

A demonstration of the undersampling technique is given in Fig. 3.9, where a decaying oscillating signal. It has to be stressed that the width and the amplitude of the Fourier transformed signals in Fig. 3.9 (b) and (c) are almost identical which will lead to a correct estimation for the coherence time. Therefore, the main advantage of the undersampling approach is the fact, that the fit of an decaying oscillating signal is more robust and yields a smaller uncertainty for the decaying parameter compared to a signal which is solely decaying at a certain rate.

However, the drawback of undersampling becomes evident, when more than one frequency is present. If the difference of two frequencies, \( \Delta \nu = \nu_2 - \nu_1 \), \( (\nu_2 > \nu_1) \) is larger than three times the selected image frequency, \( \nu_{\text{image}} \), then \( \Delta \nu \) will create aliasing effects.
3.3 Sampling as a powerful measurement tool

**Fig. 3.9**: Demonstration of undersampling. (a) The blue curve was generated by the function \( a(t) = A \sin(2\pi \nu t) \exp(-t/T_2) \), with the parameters, \( A = 1 \), \( T_2 = 1 \mu s \) and \( \nu = 20 \) MHz. This curve was undersampled (with red points) to obtain a desired image frequency of \( \nu_{\text{image}} = \nu/10 = 2 \) MHz. Following the algorithm in eqs (3.3) to (3.5), and by choosing \( N_p = 9 \) a sample rate of \( \nu_{\text{sample}} = 18 \) MHz was determined to produce the image frequency. (b) Fourier amplitude spectrum of the undersampled signal. (c) Fourier amplitude spectrum of the original signal.

Fig. 3.9: Demonstration of undersampling. (a) The blue curve was generated by the function \( a(t) = A \sin(2\pi \nu t) \exp(-t/T_2) \), with the parameters, \( A = 1 \), \( T_2 = 1 \mu s \) and \( \nu = 20 \) MHz. This curve was undersampled (with red points) to obtain a desired image frequency of \( \nu_{\text{image}} = \nu/10 = 2 \) MHz. Following the algorithm in eqs (3.3) to (3.5), and by choosing \( N_p = 9 \) a sample rate of \( \nu_{\text{sample}} = 18 \) MHz was determined to produce the image frequency. (b) Fourier amplitude spectrum of the undersampled signal. (c) Fourier amplitude spectrum of the original signal.

on the image frequency itself (a consequence from eq. (3.9)). Thus, the whole situation becomes more complicated and can create unpredicted artefacts.

Instead, another approach can be used to measure the coherence time of \( \nu_1 \) without the necessity of oversampling \( \nu_2 \) and \( \nu_1 \) for this measurement. That means, by sampling \( \nu_2 \) at a rate \( \nu_{\text{sample}} = \nu_2/k \), the frequency contribution of \( \nu_2 \) is removed from the spectrum and the second frequency, \( \nu_2 \), reveals as the beating frequency, \( \Delta \nu \), in the obtained data. Eventually, the choice of the harmonic number, \( k \), is also bound by eq. (3.1)

\[
\nu_2 - \nu_1 = \Delta \nu \\
2\Delta \nu < \frac{\nu_2}{k} \quad \forall k \in \mathbb{N}
\] (3.12)

The same approach can be applied on \( \nu_1 \), or on other frequency contribution if eq. (3.12) is satisfied.

In Fig. 3.10 such a situation with two frequency components are shown. By applying the “out-sampling” technique, one frequency component can be removed from the spectrum and the result will show just the beating between the two frequencies. Here, every second period was sampled, which reduces significantly the measurement time for the whole time trace to extract the coherence time parameter. The number of points can be even further reduces which would yield the same information content.

A more advanced approach constitutes the combination of the two presented techniques, undersampling and “out-sampling” resulting in “down-sampling” of the actual frequency
3 Measurement realization

Fig. 3.10: Demonstration of “out-sampling”. (a) The blue curve was generated by the function $a(t) = (A_1 \sin(2\pi\nu_1 t) + A_2 \sin(2\pi\nu_2 t)) \exp(-t/T_2)$, with the parameters, $A_1 = 1$, $A_2 = 0.5$, $T_2 = 1$ µs, $\nu = 20$ MHz and $\nu = 18$ MHz. Following eq. (3.12) and by choosing $k = 2$ a sample rate of $\nu_{\text{sample}} = \nu_2/2 = 10$ MHz was determined (red points) to reduce the spectrum by $\nu_2$. (b) Fourier amplitude spectrum of the “out-sampled” signal. (c) Fourier amplitude spectrum of the original signal $a(t)$.

spectrum. In order to guarantee a valid result, all the denoted constraints (eqs (3.6), (3.9) and (3.10) for the first scenario and eqs (3.7), (3.9) and (3.11) for the second one) have to be satisfied. Fig. 3.11 demonstrates the “down-sampling” of three very close frequency components to their respective image frequencies. The selected image frequency $\nu_{\text{image}}$ for this scenario has to be a multiple of one of the actual frequencies $\nu_i = k\nu_{\text{image}}$. Then, related to the selected frequency, $\nu_i$, the peaks of the other frequencies will appear with the same distance like in the original spectrum.

The last examples illustrates a rather extreme case for the application of sampling techniques, and in most of the use cases for this work, either undersampling or out-sampling was pursued. But beyond that, the potential power of the correct choice of the sampling method becomes evident. Although the presented methods have a simple underlying principle, it must be adhered to the derived limits, to ensure correct undersampling.
Fig. 3.11: Downsampling, a combination of undersampling and “out-sampling”. (a) The function \( a(t) = \sum_i^m A_i \sin(2\pi \nu_i t) \exp(-t/T_2) \), with the parameters, \( m = 3 \), \( A_i = [1, 0.5, 0.75] \), \( T_2 = 1 \mu s \) and \( \nu_i = [18, 20, 22] \) MHz generate the blue curve. This curve was downsampled (with red points) to obtain a desired image frequency of \( \nu_{\text{image}} = \nu_2/5 = 4 \) MHz. Following the algorithm in eqs (3.3) to (3.5), and by choosing \( N_p = 4 \) a sample rate of \( \nu_{\text{sample}} = 16 \) MHz was determined to produce the image frequency. (b) Fourier amplitude spectrum of the down sampled signal. (c) Fourier amplitude spectrum of the original signal. Note that zeropadding was performed before FT to smooth the spectrum.
CHAPTER 4

Qudi: A modular python suite for experiment control and data processing

Technology is a useful servant, but a dangerous master.

— Christian Lous Lange
4 Qudi: A modular python suite for experiment control and data processing

4.1 Introduction

Experiments on quantum mechanical systems are complex. They typically require the fine control of multiple hardware and precise sequencing of operations. Dedicated software is necessary to control it all and to perform specific tasks. The problem is general - all research groups face increasing complexity of their experiments - but usually the issues are addressed on a local scale. With home-built programs, implemented by non-professionals who have only little time to dedicate to that activity, only specific solutions are made adjusted to very individual needs. This creates obstacles for the transfer of knowledge, is hard to reverse engineer and usually forbids portability to similar experiments, even within the same research group.

Although, having good software is now recognized as a critical issue, researchers do not have access to IT support to help them develop a rigorous and lasting software solution. As a result, a lot of time and resources are wasted in temporary solutions.

This PhD project demanded for a different approach. As the work was carried out at two distant places (Ulm, Germany and Lyngby, Denmark) with about half-year alternating duration for a stay at each place, it accompanied some challenges on the control and measurement procedures. As a matter of fact, this initial work laid the foundation for a successful implementation and performance of the work presented in Chapter 5 and Chapter 6.

Verily, it is not true that every experiment is unique. The hardware configuration at every institute may vary significantly, but the strategy for a measurement is almost independent of the setup. It becomes apparent, that logic operations, relying on a physical idea for the measurement, have to be formulated hardware agnostic. Thus, supporting this way of thinking in a natural way by embedding it into a software framework, which led to the development of Qudi.

Qudi is a general software suite to control hardware and process data [113, 114]. It is written in Python, which has become a popular programming language for scientific work over the years [115, 116] and recently even ruled out the top programming language C and Java from its leadership position [117]. Qudi is open source and Python itself is a highly collaborative language. Hence, the core framework of Qudi relies on the free and open source character of the software to offer a maximum transparency and benefit for the scientific society. Qudi provides a large toolset for lab experiments for the quantum community and encourages community work. The software suite was originally intended for activities related to color centers in diamond. However, its underlying principle is not specific for the measurement and manipulation of color centers in diamond, and has a much broader and general application range.

The development of Qudi was supported by both institutes, the University of Ulm and the Technical University of Denmark. As being part of the core development team of
Qudi, we shaped, implemented and developed the conceptual ideas guiding this framework. The current status of the project as well as each individual contribution of all its members can be found in Ref. [113]. As a result, I had the unique opportunity to experience the benefits of our created ecosystem and to develop my PhD work on this basis. In the following, a detailed presentation of the concept of the measurement framework, termed as Qudi, is given.
Qudi: A modular python suite for experiment control and data processing

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ABSTRACT
Qudi is a general, modular, multi-operating system suite written in Python 3 for controlling laboratory experiments. It provides a structured environment by separating functionality into hardware abstraction, experiment logic and user interface layers. The core feature set comprises a graphical user interface, live data visualization, distributed execution over networks, rapid prototyping via Jupyter notebooks, configuration management, and data recording. Currently, the included modules are focused on confocal microscopy, quantum optics and quantum information experiments, but an expansion into other fields is possible and encouraged.

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If available Link to developer documentation/manual https://ulm-iqo.github.io/qudi-generated-docs/html-docs/
Support email for questions qudi@uni-ulm.de

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## 1. Motivation and significance

Modern scientific experiments typically rely on multiple hardware devices working together in a coordinated fashion. In many instances, the hardware devices are commercial products with programming interfaces for direct control via custom software. The unique combination of such devices is then specific to a given experiment. Efficient control of such experiments requires software that is capable of coordinating the operation of multiple devices. In addition, data interpretation is facilitated by rapid data processing and visualization.

These challenges are exemplified when studying color centers in diamond as solid state quantum emitters for sensing, spin manipulation and quantum information technologies. It is typical for such experiments to be performed on a “home-built confocal microscope” [1–5]. As evidenced by the 2014 Nobel Prize in Chemistry, these techniques have expanded beyond the context of physics and now this kind of microscope is pushing advances in biology [6–8] and nanotechnology [9, 10]. A wide range of hardware is used for such experiments, but there is a paucity of mature and flexible lab control software to operate the apparatus.

Here, we present Qudi, a Python software suite for controlling complex experiments and managing the acquisition and processing of measurement data. Despite being developed in the context of quantum optics laboratories, the core Qudi framework is broadly applicable to many scenarios involving coordinated operation of multiple experiment devices. The free and open-source nature of Qudi makes it possible for anyone to use and modify the software to fit their research needs, and the modular code design simplifies this task. Qudi continues to be actively developed, but it is already mature enough for reliable laboratory use [11].

## 2. Software description

### 2.1. Why Python?

Python was chosen as the programming language for Qudi because of its conceptual synergy with the goals of the project. As a dynamic, strongly typed, scripting language, Python has become a popular choice for scientific programming [12, 13] as the importance of scientific software increases [14]. Python’s high level of abstraction makes it human-readable and concise, providing a direct advantage for laboratory programming typically performed by scientists rather than dedicated software developers. Source code availability under an open-source license, the built-in modular structure of Python and good community support lower the initial hurdle to learn the language. Additionally, most laboratory hardware has at least an application programming interface (API) specified for the C programming language, which can be accessed by Python.

### 2.2. Qudi design

The Qudi suite consists of a collection of modules that are loaded and connected together by a manager component according to settings given in a configuration file as shown in Fig. 1(a). The program startup code and manager were initially derived from similar elements contained within the neurophysiology software ACQ4 [16]. Startup is initiated by a single executable python file, and the manager component provides core functions for logging, error handling, configuration reading, and remote access. Additionally, the manager also administers the other modules by providing functionality for module loading, module dependency resolution and connection, concurrent execution and network access to modules running on other computers. This core infrastructure makes it easier to rapidly develop modules for new experiments by providing structure and starting points.

A typical Qudi session will proceed as follows. On startup, the supervisor process, for example an IDE, creates a Qudi process. In this Qudi process, the manager component reads the configuration file, sets up the log file and loads the modules designated in the startup section of the configuration file. Typically, the startup section will – but does not have to – contain at least the Manager GUI and the tray icon module. Laboratory operation and experiment control are performed by science modules, which are specified in the configuration file along with any hardware-specific parameters. Science modules can be loaded for the desired measurement from the Manager GUI or a Jupyter notebook. Some of the science modules in Qudi were inspired by the p3diamond software [3–5, 17–19].

The science modules are divided into three categories: hardware interaction, experiment “logic”, and user interface. These categories and the relationships between them are illustrated in Fig. 1(b). The division into hardware, logic, and interface represents a clear separation of tasks that improves reliability and flexibility of the Qudi code. It also simplifies the implementation of new experiment modules. The fundamental three-fold distinction is at the basis of Qudi’s adaptability, and makes Qudi an experiment control software in contrast to a general software framework.

### 2.2.1. Logic modules

Logic modules control and synchronize a given experiment. They pass input parameters from the user interface to the respective hardware modules, and process measurement data in the desired way. These modules control the information exchange.
between different hardware modules and perform all necessary computations and conversions.

Logic modules are the only type of modules that are allowed to interact with each other. They are also the only type of module that has its own thread and event loop. Therefore they are the place where concurrent execution of tasks and synchronization of different devices is handled. All steps from the start of a measurement to its end, including data evaluation and storage are performed by the logic. This goes as far as producing “publication ready” plots of data that are saved together with the raw data and which provide a good overview or can be sent to collaborators without post-processing.

2.2.2. Hardware abstraction via interfaces

Today it is possible and even necessary to control most experiment hardware remotely. Unfortunately, the command structure, grammar, measurement units and connection methods differ widely between device models or devices from different suppliers of experiment hardware. To get the most re-usability out of logic modules, it must be possible to interchanger hardware modules for measurement devices that provide similar functionality, but work and communicate differently. It is the task of the hardware modules to overcome these problems by translating the commands given by the logic into the “language” of the specific hardware.

The problem is solved by defining an interface, a set of functions that a hardware module of a given type must implement, in order to make a certain measurement work. This set of functions is defined in a class (named ...Interface in a file in the interface folder) where the default implementation of each function raises an exception, if it is not replaced in the device-specific implementation. This class is then inherited by the actual implementing hardware module and all inherited functions must be overwritten.

Hardware modules can represent virtual dummy or mock hardware, which emulates the functionality of a device. Those dummies could load recorded measurement files, create arbitrary data or may perform real physical simulations of measurements, where the result is prepared according to the interface commands which the logic can access. One of the most significant uses of dummy hardware modules is to test the experiment logic without being connected to any actual hardware.

2.2.3. Advanced abstraction via “interfuses”

Building on the abstraction of interfaces, Qudi introduces an additional concept to facilitate the reuse of modules. This ability is provided by interfuse modules which interconnect (or fuse) different hardware or logic modules to modify their interface behavior or to achieve a task for which these modules were not originally designed.

An interfuse is a logic module that implements a hardware interface. In doing so, it pretends to be hardware that can connect to an experiment logic module. This allows the core experiment functions to remain in the logic module, while altering the kind of data that is measured. A tangible example helps clarify this concept. A confocal image (2D array) can represent single fluorescence values from a photon counter for each position (x, y). An interfuse makes it possible to replace the counter data with spectrometer measurements at each pixel, allowing fluorescence to be imaged with arbitrary spectral filtering. This practice improves maintainability and prevents code duplication.

The other reason to use interfuses is where a desired feature would require altering an existing interface definition. For example, an interfuse can perform the coordinate transform to correct for a tilted sample in a confocal scan. As a result, the tilted surface appears flat in the confocal image and can then be imaged at a consistent depth.

2.2.4. GUI

Qudi GUI modules create windows on the screen that a user can interact with, allowing experiment control and data visualization. Their purpose is to offer a convenient way for the user to interact with logic modules, however Qudi is fully functional without the GUI modules. The logic can also be controlled by the integrated Jupyter console or from a Jupyter notebook. For this reason, GUI modules are not allowed to interact with each other or the hardware directly and they do no data processing.

The Qudi graphical user interface (GUI) is built with Qt [20], offering users a familiar appearance. Qt is suitable due to its multi-platform GUI toolkit that provides good Python bindings [21,22] and makes it possible to separate the GUI design from the implemented functionality. Also, Qt’s multi-thread ability ensures good scalability and parallel processing, which are essential requirements for complex experiments. Furthermore, Qt implements a signal-slot mechanism [23] that is very useful for...
Table 1
Overview of science modules included in the Qudi suite.

<table>
<thead>
<tr>
<th>Name</th>
<th>Purpose</th>
</tr>
</thead>
<tbody>
<tr>
<td>Confocal (GUI + logic)</td>
<td>Confocal microscope interface for imaging and positioning scanner.</td>
</tr>
<tr>
<td>Optimizer (logic)</td>
<td>Automatically center image scanner on a local signal maximum.</td>
</tr>
<tr>
<td>odmr (GUI + logic)</td>
<td>Microwave resonance experiments.</td>
</tr>
<tr>
<td>Pulsed (GUI + logic)</td>
<td>Pulse sequence measurements (pulsed laser and/or microwave).</td>
</tr>
<tr>
<td>Poincare (GUI + logic)</td>
<td>Point-of-interest manager for keeping track of multiple measurement spots.</td>
</tr>
<tr>
<td>Magnet (GUI + logic)</td>
<td>Driving physical magnet on motorized stages to vary applied magnetic field.</td>
</tr>
<tr>
<td>Fit (logic)</td>
<td>Obtain fits for data in various common models (Gaussian, Lorenzian, sinusoidal, etc.).</td>
</tr>
<tr>
<td>Counter (GUI + logic)</td>
<td>Perform and display counting tasks of binary events either in continuous or gated way.</td>
</tr>
<tr>
<td>Wavemeter_logger (GUI + logic)</td>
<td>Record and process data as a function of laser wavelength as measured by a wavemeter.</td>
</tr>
</tbody>
</table>

concurrency, modular design, and interaction between GUI modules and logic modules. On top of this, the Python library PyQtGraph [24] makes it easy to create interactive, frequently updated 2- and 3-dimensional plots. The user interface can be edited graphically in Qt Designer and is stored as an XML file. For rapid prototyping, this file can be (re)loaded by a running Python program. The GUI design strives to adhere to the KDE Human Interface Guidelines [25], as these stress the importance of interface familiarity and they work well with the default set of Qt user interface elements.

3. Impact and reuse potential

The Qudi suite is useful for any small to medium-size computer-controlled laboratory experiment. Its modular design combined with the use of interface definitions makes it easy to integrate new hardware into an existing experiment. Moreover, this design offers the capability to easily reuse existing modules in new experiments. The Qudi core infrastructure is broadly applicable, even beyond the context of confocal microscopy or physics experiments in general.

Qudi is of more tangible impact to the quantum optics community in particular. The existing modules already offer control over confocal microscopes, electromagnets, motorized stages, lasers, (arbitrary) signal generators, and other devices used in this field of research. Table 1 lists the science modules currently included in the Qudi suite. Furthermore, typical measurement protocols and data analysis functions are already implemented. These existing modules make Qudi a ready-to-use Python-based software suite for quantum optics labs, independent from the individual hardware and measurement schemes used by different groups.

4. Illustrative example

The measurement of optically detected magnetic resonance (ODMR) on single color centers in diamond [26,27] requires the coordinated operation of a scanning confocal microscope and a microwave source. This section describes how such a measurement is performed with Qudi, illustrating the convenience arising from the software design outlined in this paper. The interested reader can perform this process using the default config distributed with Qudi that loads dummy hardware modules to provide representative data. This experiment makes use of the “confocal” and “ODMR” science modules.

The first step is to find a single color center inside the diamond. The Qudi confocal GUI and logic modules are used to move a diffraction-limited focal spot through the diamond sample in three dimensions [28–31]. This is achieved by scanning hardware that is controlled by the confocal logic. A photon counter records the fluorescence measured by the confocal microscope, and this hardware device sends the data to the confocal logic. The confocal logic produces an image of fluorescence as a function of position, and the GUI presents this image to the user. Fig. 2 shows the confocal GUI with an x-y image on the left and an x-z image on the right.

In order to focus on a single center, the user places the confocal cursor near a promising spot. An optimizer module performs a series of close-range scans around the cursor, and the optimal position of maximum fluorescence is found via a fitting module built on the Imfit package [32]. A 2D gaussian fit is performed on the x-y plane scan and for the third dimension a 1D gaussian fit on the z line scan. These are shown in Fig. 2 on the lower right of the Confocal GUI. Finally, the optimizer module moves the scanning hardware to the optimal position focussed on the desired single color center.

In addition to spatial alignment, a microwave resonance condition has to be matched in order to detect the desired change in optical signal [26,27,33–35]. The ODMR logic module controls the frequency of a microwave source while recording the fluorescence level. The design of Qudi means that the ODMR logic is easily capable of driving a variety of microwave source hardware, increasing flexibility in the laboratory.

An ODMR experiment is performed by sweeping the microwave frequency and recording the fluorescence. Recorded data are shown live on screen in the ODMR GUI as both the fluorescence sum of all frequency sweeps and as a matrix plot containing each sweep (Fig. 2, lower right). ODMR scans of several spots can be measured automatically by saving the color center positions and then using a script to move from spot to spot, optimizing the position on each site and recording an ODMR spectrum.

5. Conclusions and future directions

Qudi is a generally applicable experiment control software suite, with infrastructure to support modular design of experiments, significantly reducing the effort involved in constructing new experiments. Qudi already offers a developed quantum-optics tool set capable of reliable laboratory operation, and a modern user interface.

There is continuing effort to expand the library of available science modules. One priority for the future is to simplify the setup of Qudi by providing a graphical configuration editor. Furthermore, it would be convenient to make Qudi installable from the Python
Package Index. In the context of experiment operation, enhanced automation capabilities are desired to allow a user to rearrange the existing functionality without programming.

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We would like to thank Boris Naydenov for advocating the use of a software platform that can be used by the whole Quantum Optics institute and for maintaining the predecessor to this software. Furthermore, we would like to thank Ou Wang, Gerhard Wolff, Samuel Müller and Andrea Filipovskifor contributions to the software, testing and reporting bugs.

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References


Fig. 2. Simplified illustration of Qudi used to perform ODMR experiments in the laboratory. The experimental setup consists of three main parts. The confocal microscope is used to image the red fluorescence of color centers in diamond using a green excitation laser. The objective can be scanned in all three dimensions by scanner hardware. An avalanche photodiode (APD) detects red fluorescence photons which are counted by digital data acquisition hardware. In addition, a signal generator exposes the color centers to a microwave field which lowers the fluorescence at certain resonance frequencies (ODMR). The Confocal GUI shows the fluorescence images used to position the optical focal spot and the ODMR GUI displays the microwave resonance spectra. This figure illustrates the experience of a user, and does not show the logic modules which perform experimental functions. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)


Narrow-bandwidth sensing of high-frequency fields with continuous dynamical decoupling

Whatever you can do, or dream you can, begin it.

Boldness has genius power and magic in it.

JOHANN WOLFGANG VON GOETHE
5 Narrow-bandwidth sensing of high-frequency fields with continuous dynamical decoupling

5.1 Introduction

High frequency spectroscopy with the NV center is challenging. Weak oscillating signals are typically out of reach for ordinary detection schemes and dynamical decoupling becomes the method of choice. Their range of application covers pulsed and continuous wave protocols, where both were restricted up to MHz frequencies. High frequency spectroscopy with high spectral resolution of magnetic fields in the GHz regime remained a challenging avenue not only for the NV center community. Only relaxometry measurements offered the ability to detect high-frequency fields, but with a $T_2^*$ limited bandwidth.

In the following, a general scheme is shown which relaxes this limitation and allows for the detection of high-frequency signals in the GHz range with a bandwidth of $T_2^*$. In addition, continuous dynamical decoupling is integrated into a sensing task. The basic idea relies on continuous concatenated drive which turns a given two-level system, with energy separation in the regime of the signal, into a narrow-bandwidth high-frequency sensor.

In continuous dynamical decoupling protocols, the sensor is permanently subjected to drive fields which constitute the largest noise source in the scheme. Concatenation of drive fields reduces significantly the impact of drive noise by creating a protected subspace. The mere drive of the system realizes at the same time a decoupling of the NV sensor from its environmental noise by creating dressed states.

The characteristic of this approach reveals that the interaction with a high-frequency field occurs at restrictive resonance conditions. That means the frequency of the measured field is detected by a narrow-bandwidth condition imposed by the drive fields and the bare energy separation of the NV sensor. The interaction strength of the high-frequency field is imprinted as oscillations in the population between the protected states.

The coherence time of the sensor, $T_2^{\Omega_i}$, under a drive with strength $\Omega_i$, gives the timescale in which an oscillation between the protected states can be detected. The smallest detectable interaction strength relies therefore on the ability to record at least one oscillation within $T_2^{\Omega_i}$ of the protected states. Hence, the measurement of the signal strength of the high-frequency field is very similar to the nature of Rabi oscillations.

A very unique feature of this protocol is the possibility that the high-frequency signal itself can act as a weak driving field (of the protected states), thereby creating an even better protected subspace. In this case, the signal can be regarded as an additional weak drive, thereby prolonging non-linearly the coherence time of the sensor.

A rigorous and extensive introduction in the theoretical formalism as well as some background information is given in the supplementary information, in Sec. 5.3 The main publication in Sec. 5.2 summarizes the findings.
Narrow-bandwidth sensing of high-frequency fields with continuous dynamical decoupling

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State-of-the-art methods for sensing weak AC fields are only efficient in the low frequency domain (<10 MHz). The inefficiency of sensing high-frequency signals is due to the lack of ability to use dynamical decoupling. In this paper we show that dynamical decoupling can be incorporated into high-frequency sensing schemes and by this we demonstrate that the high sensitivity achieved for low frequency can be extended to the whole spectrum. While our scheme is general and suitable to a variety of atomic and solid-state systems, we experimentally demonstrate it with the nitrogen-vacancy center in diamond. For a diamond with natural abundance of 13C, we achieve coherence times up to 1.43 ms resulting in a smallest detectable magnetic field strength of 4 nT at 1.6 GHz. Attributed to the inherent nature of our scheme, we observe an additional increase in coherence time due to the signal itself.
Improving the sensitivity of high-frequency sensing schemes is of great significance, especially for classical fields sensing1–3, detection of electron spins in solids4–5, and nuclear magnetic resonance spectroscopy6. The common method to detect high-frequency field components is based on relaxation measurements, where the signal induces an observable effect on the lifetime, \( T_\text{1} \), of the probe system. \( T_\text{2} \). Nevertheless, the sensitivity of this method is limited by the pure dephasing time \( T_\text{2} \) of the probe system.

Pulsed dynamical decoupling8–10 can substantially increase the coherence time11–18. In order to carry out sensing with a decoupling scheme, the frequency of the decoupling pulses has to be matched with the frequency of the target field19, 20. This largely restricts the approach to low frequencies, as the repetitive application of pulses is limited by the maximum available power per pulse21. The same power restrictions are present for very rapid and composite pulse sequences aimed to decrease both external and controller noise22–28. With continuous dynamical decoupling (CDD)21, 27–29, robustness to external and controller noise can be attained, especially for multi-level systems30–33. However, the significance of CDD for sensing high-frequency fields remains elusive. Indeed, it was unclear whether it is possible to incorporate such a protection into the metrology task of sensing frequencies in the GHz domain. The first step towards this goal was done recently by integrating CDD in the sensing of high-frequency fields with three level systems32.

In this article, we propose, analyze, and experimentally demonstrate a sensing scheme that is capable of probing high-frequency signals with a coherence time, \( T_\text{2} \), limited sensitivity. Unlike relaxation measurements comprising a bandwidth \( \Omega_1/T_\text{2} \), determined by the pure dephasing time, \( T_\text{2} \), of the sensor (up to the MHz range), our protocol overcomes the imposed limitation by protecting the addressed two-level system (TLS) with an adapted concatenated CDD approach. We use and adjust it such that high-frequency sensing becomes feasible even for not phase-matched signals. As a result, the proposed scheme is generic and works for many atomic or solid-state TLS, in which the energy gap matches the frequency of the signal under interrogation. A remarkable feature of our scheme is the fact that the signal to be probed also works partially as a decoupling drive and thus improves further the sensitivity of the sensor.

We demonstrate the performance of CDD by applying it to a nitrogen-vacancy (NV) center in diamond with natural abundance of 1\%34, 35. Here, we utilize two of its ground sub-levels as the TLS. The states of the NV center can be read out and initialized by a 532 nm laser, which reveals spin-dependent fluorescence between the two levels36–38. Moreover, we report on the measurement of a weak high-frequency signal with strength \( g \), which relates to a smallest detectable magnetic field amplitude of \( \delta B_{\text{min}} \approx 4 \) nT.

### Results

#### The sensing scheme

The basic idea of utilizing concatenated continuous driving to create a robust qubit is illustrated in Fig. 1a, b. The concatenation of two phase-matched driving fields results in a robust qubit36, 42. In what follows we show that such a robust qubit can be utilized as a sensor for frequencies in the range of the qubit’s energy separation and hence, dynamical decoupling can be integrated into the sensing task. By the concatenated driving, the qubit is prepared in a state that allows for strong coherent coupling to the high-frequency signal to be probed (corresponding to the last TLS in Fig. 1c). In the total Hamiltonian, \( H \), we consider the concatenation of two driving fields of strength (the Rabi frequency) \( \Omega_1 \) and \( \Omega_2 \), respectively. The Hamiltonians of the TLS, \( H_\text{TLS} \), the protecting driving fields, \( H_\text{D1}, H_\text{D2} \), and the signal, \( H_\text{g} \), are given by

\[
H = H_\text{TLS} + H_\text{D1} + H_\text{D2} + H_\text{g},
\]

where \( \Omega_0 \) is the energy gap of the bare states (\( \hbar = 1 \)), \( \omega_0 \) is the frequency of the signal, and \( g \) is the signal strength which we want to determine. We tune the system, i.e., \( \omega_0, \Omega_1, \) and \( \Omega_2 \), such that \( \omega_0 = \Omega_1 + \Omega_2/2 \).

It is an important feature that phase matching between the signal and the control is not required, which means that the signal phase \( \varphi \) can be unknown and moreover, it may vary between experimental runs. In addition, we make the assumption that \( \omega_0 > \Omega_1 > \Omega_2 > g \). Moving to the interaction picture (IP) with respect to \( H_0 = \hbar/2 \sigma_\text{z} \), and making the rotating-wave approximation, we obtain

\[
H_I = \frac{\Omega_1}{2} \sigma_x + \frac{\Omega_2}{2} \sigma_x \cos(\Omega_0 t) + \Omega_2 \sigma_x \cos(\omega_0 t) \cos(\Omega_0 t) + g \sigma_x \cos(\omega_0 t + \varphi) ,
\]

This picture incorporates the effect of \( \Omega_0 \) onto a TLS and express the new system in eigenstates of \( \sigma_x \), the \( \{ \pm \} \) (dressed) states, which separates the contributions from \( \Omega_2 \) and \( g \). For a large enough drive \( \Omega_1 \), the \( \{ \pm \} \) eigenstates are decoupled (in first order) from magnetic noise, \( \delta B_{\text{tot}} \), because \( \langle \pm | \sigma_x | \pm \rangle = 0 \).

However, power fluctuations \( \delta \Omega_1 \) of \( \Omega_1 \) limit the coherence time of the dressed states. The resulting IP is illustrated in the second TLS in Fig. 1c.

We continue by moving to a second IP with respect to \( H_{01} = \hbar/2 \sigma_x \), which leads to

\[
H_{II} = \frac{\Omega_1}{4} \sigma_y + \frac{g}{4} \left( -i \sigma_z e^{-i(\Omega_1/2 + \varphi)t} + i \sigma_z e^{i(\Omega_1/2 + \varphi)t} \right) - \frac{\Omega_2}{2} \sigma_x \cos(\omega_0 t + \varphi).
\]

Once again, we incorporate \( \Omega_1 \) into the dressed states, so that solely the contribution of the signal \( g \) becomes obvious, which is depicted in the last TLS of Fig. 1c. The second drive, \( \Omega_2 \), which is larger than \( \delta \Omega_1 \), creates effectively doubly dressed states (the \( \sigma_y \) eigenstates). These doubly dressed states are immune to power fluctuations of \( \Omega_1 \) and hence prolong the coherence time (see Supplementary Note 2 for more details). Moving to the third IP with respect to \( H_{02} = \hbar \sigma_y \), results in

\[
H_{III} = \frac{g}{8} \left( \sigma_y e^{-i \omega_0 t} + \sigma_y e^{i \omega_0 t} \right) ,
\]

where we can clearly see that the signal \( g \) induces rotations in the robust qubit subspace (either with \( \sigma_y \) or \( \sigma_z \)). These rotations are obtained for any value of an arbitrary phase \( \varphi \) and the bandwidth (\( \Omega_1/2 \Omega_2 \)) is now limited by the coherence time, \( T_\text{2} \), of the sensor. Hence, if a given TLS exhibits the possibility of manipulating it via drive fields \( H_\text{D1} \) and \( H_\text{D2} \), we can achieve a high-frequency sensor in the range of \( \delta B_{\text{min}} \).
By this, we overcome the low frequency limit that is common to state-of-the-art pulsed dynamical decoupling sensing methods. In addition, we present an analog pulsed version of our scheme, where the pulsing rate is much lower than the frequency of the signal (Supplementary Note 9). However it is not a direct measurement of the signal, but based on a signal demodulation approach. Compared to the pulsed schemes, CDD does not suffer from being susceptible to higher harmonics of the decoupling window appearing naturally from the periodic character of the pulsed sequence. Eventually, less power per unit time is used in the continuous scheme leading to a smaller overall noise contribution from the drive.

Implementation and analysis of the presented scheme. After determining the optimal drive parameters, \( \Omega_1 \) and \( \Omega_2 \), for the concatenated sensing sequence, and thereby maximize the contribution from the drive.

The measurements take place in the laboratory frame, i.e., all three contributions \( \Omega_1 \), \( \Omega_2 \), and \( g \) to the population dynamics of the TLS will be visible. In order to see solely the effect of \( g \) on the TLS, we alter the modulation of the second drive in Eq. (2) to \( \cos (\Omega t + \pi/2) \). This does not change the performance of the scheme, but only changes the axis of rotation to \( \sigma_z \) for the second drive \( \Omega_2 \). Since the readout laser is effectively projecting the population in the \( \sigma_z \) eigenbasis, we can make the Rabi rotations of \( \Omega_2 \) invisible to the readout. To remove the effect of \( \Omega_1 \) in the data, we can simply sample the measurement at multiple times of \( \tau_{\Omega_1} = 2\pi/\Omega_1 \), i.e., we measure at times \( t = N\tau_{\Omega_1} \) \( (N \in \mathbb{N}) \). This procedure reveals directly \( g' \) as the signal induces Rabi oscillations of the robust qubit under double (single) drive (Fig. 2). Alternatively, we could have applied at the end of the drive a correction pulse in order to complete the full \( \Omega_1 \) and \( \Omega_2 \) rotations, so that just the effect of \( g' \) remains.

Without a signal, \( g \), we achieve coherence times of \( T_2^\sigma \approx 60 \mu s \) with a single drive (\( \Omega_2 = 0 \), compare Supplementary Fig. 3 in Supplementary Note 6A) and \( T_2^\sigma = 393 \mu s \) with a double drive (compare Supplementary Fig. 4 in Supplementary Note 6B). The results for long and slow Rabi oscillations induced by an external signal, \( g \), under single and double drive (\( \Omega_2/\Omega_1 \approx 0.15 \)) are shown in Fig. 2. These illustrate a significant increase of the coherence time of the sensor by two orders of magnitude, from \( T_2^\sigma = 60 \mu s \) to a lifetime limited coherence time of \( (T_1/2\pi) = 1.43 \mathrm{ms} \). It should be noted that the signal itself can be considered as an additional drive (cf. Eq. (3)), correcting external errors \( \delta \Omega \) of the previous drive and thereby prolonging the coherence time even further. Consequently, we can improve the bandwidth for high-frequency sensing by almost three orders of magnitude from \( \approx 900 \mathrm{kHz} \) (for \( T_2^\sigma \approx 1.1 \mu s \)) to \( \approx 700 \mathrm{Hz} \) (for a \( T_2^\sigma = 1.43 \mu s \)). Moreover, in Supplementary Note 3 we discuss an improved version of our scheme which has the potential to push the coherence time of the sensor further towards the lifetime limit.

To benchmark the double drive scheme against a standard single drive approach, we determine the smallest magnetic field which can be sensed after an accumulation time \( t \). The smallest measurable signal \( S \) is eventually bounded by the smallest measurable magnetic field change \( \delta B_{\min} \), which is found to be

\[
\delta B_{\min}(t, \tau) = \frac{\delta S}{\max|\delta B|} = \frac{1}{\tau_{\sigma} T_{1\mathrm{NV}}/\alpha C}.
\]

Here, \( \gamma_{\sigma \mathrm{NV}}/2\pi = 28.8 \mathrm{GHz T}^{-1} \) is the gyromagnetic ratio of the NV defect, \( \sigma(t) \) is the standard deviation of the measured normalized fluorescence counts after time \( t \), \( \alpha \) accounts for a different phase accumulation rate depending on the decoupling scheme, and \( C \) is the contrast of the signal (see Supplementary Note 5 for detailed derivation). Since the photon counting is shot noise limited, we have \( \sigma(t) = 1/\sqrt{N_{\mathrm{ph}} \cdot N} \), with \( N_{\mathrm{ph}} \) being the number of photons measured in \( t \) and \( N = t/\tau \) is the number of sequence repetitions. With this, Eq. (5) will transform in the commonly known form, with some measurement dependent constants.

We recorded \( \sigma(t) \) as a function of time and use this to determine \( \delta B_{\min} \). The results of this measurement for both the single and double drive are summarized in Fig. 3. The sensitivity

![Fig. 1 Schematic representation of our setup. a]a The NV center probes an external signal while it is being manipulated by the control fields. b Schematic representation of the sequence applied in this work. c A The protected TLS; the bare system, \( H_s \), is subjected to strong environmental noise \( \delta \). Applying a strong drive, \( \Omega \), opens a protected gap, now subjected mainly to drive fluctuations \( \delta \Omega \). A second drive, \( \Omega_2 \), is then applied to protect the TLS, \( H_s \), from these fluctuations, resulting in a TLS, \( H_s \), on resonance with the signal, \( g' \approx g/4 \), with noise mainly from the second weak drive \( \delta \Omega_2 \approx \delta \Omega \).
can be obtained by $\eta(\tau) = \delta B_{\text{min}}(t, \tau) \sqrt{T}$, which is optimal in the vicinity of the coherence time of the sensor, $\tau \approx T_2$. With our system, we achieve a sensitivity of $\eta_{\Omega, \omega, \theta} \approx 5 \mu T$ Hz$^{-0.5}$ in the double drive case at $-1.6$ GHz, which should be compared to $\eta_{\Omega, \omega, \theta} \approx 20 \mu T$ Hz$^{-0.5}$ for a single drive approach.

Both traces in Fig. 3 were recorded while a signal $g$ was applied. Apart from the mere fact, that the number of driving fields are different, the specific choice for $\tau$ will also determine the magnitude of the smallest measurable magnetic field change $\delta B_{\text{min}}$. Obtaining the coherence time without a signal, $g$ (which are $T_{2,\text{r}}$ and $T_{2,\text{gd}}$), is a common practice in the field, but will not result in a correct choice of $\tau$ for the sensitivity measurement and also for Eq. (5), since the signal has an impact on the sensor’s sensitivity. However, if $\delta B_{\text{min}}$ shall be evaluated correctly, then the non-linearity, i.e., the coherence time prolonging effect of the signal, has to be taken into account. Otherwise an even worse non-linearity, i.e., the coherence time prolonging effect of the single drive case in Fig. 3, where $\tau \equiv \frac{1}{\Omega} \approx 1.43$ ms, was recorded.

Figure 3 illustrates the smallest measurable magnetic field change $\delta B_{\text{min}} = (2 \pi \delta \theta_{g,\text{min}})/\gamma \tau$ as a function of total measurement time. To show the total improvement, we obtain $\sigma(t) \approx T_{2,\text{r}} \approx 60 \mu s$ in the single drive case and $\sigma(t) \approx T_{2,\text{gd}} \approx 1.43$ ms in the double drive case. Note, that for both data traces a signal was always present, $g/2\pi = 26.9$ kHz and $g/2\pi = 62.9$ kHz in the single and in the double drive, respectively. But only in the double drive the coherence time prolonging effect of $g$ was included into the choice of $\tau$ for Eq. (5) (i.e., the measurement was performed at $\tau = T_{2,\text{gd}}$ instead of $\tau = T_{2,\text{r}} \approx 393 \mu s$).

This effect on the coherence time of the sensor (i.e., we measure at $\tau = T_{2,\text{gd}}$ and not at $\tau = T_{2,\text{r}}$). This effect was included in the double drive case.

To examine the signal protection effect more in detail, the coherence time of the sensor is measured as a function of signal strength, $g$, in a single drive configuration (Supplementary Note 7). From these measurements we project the sensitivity associated with a specific signal strength (Fig. 4), assuming $\sigma(t)$ is unchanged for the same repetition $N$. This is a reasonable assumption given that the only difference between measurements is the signal strength, $g$, and sequence length, $\tau$.

Eventually, this phenomenon, which seems to be an inherent part of this continuous scheme, can be used to further increase the performance of the sensor by fine tuning the controlled parameters (static bias field $B_{\text{bias}}$ and thereby changing $\omega_0$, $\Omega_1$ and $\Omega_2$) to match the signal frequency, $\omega_s$, and strength, $g$ (see Supplementary Note 3 for further discussions).

**Discussion**

We have demonstrated that dynamical decoupling can be used in the context of sensing high frequency fields. In contrast to state-of-the-art pulsed dynamical decoupling protocols, we can show that CDD can be simultaneously integrated into the sensing task. By utilizing a NV center in diamond we have demonstrated by pure concatenation of two drives a coherence time of $\sim 393 \mu s$ which constitutes an improvement of more than two orders of magnitude over $T_2$, and an increase of resolution from the MHz to a few kHz. The application of this method for wireless communication could have a transformative effect due to the high resolution of the protocol. Since the protocol is applicable to a variety of solid-state, molecular, and atomic systems, we believe that it has a great potential to have a significant impact on many fields and tasks that involve high frequency sensing (up to frequencies in the THz range). Eventually, this method could also be used to improve the coupling to quantum systems. We would like to note that during the preparation of this manuscript we became aware of a related independent work by Joas et al.20.
**Data availability.** The authors declare that all relevant data supporting the findings of this study are available within the paper (and its Supplementary Information file). Any raw data can be obtained from the corresponding authors on reasonable request.

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**References**


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**Author contributions**

N.A. and A.R. conceived the idea and developed the theory. A.H., A.R., U.L.A. and F.J. designed and supervised the project. A.S., T.U. and D.L. performed, planned and developed the concept of the experiment. A.S., N.A., T.U. and D.L. analyzed the data. N.A. and D.L. planned and carried out the simulations. A.S. and N.A. took the lead in writing the manuscript. All authors contributed to the interpretation of the results, provided critical feedback and helped to shape the research, analysis and manuscript.

**Additional information**

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5.3 Supplementary Information

Supplementary Note 1: Sensing of a high frequency signal with a single drive

In this section we describe in detail how the enhanced sensing of high frequency signals can be performed with a two-level system (TLS) by employing a single continuous driving field. The Hamiltonian of the TLS is given by

\[ H = \frac{\omega_0}{2} \sigma_z + \Omega_1 \left( 1 + \delta\Omega_1(t) \right) \cos(\omega_0 t) \sigma_x + g \cos(\omega_0 t + \varphi) \sigma_z + \delta_B(t) \sigma_z, \]

where \( \omega_0 \) corresponds to the energy gap of the bare states, \( \Omega_1 \) is the Rabi frequency of the on-resonance driving field, \( g \) is the Rabi frequency of the signal, \( \varphi \) is a random phase, which indicates that the signal field and the driving field are not phased-matched, and therefore \( \varphi \) has a different value in each experiment. We tune the system, i.e., \( \omega_0 \) and \( \Omega_1 \), such that \( \omega_0 = \omega_0 + \Omega_1 \). In addition, \( \delta\Omega_1(t) \Omega_2 \) represents the fluctuations of the driving field, and \( \delta_B(t) \) is the magnetic noise. Moving to the interaction picture (IP) with respect to \( g \), making the rotating-wave-approximation (RWA), we get

\[ H_I = \frac{\Omega_1}{2} \left( 1 + \delta\Omega_1(t) \right) \sigma_z + g \left( \sigma_x e^{-i(\Omega_1 t + \varphi)} + \sigma_x e^{i(\Omega_1 t + \varphi)} \right) + \delta_B(t) \sigma_z. \]

In the basis of the dressed states, the \( \sigma \) eigenstates, the Hamiltonian reads \((x \rightarrow z, z \rightarrow -x, y \rightarrow y)\)

\[ H_I = \frac{\Omega_1}{2} \left( 1 + \delta\Omega_1(t) \right) \sigma_z + g \left( \cos(\Omega_1 t + \varphi) \sigma_x + \sin(\Omega_1 t + \varphi) \sigma_y \right) - \delta_B(t) \sigma_z. \]

Because the magnetic noise couples between the dressed states, in first order, the noise induces a longitudinal relaxation (decay) rate of \( S_B(\Omega_1) \), where \( S_B \) is the power spectrum of the noise. A large enough \( \Omega_1 \) ensures that the longitudinal relaxation rate is negligible \( (S_B(\Omega_1) \ll 1/T_1) \). In this case, within the experiment time the noise does not induce transitions between the dressed states, but does result in a second order fluctuating phase shift of \( \sim \delta_B(t)/\Omega_1 \). The resulting dephasing rate is considerably diminished with an increasing \( \Omega_1 \) [1]. The main limitation on the coherence time of the dressed states, \( T_2^D \), is due to power fluctuations of the driving field, \( \delta\Omega_1(t) \Omega_1 \), which limit the coherence time to \( T_2^D \sim 1/(\delta_B(t) \Omega_1) \). For typical experimental setups \( \delta\Omega_1(t) \sim 0.1 - 1\% \) implies an improvement of \( \sim 1 \) order of magnitude in the coherence time compared to \( T_2 \).

Note, that throughout this manuscript and in the main text, we use the following notation: \( T_1 \), known as the longitudinal relaxation time of the qubit, is called the lifetime of the sensor. \( T_2 \) describes the pure dephasing time of the sensor, if no protection or drive is applied to the sensor. \( T_2 \) denotes the transverse relaxation time, which is the coherence time in a pulsed dynamical decoupling experiments. \( T_2^D \) expresses the coherence time under drive \( \Omega_1 \) and \( T_2^{D,\varphi} \) characterize a coherence time under drive \( \Omega_1 \) with an externally applied signal \( g \).

We continue by moving to the interaction picture (IP) with respect to \( H_{II} = \frac{\Omega_1}{2} \sigma_z \) (in the basis of the dressed states) and, assuming that \( \Omega_1 \gg g \), making the rotating-wave-approximation (RWA), which leads to

\[ H_{II} = \frac{g}{4} \left( e^{-i\varphi} \sigma_x + e^{i\varphi} \sigma_x \right) + \frac{\delta\Omega_1(t) \Omega_1}{2} \sigma_z, \]

where we neglected the (fast rotating) terms of the magnetic noise. The signal in \( H_{II} \) corresponds to the on-resonance coupling between the dressed states, and hence the signal induces rotations of the dressed qubit, regardless to the value of \( \varphi \) (so long as \( \varphi \) is constant during a single experiment).

Supplementary Note 2: Sensing of a high frequency signal with a double drive

In order to mitigate the driving fluctuations of the (first) driving field we introduce a second drive, polarized along a perpendicular direction with respect to the polarization of the first driving field. We therefore consider the Hamiltonian

\[ H = \frac{\omega_0}{2} \sigma_z + \Omega_1 \left( 1 + \delta\Omega_1(t) \right) \cos(\omega_0 t) \sigma_z + \Omega_2 \left( 1 + \delta\Omega_2(t) \right) \cos(\omega_0 t) \cos(\Omega_1 t) \sigma_y + g \cos(\omega_0 t + \varphi) \sigma_z + \delta_B(t) \sigma_z. \]
where $\Omega_2$ is the Rabi frequency of the second drive, and we tune the system such that $\omega_s = \omega_0 + \Omega_1 + \Omega_2$. In the first IP with respect to $H_0 = \frac{g}{2} \sigma_z$, and after making the RWA and moving to the basis of the dressed states ($x \rightarrow z$, $z \rightarrow -x$, $y \rightarrow y$) we have that

$$H_I = \Omega_1 \left( 1 + \delta_{\Omega_1}(t) \right) \sigma_z + \Omega_2 \left( 1 + \delta_{\Omega_2}(t) \right) \cos (\Omega_1 t) \sigma_y$$

$$+ \frac{g}{2} \left[ \cos \left( \left( \Omega_1 + \frac{\Omega_2}{2} \right) t + \phi \right) \sigma_x + \sin \left( \left( \Omega_1 + \frac{\Omega_2}{2} \right) t + \phi \right) \sigma_y \right] \sigma_z - \delta_B(t) \sigma_z. \tag{6}$$

We continue by moving to the second IP with respect to $H_0 = \frac{\Omega_2}{2} \sigma_z$ (in the basis of the dressed states) and taking the RWA to obtain

$$H_{II} = \frac{\delta_{\Omega_1}(t) \Omega_1}{2} \sigma_z + \frac{\Omega_2}{4} \left( 1 + \delta_{\Omega_2}(t) \right) \sigma_y + \frac{g}{4} \left[ \sigma_z e^{-i\left( \frac{\Omega_2}{2} t + \phi \right)} + \sigma_x e^{i\left( \frac{\Omega_2}{2} t + \phi \right)} \right], \tag{7}$$

where we neglected the (fast rotating) terms of the magnetic noise. In the basis of the doubly-dressed states, the $\sigma_y$ eigenstates ($y \rightarrow z$, $z \rightarrow -y$, $x \rightarrow x$), the Hamiltonian is given by

$$H_{II} = \frac{\delta_{\Omega_1}(t) \Omega_1}{4} \sigma_z - \frac{\delta_{\Omega_2}(t) \Omega_2}{4} \sigma_x + \frac{g}{4} \left[ \sigma_z e^{-i\phi} + \sigma_x e^{i\phi} \right] - \frac{\delta_{\Omega_2}(t) \Omega_2}{4} \sigma_z. \tag{8}$$

Finally, in the third IP with respect to $H_{02} = \frac{\Omega_2}{4} \sigma_z$ (in the basis of the doubly-dressed states), and after making the RWA we have that

$$H_{III} = \frac{g}{8} \left( \sigma_z e^{-i\phi} + \sigma_x e^{i\phi} \right) + \frac{\delta_{\Omega_2}(t) \Omega_2}{4} \sigma_z. \tag{9}$$

where we neglected the (fast rotating) terms of the driving fluctuations of $\Omega_2$, which now contribute only as a second order effect. We therefore conclude that the signal in $H_{III}$ corresponds to the on-resonance coupling between the doubly-dressed states, and hence the signal induces rotations of the doubly-dressed qubit. The doubly-dressed states are vulnerable to fluctuations of $\Omega_2$, but since $\Omega_2 \ll \Omega_1$ these fluctuations have a smaller effect than fluctuations in $\Omega_1$; with $\delta_{\Omega_2}(t) \sim 0.1 - 1\%$ the coherence time is improved by $\sim 1$ order of magnitude with respect to the coherence time of the dressed states (an improvement of $\sim 2$ orders of magnitude with respect to $T_2^\ast$). In principle, the robustness to driving fluctuations can be further improved by the concatenation of more driving fields [2].

**Supplementary Note 3: Improved scheme**

In our scheme, a weak signal may further prolong the coherence time of the probe qubit. To see this we continue from equation (9) and assume for simplicity that $\phi = 0$. In this case

$$H_{III} = \frac{g}{8} \sigma_z + \frac{\delta_{\Omega_2}(t) \Omega_2}{4} \sigma_z. \tag{10}$$

However, the difference between the signal, $g \cos (\omega_1 t + \phi) \sigma_z$, and a concatenated third drive, which in our case could be given by $\Omega_3 \cos (\omega_1 t) \cos \left( (\Omega_2/2) t \right) \sigma_z$, comes from the counter-rotating terms that we usually neglect when making the RWA. Although that the condition $\Omega_1 \gg \Omega_2$ holds, examination of the counter-rotating terms of the signal $g$, the magnetic noise $\delta_B(t)$, and the driving noise of the first drive $\delta_{\Omega_1}(t)$, in the third IP, reveals that there are counter-rotating terms of $g$ and $\delta_B(t)$ and of $g$ and $\delta_{\Omega_1}(t)$ that have identical frequencies. This implies that these terms result in an effective time-independent Hamiltonian [3] that should be included in equation (10), and hence,

$$H_{III} = \frac{g}{8} \sigma_z + \frac{\delta_{\Omega_2}(t) \Omega_2}{4} \sigma_z + \frac{g \delta_B(t)}{8 \Omega_1} \sigma_x + \frac{g \delta_{\Omega_1}(t)}{4 \Omega_2} \sigma_z. \tag{11}$$

The added effective terms imply dephasing rates of $\Gamma_{\delta_B}(t) = \frac{g}{8 \Omega_1} S_{BB}(0)$ and $\Gamma_{\delta_{\Omega_1}}(t) = \frac{g}{8 \Omega_1} S_{\Omega_1 \Omega_1}(0)$, where $S_{BB}(\omega)$ and $S_{\Omega_1 \Omega_1}(\omega)$ are the power spectra of the magnetic noise and the noise of the first driving field, respectively. Hence, increasing values of $g$ result in increasing dephasing rates. This was verified in simulations (see Supplementary Note 8), and to
some extent experimentally as shown in Supplementary Fig. 5 and in Fig. 4 of main text. Indeed, for small enough values of $g$ this second-order effect is negligible and the signal prolongs the coherence time. For large values of $g$ the coherence time is decreased, even below the coherence time of the doubly-dressed states.

In order to circumvent this problem, we tune the system such that $\omega_s = \omega_0 + \Omega_1 + \Omega_2$ instead of $\omega_s = \omega_0 + \Omega_1 + \Omega_2^2$, which means that we now utilize a different transition in order to couple between the doubly-dressed states (see Supplementary Fig. 1b). In this case, in the first IP with respect to $H_0 = \frac{\Omega_1}{2} \sigma_z$, and after making the RWA and moving to the basis of the dressed states we have that

$$H_I = \frac{\Omega_1}{2} \left( 1 + \delta\Omega_2 (t) \right) \sigma_z + \frac{\Omega_2}{2} \left( 1 + \delta\Omega_2 (t) \right) \cos (\Omega_1 t) \sigma_y + \frac{g}{2} \left( \cos \left( \frac{\Omega_2}{2} t + \varphi \right) \sigma_z + \sin \left( \frac{\Omega_2}{2} t + \varphi \right) \sigma_y \right) - \delta\Omega_1 (t) \sigma_z. \quad (12)$$

Moving now to the second IP with respect to $H_{01} = \frac{\Omega_1}{2} \sigma_z$ (in the basis of the dressed states) and taking the RWA we obtain

$$H_{II} = \frac{\delta\Omega_1 (t) \Omega_1}{2} \sigma_z + \frac{\Omega_2}{4} \left( 1 + \delta\Omega_2 (t) \right) \sigma_y + \frac{g}{2} \cos \left( \frac{\Omega_2}{2} t + \varphi \right) \sigma_z, \quad (13)$$

where we neglected the (fast rotating) terms of the magnetic noise. In the basis of the doubly-dressed states, the $\sigma_z$ eigenstates ($y \rightarrow z, z \rightarrow -y, x \rightarrow x$), the Hamiltonian is given by

$$H_{II} = \frac{\Omega_1}{4} \left( 1 + \delta\Omega_2 (t) \right) \sigma_z - \frac{\delta\Omega_1 (t) \Omega_1}{4} \sigma_y - \frac{g}{2} \cos \left( \frac{\Omega_2}{2} t + \varphi \right) \sigma_z. \quad (14)$$

Finally, in the third IP with respect to $H_{02} = \frac{\Omega_1}{2} \sigma_z$ (in the basis of the doubly-dressed states), and after making the RWA we have that

$$H_{III} = \frac{g}{4} \left( i\sigma_x e^{-i\varphi} - i\sigma_x e^{+i\varphi} \right) + \frac{\delta\Omega_2 (t) \Omega_2}{4} \sigma_z, \quad (15)$$

Here, the calculation of the effective Hamiltonian of the counter-rotating terms yields (taking $\varphi = 0$ again)

$$H_{III} = -\frac{g}{4} \sigma_x + \frac{\delta\Omega_2 (t) \Omega_2}{4} \sigma_z + \frac{\delta\Omega_1 (t) \Omega_1}{8 \Omega_1} \sigma_z. \quad (16)$$

In this case the second order contribution of the driving noise, $g\delta\Omega_2 (t)$, vanishes, and because the second order contribution of the magnetic noise, $g\delta\Omega_1 (t)$, is perpendicular to the signal, it contributes in higher orders only (assuming $\Omega_1 \gg |\delta\Omega_1 (t)|$). Therefore, the improved scheme results in even prolonged coherence times and enables the measurement of stronger signals. Moreover, the measured signal in this scheme is stronger by a factor of 2 compared to the original scheme. A comparison between the schemes is shown in Supplementary Note 8 where the results of the simulations suggest that the improved scheme may increase the coherence time by an additional order of magnitude.
Supplementary Note 4: Detailed level scheme and setup

The diamond used for this measurements contains natural abundance of $^{13}\text{C}$ (1.1%) and the selected nitrogen-vacancy (NV) was situated roughly 2 µm below the surface. All the measurements are performed at a static magnetic field of $B_{\text{bias}} = 446$ G, where the Nitrogen nuclear spin of the NV is polarized [4], so that the hyperfine transitions appearing from the coupling to the $^{14}\text{N}$ do not intervene with the protocols. The static magnetic field is aligned parallel to the quantization axis of the NV centre (connecting the nitrogen and the vacancy) and is defined as the z-axis of the system. Supplementary Fig. 1a shows an optically detected magnetic resonance (ODMR) measurement from where $\omega_0$ is determined. A more detailed level scheme containing the effect of two applied drives to the TLS of the NV centre is depicted in Supplementary Fig. 1b.

By switching on the first drive $\Omega_1$ a dressed state configuration is obtained, where the new eigenstates are separated by $\Omega_1$. In the single drive configuration the energy levels $\omega_1$ and $\omega_0$ are susceptible to external signal. The dressed states are decoupled from external magnetic noise $\delta B$, but suffer mainly from the drive noise $\delta \Omega_1$. To decouple the sensor from $\delta \Omega_1$ noise, a second drive $\Omega_2$ of the order of $\delta \Omega_1$ is applied, which drives effectively the appearing transitions $\omega_1$ and $\omega_0$ due to

$$\sin \left( \frac{\omega_0 + \pi}{2} \right) \sin \left( \frac{\Omega_1 + \pi}{2} \right) = \frac{1}{2} \left( \cos \left( \omega_0 - \Omega_1 \right) + \cos \left( \omega_0 + \Omega_1 \right) \right)$$

leading to a doubly-dressed state configuration. Here, four new energy gaps $\omega_j = \omega_0 \pm \omega_1 \pm \omega'_2$ are opened, which can be addressed by an external signal. Note that due to a change in interaction pictures $\omega'_2 = \omega_2/2$. Consequently, the doubly-dressed state suffers mainly from noise contributions of $\delta \Omega_2$.

This concatenation of drives can be continued and will increase the coherence time of the sensor until the noise $\delta \Omega_2$ of the additional applied i-th drive field is on the order of the drive $\Omega_i$ itself, or until $T_1$ time of the bare states of the sensor is reached.

Both drives, $\Omega_1$ and $\Omega_2$, are sampled and outputted from one channel of an AWG (arbitrary waveform generator, Keysight M8195A) with a time resolution of 65 GS/s. The DAC (digital analog converter) in this device has a resolution of 8 bit per channel at maximum 1 V (peak-to-peak). As an external signal source, the Rohde&Schwarz SMIQ03B was used.
Supplementary Note 5: Calculating the slope

The sensor obtains a phase $\phi$ during a time $t$ which can be written as

$$\phi(t) = \int_0^t \gamma_N \alpha B \, d\tau = \gamma_N \alpha B \cdot t$$

(18)

if $B$ remains constant over time. $\alpha$ is a constant factor, which depends on the measurement scheme (and determines the rate of phase accumulation). For a measurement of $B$ under single drive we can set $\alpha_{sd} = 1/2$ and for the double drive case $\alpha_{dd} = 1/4$. This can be seen in equation (9), where the rate at which the signal is recorded is reduced in the double drive case to $(g/2) \cdot \alpha_{dd} = g/8$.

In the measurement a normalized fluorescence signal $S$ is recorded, where the state $\phi = 0$ is associated with the bright state of the NV (which corresponds to a normalized fluorescence value $b$). The state $\phi = \pi$ is the dark state of the NV (which corresponds to a normalized fluorescence value $d$). Therefore the signal accumulation corresponds to

$$S = \left(\frac{b+d}{2}\right) + \left(\frac{b-d}{2}\right) \cos(2\phi)$$

(19)

since for a TLS we have $S \propto \cos^2(\phi)$.

An unknown magnetic field amplitude $B$ can be extracted by performing a measurement of $S$. The uncertainty $\delta S$ of the signal $S$ will eventually determine the error in $\delta B$, which are connected by

$$\delta S = \frac{\partial S}{\partial B} \delta B$$

(20)

The sensor is the most sensitive to a small change in the magnetic field at the point where the signal $S$ has the maximum change, which is the maximal slope

$$\max \left| \frac{\partial S}{\partial B} \right| = \max \left| \left(\frac{b-d}{2}\right) \sin(2\gamma_N \alpha B \tau) \cdot 2\gamma_N \alpha \tau \right| = (b-d)\gamma_N \alpha \tau = \omega_S C$$

(21)

Here the full amplitude $C = (b-d)$ of the signal and the rate of change $\omega_S = \gamma_N \alpha \tau$ in $S$ were introduced. Since $S$ represents a normalized value (normalized with the bright state of the sensor) $C$ directly corresponds to the contrast of the signal. Note that the time (for one measurement run) $\tau$ will determine the magnitude of the slope.

To obtain the state of the NV we will count photons. Since the number of photons in one measurement run $\tau$ is very small for a NV, we will have to repeat the measurement $N = t/\tau$ times. Each photon record becomes an independent measurement and the uncertainty of the state signal $S$ is Poissonian distributed, $\delta S(t) = 1/\sqrt{N_{ph}} \cdot N = \sigma(t)$. $N_{ph}$ represent the amount of photons counted in one experimental run $\tau$, which can be understood as $N_{ph} = \Gamma c \cdot \tau$, with $\Gamma c$ being the count rate.

In the measurement we will accumulate a signal $S$ for a time $t$ and determine the standard deviation $\sigma(t)$ of the accumulated signal over time. Therefore the minimum resolvable magnetic field writes

$$\delta B_{\text{min}}(t, \tau) = \frac{\delta S}{\max \left| \frac{\partial S}{\partial B} \right|} = \frac{1}{\gamma_N} \frac{\sigma(t)}{\alpha \tau C} = \frac{1}{\gamma_N} \frac{\alpha C}{\sqrt{N_{ph}}} \frac{1}{\sqrt{\tau}}$$

(22)

The sensitivity $\eta$ for a repetitive measurement at $\tau$ after time $t$ (i.e. after repeating the measurement $N = t/\tau$ times) is consequently

$$\eta(\tau) = \delta B_{\text{min}}(t, \tau) \sqrt{\tau} = \frac{1}{\gamma_N} \frac{1}{\alpha C} \frac{1}{\sqrt{N_{ph}}} \frac{1}{\sqrt{\tau}}$$

(23)

As the signal of the sensor decays with $t$ resulting in a reduced contrast $C$, the optimal measurement point $\tau$ will be situated at the coherence time $T_2$, where also the contrast $C$ of the signal should be determined and we will end up in

$$\eta(T_2) = \frac{\hbar}{g \mu_B \alpha C} \frac{1}{\sqrt{N_{ph}}} \frac{1}{\sqrt{T_2}}$$

(24)
where $\gamma_{NV} = g\mu_B/\bar{h}$ was used. A similar derivation can be found in [5]. Note that in general the contrast $C = C(T_2)$ of the signal is depending on the coherence time of the sensor, which mean that it will decay for a time $t > T_2$, which will cause a worst sensitivity $\eta$. The appearing constants $1/(\alpha C)$ are measurement related values.

Supplementary Fig. 2a shows a measurement to determine the maximal slope $|\partial S/\partial B|$ in the double drive case, where an externally applied signal was varied in strength $g$. With this result it is possible to obtain via $\delta B_{\text{min}} = (2\pi g_{\text{min}})/\gamma_{NV}$ directly $\delta B_{\text{min}}(t, \tau)$ by measuring at the point of the maximal slope $\sigma(t)$ over time $t$. The result is plotted in Fig. 3 of the main text.

**Supplementary Figure 2.** Two different approaches to determine $\delta B_{\text{min}}$. (a) Direct slope measurement in double drive at $\tau = 250\mu s$ by varying the applied signal strength $g$. From this measurement the maximal slope $|\partial S/\partial B|_{\text{max}}$ can be obtained. (b) Coherence time measurement in double drive with a signal $g$. A signal of strength $g/2\pi \approx 41$ kHz was applied during a double drive measurement with $\Omega_1/2\pi = 3.366$ MHz and $\Omega_2/2\pi = 519.5$ kHz. In the double drive $g'' = g/4$ is measured. The total measurement took about 1 day (where a signal-to-noise ratio of $\approx 66$ was obtained).

Alternatively, it is possible to obtain $\delta B_{\text{min}}(t, \tau)$ by measuring the coherence time under drive with a signal, $\tau = T_{\Omega_1,\Omega_2,g}$, and the resulting contrast, $C$, at the time $T_{\Omega_1,\Omega_2,g}$ like in Supplementary Fig. 2b, to obtain the denominator of equation (22).
Supplementary Note 6: Determine the optimal drive parameters

A. Optimal drive parameter in single drive

Before an external high frequency signal can be measured suitable values for the drive fields \( \Omega_1 \) and \( \Omega_2 \) have to be chosen, to obtain the maximal coherence time of the sensor. Since the first drive \( \Omega_1 \) increases the decoupling of the sensor from the environment but introduces drive noise \( \delta \Omega_1 \) with a stronger drive \( \Omega_1 \), an optimal value has to be selected. The optimal value should prolong the coherence time of the sensor as much as possible for a given drive configuration. Supplementary Fig. 3 shows Rabi (= single drive) measurements with different drive strength, \( \Omega_1 \), displayed against the extracted coherence time of the sensor under drive, \( T_{2\Omega_1} \). It becomes obvious that for a slower drive (<1 MHz) the coherence time of the sensor, \( T_{2\Omega_1} \), is not increasing monotonically. One of the reason for this is a less efficient decoupling by \( \Omega_1 \) allowing various interactions between individual nearby \( ^{13}\text{C} \) nuclear spins and the overall \( ^{13}\text{C} \) bath contribution. For a static magnetic field of \( B = 446 \) G and \( \gamma_{^{13}\text{C}}/2\pi = 1.0705 \) kHz/G we expect the Larmor frequencies of the \( ^{13}\text{C} \) bath to be around \( \nu_{\text{Larmor}^{13}\text{C}} = \gamma_{^{13}\text{C}}B \approx 477 \) kHz, which fits well with the large dip at around 500 kHz. To use the dressed states as a high frequency sensor under a single drive, we select \( \Omega_1/2\pi > 2.5 \) MHz based on Supplementary Fig. 3 to make sure that a sufficient decoupling from the environment can be guaranteed. Compared to the pure dephasing time of this NV with \( T_{2}^* = 1.1 \) µs, the single drive with \( \Omega_1/2\pi = 2.5 \) MHz improves the coherence time already by about a factor of 60.
B. Optimal drive parameter in double drive

In principle, we could select for the double drive experiments a very strong first drive $\Omega_1$, but then a stronger second drive $\Omega_2$ is required to correct the noise on the order of $\delta \Omega_1$. Consequently, a balanced first drive $\Omega_1$ (which maximizes Supplementary Fig. 3) will eventually introduce less drive noise $\Omega_2$, and prolong the coherence time of the sensor. For a moderate first drive $\Omega_1/2\pi = 3.363\text{MHz}$, the second drive $\Omega_2$ was scanned in Supplementary Fig. 4 in order to find the optimal choice for $\Omega_2$. This scan of $\Omega_2$ has to be done, if the exact magnitude or characteristics of $\delta \Omega_1$ is not known. For each measurement point

![Supplementary Figure 4. Measurement of the coherence time, $T_{2}^{\Omega_1,\Omega_2}$, under two drive fields, $\Omega_1$ and $\Omega_2$. The first drive was set to $\Omega_1/2\pi = 3.363\text{MHz}$ and the second drive, $\Omega_2$, is incrementally increased. Since the measurement is sampled at $\tau_{\Omega_1}$ (see text), we obtain solely a decaying oscillation with $\Omega_2 = \Omega_2/2$, hence, the recorded curve was fitted to $f(t) = c + \alpha \sin (\Omega_2 t + \phi) \exp(t/T_{2}^{\Omega_1,\Omega_2})$ to extract the coherence time $T_{2}^{\Omega_1,\Omega_2}$. The error bars represent the standard deviation $\Delta T_{2}^{\Omega_1,\Omega_2}$ of the coherence time in the fit. The optimal second drive, $\Omega_2$, maximizes eventually the coherence time $T_{2}^{\Omega_1,\Omega_2}$ of the sensor.

A decaying trace was recorded, which was (under)sampled at multiples of $\tau_{\Omega_1} = 2\pi/\Omega_1$, i.e. we measure at times $t = N\tau_{\Omega_1}$ ($N \in \mathbb{N}$), which removes the effect of $\Omega_1$ on the recorded traces.

Note that the measured coherence time in Supplementary Fig. 2b is longer compared to the maximal coherence time obtained in Supplementary Fig. 4. In Supplementary Note 7 it is shown that an applied signal with strength, $g$, acting partially as a third drive, can further increase the coherence time of the sensor for certain values of $g$.}
Supplementary Note 7: Determine the coherence time of the single drive under an increasing signal

To show the impact of an external signal strength $g$ on the coherence time of the sensor under drive, we perform a single drive measurement (corresponding to one point in Supplementary Fig. 3) by now varying the external signal strength $g$. We set $\Omega_1/2\pi = 3.002$ MHz and increase gradually $g$. For each measurement point a decaying trace was recorded, which was (under)sampled at multiples of $\tau_0 = 2\pi/\Omega_1$, i.e. we measure at times $t = N\tau_0$ ($N \in \mathbb{N}$), which removes the effect of $\Omega_1$ on the recorded traces. Consequently, $g'$ was obtained by fitting the data to an exponential decaying sine function. For small values of $g$, recording the oscillations of $g'$, and subsequently the coherence time proved to be a challenging measurement, mainly because the period of $g'$ was longer than the coherence time $T_{2g}'$, so it was difficult to distinguish the two. This caused the measured coherence time to be shorter than expected, given we would expect the coherence time at small $g$ to converge with the measurements without signal (Supplementary Fig. 3), thus having larger error bars, $\Delta T_{2g}'$, to compensate the ambiguity. Interestingly, the signal acts to a certain extend as a double drive and prolongs the coherence time which is pointed out in Supplementary Note 3. Comparing Supplementary Fig. 4 and Supplementary Fig. 5 two main differences arise.

First, the maximal coherence times in Supplementary Fig. 5 is reached at a different ratio ($g/\Omega_1 \approx 0.0083$) and second, the measured coherence time at this point is about the half it was by using $\Omega_2$. This can be explained by the following.

The signal $g$ is not phase locked to the sensor, therefore we would expect that on average $\Sigma G(t) = \Sigma g \cos^2(\omega_0 t + \langle \phi \rangle) \approx \frac{1}{2} g$ is contributing to the sensor. A phase locked signal, however, will completely contribute to the decoupling with $g$, since $\phi$ does not vary between each measurement run, which justifies the discrepancy on the $y$ axis in the graphs.

Furthermore, it seems that $\Omega_2$ introduces more noise to the system than $g$ does. As a consequence, $\delta \Omega_2$ is larger and a stronger second drive is needed to cope for the resulting overall noise level.

On the basis of a very rough estimation, we can give some upper limits for the amplitude noise of the microwave fields, which are mentioned in Supplementary Note 4. For the maximal output range of 1 Vpp we can assume 1 Vpp/2$^8 \approx 0.004$ Vpp to be the absolute noise level for this device (Keysight M8195A). $\Omega_1$ was created by 0.75V pp and based on Supplementary Fig. 4 the optimal second drive has to be $\Omega_2 = 0.15 \Omega_1 \approx 0.1125$ Vpp. Eventually, $\Omega_2$ suffers from a larger relative noise level (0.004 Vpp/0.1125 Vpp $\approx 3.5\%$) then the first drive $\Omega_1$ (0.004 Vpp/0.75 Vpp $\approx 0.4\%$). This statement can be further...
confirmed by comparing the relative noise \( \delta \Omega_2 / \Omega_2 \) with \( \delta g / g \). The external signal was produced by a microwave generator (R&S SMIQ03B), which runs continuously during the measurements and has a more than 100 times cleaner signal (-70 dBC, relative amplitude noise ratio 0.0003), compared to \( \delta \Omega_2 / \Omega_2 \). This indicates also why the coherence time in Fig. 2 of the main text could be prolonged with \( g \) by such a great amount, since the relative (and absolute) noise of the signal \( g \) seems to be much smaller than \( \delta \Omega_2 \). Eventually, a coherence time of 1.5 ms should be almost the limit to which we could extend the coherence time, since the lifetime of the sensor, \( T_1 \), was about 3 ms.

### Supplementary Note 8: Simulations

In this section we present the results of simulations aimed at reproducing the experimental results and verifying our theoretical model.

#### A. Magnetic noise

The NV centre spin used in our experiment had a pure dephasing time of \( T^*_2 = 1.1 \mu s \), and under a Hahn Echo pulse, a coherence time of \( T_2 = 515 \mu s \). In our model, the magnetic noise has two components. One component, \( B_r \), is a random field that is static within each experiment, but has a different value in different experiments, and a second component, \( B(t) \), which is an Ornstein-Uhlenbeck process \([6, 7]\) with a zero expectation value, \( \langle B(t) \rangle = 0 \), and a correlation function \( \langle B(t) B(t') \rangle = \frac{\sigma^2}{2} e^{-|t-t'|/\tau} \). \( B_r \) is normally distributed with a variance of \( \sigma^2 \approx 0.96 \times 10^{-2} \), where \( T^*_2 = 1.1 \mu s \). An exact simulation algorithm \([8]\) was employed to realize the Ornstein-Uhlenbeck process, which according to

\[
B(t + \Delta t) = B(t)e^{-\frac{\Delta t}{\tau}} + n\sqrt{\frac{c}{2}} \left(1 - e^{-\frac{2\Delta t}{\tau}}\right),
\]

(25)

where \( n \) is a unit Gaussian random number. The correlation time of the noise was set to \( \tau = 1/\gamma = 10 \mu s \), where the diffusion constant is given by \( c \approx 2 T^*_2 \) and corresponds to \( S_{BB}(0) \approx \frac{2}{\pi T^*_2} \). For pure dephasing, we simulated the Hamiltonian

\[
H = \frac{\omega_0}{2} \sigma_z + \left(B_r + B(t)\right) \sigma_z,
\]

(26)

where we used \( \omega_0 = 100 \text{ MHz} \) and the qubit was initialized to \(| \uparrow \rangle \). The result of the simulation is shown in Supplementary Fig. 6a and corresponds to a pure dephasing time of \( T^*_2 = 1.1 \mu s \). For the Hahn echo pulse we run the same simulation but flipped the sign of \( (B_r + B(t)) \sigma_z \) at \( t = 515/2 \mu s \). The result is shown in Supplementary Fig. 6b and corresponds to a Hahn echo time of \( T_2 = 515 \mu s \).

#### B. Single drive

We simulated the single-drive scenario under the RWA, so the Hamiltonian is given by

\[
H = \frac{\Omega_1}{2} \left(1 + \delta \Omega_i(t)\right) \sigma_z + \left(B_r + B(t)\right) \sigma_z.
\]

(27)

Driving fluctuations were also modelled by an an Ornstein-Uhlenbeck process with a zero expectation value. We chose a correlation time of \( \tau_{\Omega_1} = 500 \mu s \), and a relative amplitude error of \( \delta \Omega_1 = 0.15\% \) so the diffusion constant is given by \( c_{\Omega_1} = 2 \delta \Omega_1 / \tau_{\Omega_1} \). We set \( \Omega_1 = (2\pi) \cdot 3.4 \text{ MHz} \), and the qubit was initialized to \(| \uparrow \rangle \). The result of the simulation is shown in Supplementary Fig. 6c and indicates a coherence time of \( T^*_2 \approx 33 \mu s \), which is in agreement with the experimental results.
Supplementary Figure 6. Simulations of coherence times. (a) Oscillations between |↑⟩ and |↓⟩ averaged over 5000 trails. $1 + \exp(-g^2/2)$ is plotted in green ($g^2 = 2/(T_2^2)^2$). (b) Probability of being in the initial |↑⟩ state averaged over 2400 trails. Hahn echo pulse at $t = 515/2 \mu s$ and refocusing at $T_2 = 515 \mu s$. Dashed horizontal line at $P = (1 + 1/e)/2$. (c) Coherence time under a single drive with a Rabi frequency of $\Omega_1 = 2\pi \cdot 3.4$ MHz. Oscillations between |↑⟩ and |↓⟩ averaged over 800 trails. Dashed horizontal line at $P = (1 + 1/e)/2$. The simulation indicates a coherence time of $T_1^\Omega_1 \approx 33 \mu s$. (d) Coherence time under a double drive with Rabi frequencies of $\Omega_1 = 2\pi \cdot 3.4$ MHz and $\Omega_2 = 0.15 \cdot \Omega_1$. Oscillations between |↑⟩ and |↓⟩ averaged over 800 trails. Dashed horizontal line at $P = (1 + 1/e)/2$. The simulation indicates a coherence time of $T_2^{\Omega_1,\Omega_2} \approx 380 \mu s$.

C. Double drive

We simulated the double drive scenario in the first IP, making the RWA with respect to $\omega_0$ only. The Hamiltonian is given by

$$H = \frac{\Omega_1}{2} \left( 1 + \delta_{\Omega_1}(t) \right) \sigma_x + \frac{\Omega_2}{2} \left( 1 + \delta_{\Omega_2}(t) \right) \cos(\Omega t) \sigma_y + B_A + B(t) \sigma_z. \quad (28)$$

Due to some technical issues, in our experimental system the noise in $\Omega_2$ was stronger than the noise in $\Omega_1$. We therefore set $\delta_{\Omega_1} = 0.15\%$ and $\delta_{\Omega_2} = 0.21\%$, where $\Omega_1 = 2\pi \cdot 3.4$ MHz (like in the single-drive case) and $\Omega_2 = 0.15 \cdot \Omega_1$. The spin was initialized to $|\uparrow\rangle$. The result of the simulation is shown in Supplementary Fig. 6d and indicates a coherence time of $T_2^{\Omega_1,\Omega_2} \approx 380 \mu s$, which is in agreement with the experimental results.
D. Double drive with a signal

In this section we used the experimental parameters used in the experiment of Supplementary Fig. 2b, where a signal of \( g/2\pi = 41.084 \text{ kHz} \) is sensed by the doubly-dressed qubit. The simulation result is shown in Supplementary Fig. 7a and indicates a coherence time of \( T_{\Omega_{1,\Omega_{2}}}^{\Omega_{2}} \approx 450 \mu\text{s} \), which is shorter than the experimental value of \( T_{\Omega_{1,\Omega_{2}}}^{\Omega_{2}} \approx 600 \mu\text{s} \). This could be because our theoretical model of the magnetic noise does not provide a complete characterization of the actual noise, and in fact describes a more severe situation. In principle, the actual spectrum of the noise can be measured [9], which would allow for a theoretical optimization of the parameters used in the sensing protocol. We then varied the value of \( g \) in order to verify its effect on the coherence time. For Supplementary Fig. 7b (Supplementary Fig. 7c) we decreased (increased) the value of \( g \) by a factor of 2, and used \( g/2\pi = 0.5 \cdot 41.084 \text{ kHz} \) (\( g/2\pi = 2 \cdot 41.084 \text{ kHz} \)). For the smaller (larger) value of \( g \) the coherence time was increased (decreased), as expected. We then used the original \( g \) value of \( g/2\pi = 41.084 \text{ kHz} \), and simulated the improved scheme with \( \omega_s = \omega_0 + \Omega_{2}^{\Omega_{2}} \). The result of this simulation is shown in Supplementary Fig. 7d. The simulation confirms the improved performance of the scheme, and indicates an improvement of the coherence time by \( \sim 1 \) order of magnitude.

**Supplementary Figure 7.** High-frequency signal sensing by a double drive. Dashed horizontal line at \( P = (1 \pm 1/e)/2 \). (a) Simulation of high-frequency signal sensing by a double drive with the experimental parameters used in the experiment of Supplementary Fig. 2b. (b) A weaker signal increases the coherence time. All parameters are identical to those of Supplementary Fig. 7a except \( g \), which is decreased by a factor of 2. (c) A stronger signal decreases the coherence time. All parameters are identical to those of Supplementary Fig. 7a except \( g \), which is increased by a factor of 2. (d) Improved scheme. All parameters are identical to those of Supplementary Fig. 7a.
Supplementary Note 9: Pulsed analog of the scheme

The sensing of high frequency fields with a TLS could also be achieved with a pulsed dynamical decoupling analog of our scheme. This could be seen by considering an AC signal with a frequency of $\omega_s = \omega_0 + \Omega$ (similar to the single-drive case). The Hamiltonian of the system under the AC signal is given by

$$H = \frac{\omega_0}{2} \sigma_z + g \sigma_x \cos(\omega_s t),$$

and in the interaction picture with respect to $H_0 = \frac{\omega_0}{2} \sigma_z$ we have that

$$H_I = \frac{g}{2} \left( \sigma_x e^{-i\Omega t} + \sigma_- e^{+i\Omega t} \right) = \frac{g}{2} (\cos(\Omega t) \sigma_x + \sin(\Omega t) \sigma_y).$$

Hence, the signal can be measured by initializing the spin to $|\uparrow_x\rangle$ and applying a CPMG pulse sequence, where the pulses correspond to $\pi$ rotations around the $\hat{y}$ axis (so the $\sigma_y$ part of the signal is measured). Similar to the common pulsed dynamical decoupling sensing methods, the rate of the pulses should match $\Omega$ in order for the signal $g$ to be observed.

Supplementary References

Chapter 6

Concatenated continuous dynamical decoupling of a three-level system

If it is important enough to you, you will find a way.
If it is not, you will find an excuse.

– Ryan Blair
6 Concatenated continuous dynamical decoupling of a three-level system

6.1 Introduction

The present chapter focuses on continuous dynamical decoupling and the creation of doubly-dressed states to protect a qubit from the drive and environmental noise. However, the idea followed in this chapter reflects an alternative approach to Chapter 5, in order to avoid the necessity of having many concatenated drive fields to create the robust qubit. Only two subsequent concatenations ensure a protected subspace by utilizing all three available spin sub-levels of the NV center.

In this way, the robustness of the created qubit against environmental noise is solely controlled by the strength of the applied microwave fields, whereas the scheme itself ensures robustness to drive noise. In addition, high frequency sensing remains possible and faster qubit gate operations are predicted. The new approach utilizes AC-Stark shifted energy gaps produced by off-resonant driving fields.

First, we investigated a scheme that utilizes only off-resonant driving fields (see. Sec. 6.4). It turned out that the energy gaps created by these fields are too small and thus further increase of the drive amplitude would face limitations imposed by the actual experimental configuration. Hence, we combined on-resonant and off-resonant drive fields to circumvent the experimental constraints thereby allowing a larger energy gap (and thereby decoupling it from environmental noise) while maintaining robustness against drive noise.

Sec. 6.2 summarizes the findings and demonstrates an implementation, which prolongs the coherence time of a protected qubit state realized by four drive fields applied on the three level system of the NV ground state. The supplementary part in Sec. 6.3 contains the background information for the measurement setup and the parameters characterizing the system.
INTRODUCTION

The reliable and efficient construction and manipulation of qubits is necessary for the implementation of quantum technology applications and quantum information processing. In solid state and atomic systems, ambient magnetic field fluctuations constitute a serious impediment, which usually limits the coherence time to a small fraction of the lifetime limit. Pulsed dynamical decoupling [1–3] has proven to be very useful in prolonging the coherence time [4–12]. However, in order to mitigate both, environmental and controller noise, very rapid and composite pulse sequences must be applied [13–17], which are not easily incorporated into other operations and require a large field strengths [18]. Continuous dynamical decoupling [18–26] shows another way of suppressing environmental noise by diminishing the effect of the controller noise. For instance, a rotary echo scheme [27, 28] can be viewed as an analog to pulsed dynamical decoupling. The concatenation of several on-resonance driving fields [29–33] is another concept, but inherently connected to a reduction of the dressed energy gap, eventually limiting the performance of the scheme. All these techniques have in common that the operation time of qubit gates is reduced. Multi-state systems allow for a different approach.

In [34], a fully robust qubit - a qubit that is robust to both external and controller noise - was realized by the application of continuous driving fields on a specific hyperfine structure. Subsequently, a general scheme for the construction of a fully robust qubit was introduced in [35]. However, the multi-state schemes, which utilize on-resonance driving fields, are not applicable to a three-level system. A protected qubit subspace within that configuration can be realized by the application of off-resonant driving fields [36], but such a construction necessitates the application of four strong driving fields making the experimental realization challenging.

In this letter we demonstrate how a fully robust qubit can be constructed by utilizing a three-level system through the application of both, continuous on-resonant and off-resonant driving fields. The on-resonant driving fields result in robustness to magnetic noise whereas off-resonant driving fields facilitate robustness against driving noise, which typically is the problem of continuous dynamical decoupling schemes. Moreover, this scheme is applicable to both optical and microwave domain and enables the implementation of fast and simple qubit gates [36]. Three level-systems are widely available and appear in many atomic and solid state systems, such as trapped ions, rare-earth ions, defect centers, and in particular, the NV-center in diamond. We demonstrate concatenated continuous dynamical decoupling of a three-level system realized by the ground state spin level of the nitrogen-vacancy center (NV) in diamond. The states are addressed by a combination of four microwave fields, tuned to the same Rabi frequency, Ω, at different transitions frequencies, ω1 and ω2, and detuning, Δ. The states of the NV center are read out and initialized by a 532nm laser identifying spin dependent fluorescence between its spin states [37–40]. We are operating in the vicinity of the excited state level anti-crossing [41, 42] at a magnetic field of 35.8 mT. At this field the intrinsic nitrogen nuclear spin becomes polarized and does not contribute to the level structure. In the following, we establish a theoretical model, which predicts an improvement in coherence time of the robust qubit. An experimental real-
 Concatenated continuous dynamical decoupling of a three-level system

THE SCHEME

We consider a three-level system with states $|0\rangle$ and $|\pm 1\rangle$, where the $|\pm 1\rangle$ states are dipole coupled to the $|0\rangle$ state, as illustrated in Fig. 1a. The energy gaps ($h = 1$) between the $|0\rangle$ state and the $|\pm 1\rangle$ states are $\omega_1$ and $\omega_2$. Our scheme utilized four drive fields which can be expressed by the driving Hamiltonian

$$H = \sqrt{2}\Omega_1 \left( \cos(\omega_1 t) |0\rangle\langle -1| - \cos(\omega_2 t) |0\rangle\langle 1| + \text{h.c.} \right) + \sqrt{2}\Omega_2 \left( \cos((\omega_1 + \Delta t)/2) |0\rangle\langle -1| + \cos(\omega_2 + \Delta t)/2 |0\rangle\langle 1| + \text{h.c.} \right).$$

Moving to the interaction picture (IP) with respect to $H_{01} = \omega_1 |1\rangle\langle -1| + \omega_2 |1\rangle\langle 1| and changing the basis to $\{|0\rangle, |B\rangle, |D\rangle\}$, with $|B\rangle = (|1\rangle + |\pm 1\rangle)/\sqrt{2}$ and $|D\rangle = (|1\rangle - |\pm 1\rangle)/\sqrt{2}$, we obtain after applying the rotating-wave approximation the Hamiltonian

$$H_I = \Omega_1 (|0\rangle\langle D| + |D\rangle\langle 0|) + \Omega_2 (|0\rangle\langle B| e^{i\Delta t} + |B\rangle\langle 0| e^{-i\Delta t}),$$

which can be illustrated by the level scheme depicted in Fig. 2a. The states $|0\rangle$ and $|D\rangle$ are on-resonantly coupled in a single lambda (SL) drive with a strength $\Omega_{SL} = \sqrt{2}\Omega_1$ and, under the assumption of $\Delta > \Omega_1$, an off-resonant coupling between $|0\rangle$ and $|B\rangle$ by $\Omega_{SL}^\text{off}$ is obtained.

The drive, $\Omega_{SL}$, in Fig. 2a transforms into the dressed states $\{|u\rangle, |B\rangle, |D\rangle\}$, as schematically illustrated in Fig. 2b, with the corresponding eigenvalues $\{+\Omega_1, 0, -\Omega_1\}$, respectively. Here, we introduced the $|u\rangle = (|0\rangle + |D\rangle)/\sqrt{2}$ and $|d\rangle = (|0\rangle - |D\rangle)/\sqrt{2}$.

For a large enough drive, $\Omega_1$, robustness to magnetic noise is obtained. As the magnetic noise, $\delta B$, couples between the $|B\rangle$ state and the $|u\rangle$ and $|d\rangle$ states, we can set $\Omega$ such that $S_{BB}(\omega) \ll 1/\Gamma_1$, to ensure that the first order effect of the noise is negligible. In this case $S_{BB}(\omega)$ denotes the power spectrum of the noise and $\Gamma_1$ the lifetime of the system.

The coherence time of the dressed states is mainly limited by driving amplitude fluctuations, $\Omega \rightarrow \Omega(1 + \delta(t))$, where $\delta(t)$ represents the random noise contribution. In order to obtain as well robustness to driving fluctuations, we consider the effect of the second detuned drive, $\Omega_{SL}^\text{off}$, on the dressed states. Therefore, we continue by moving to the basis of the dressed states and to the IP with respect to $H_{02} = \sqrt{2}|B\rangle\langle B|$ and obtain

$$H_{11} = k (|u\rangle\langle u| - |d\rangle\langle d| - \Delta |B\rangle\langle B| + \Omega_2 \sqrt{2} (|B\rangle\langle u| + |B\rangle\langle d|) + \text{h.c.}).$$

The eigenstates of $H_{11}$, denoted by $\{|u\rangle, |B\rangle, |d\rangle\}$, are termed as the doubly-dressed states and are illustrated in Fig. 2c.

Drive fluctuations are introduced in $H_{11}$ by replacing $\Omega_1$ by $\Omega_1 + \delta_1$, $\Omega_2$ by $\Omega_2 + \delta_2$, $\delta_1$, and $\delta_2$ of the expansion of eigenvalue $k$. For a given driving

\begin{align*}
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\end{align*}
The experimental realization of the proposed scheme follows the illustration in Fig. 1. Four drive fields are applied on the bare basis states \(\{|-1\rangle, |0\rangle, |1\rangle\}\) of the NV center (cf. Fig. 1a). The field amplitudes of the on-resonant drives are adjusted to represent the same Rabi frequency, \(\Omega\), for both transitions, \(\omega_1\) and \(\omega_2\) (see Suppl. B). The off-resonant drives are obtained by adding a detuning to the resonant ones, resulting in the total field

\[
\Omega_{\text{tot}}(t) = \Omega \cdot \left( \cos(\omega_1 t) - \cos(\omega_2 t) + \cos((\omega_1 + \Delta t) + \cos((\omega_2 + \Delta t) t) \right).
\]

synthesized by the signal generator (\(\Omega_1 = \Omega_2 = \Omega\)) and implements eq. (1). Note that the phase shift of \(\pi\) changes the sign of the second cosine term.

The interaction with the microwave field in Fig. 1a couples the drive, \(\Omega\), to the spin states. On-resonant drives in Fig. 1a create Rabi oscillations at a rate, \(\Omega_{\text{M1}}\), in Fig. 2a, and positive (negative) detuning is reflected in ‘red’ (‘blue’) detuned AC-Stark shifted energy levels of the \(|d\rangle\) and \(|B\rangle\) states in Fig. 2b, which creates in total the doubly-dressed states, depicted in Fig. 2c. The appearing energy levels in the doubly-dressed states, \(|\tilde{a}\rangle\), \(|\tilde{d}\rangle\), and \(|\tilde{B}\rangle\), are eventually all coupled to the drive fields.

![Figure 3](image-url) Expected coherence times for a drive \(\Omega_1 = \Omega_2 = 2\pi \cdot 10\, \text{MHz}\) and \(\delta_1 = \delta_2 = 0.005\). Under the assumption of no magnetic noise, the blue dotted curve predicts the position of detuning, \(\Delta/2\pi \approx 32.3\, \text{MHz}\), where drive noise of the system is eliminated. By including magnetic noise (solid orange line) with a pure dephasing time of \(T_2 = 2\, \mu\text{s}\), the coherence time reaches a limit, which is set by the second order effects of the magnetic noise, \(\delta B\), and constitutes the maximal improvement of the scheme. Inset: Plot of the maximal coherence time obtained for different drive fields, \(\Omega\).

![Figure 4](image-url) Coherence time measurement and theoretical description in a double lambda drive. The record of the coherence time relies on a Ramsey type measurement, performed in the \(|0\rangle, |1\rangle, |D\rangle\)-basis, with on-resonant and off-resonant drives, at \(\Omega/2\pi = (2.869 \pm 0.011)\, \text{MHz}\) (cf. Fig. 1b). The theory reproduces the measurement results by assuming three times greater drive noise on \(|B\rangle\) compared to \(|D\rangle\) (cf. Fig. 2a). The error bars in the graph represent \(\Delta T_2^{\Omega_B, |\rangle, \langle|}\) and originate from fitting the coherence time to \(\propto \sin(\omega t + \phi) \exp \left\{ -t/T_2^{\Omega_B, |\rangle, \langle|} \right\} \).
(see Fig. 2). The robustness of the scheme against drive noise, $\delta \Omega$, is reflected by having energy levels, which experience the same drive fluctuations. On top of that, a large energy gap, $\Omega_{RQ}$, in the doubly-dressed states, originating from on-resonant drives, ensures an efficient decoupling from external magnetic noise contributions, $\delta B$. Thus, increasing the drive fields, $\Omega$, would provide a better protection against $\delta B$ noise.

To determine the performance of the scheme the coherence time of the protected states has to be recorded as a function of detuning, in order to find the optimal AC-Stark shifted energy levels, which minimize the drive fluctuations. The measurement is performed in the dressed state basis and is analogous to a free induction decay (FID) or Ramsey measurement (cf. Fig. 1b). Here, by the application of an on-resonant $\pi/2$ pulse with $\Omega_{SL}$, a superposition is creates between the $|B\rangle$ and $|0\rangle$ states (which is also a superposition of the $|D\rangle$ and $|0\rangle$ states, just with different initial phase factor in the $|-1\rangle$ state). In the next step, the double lambda drive (depicted in Fig. 1a) is applied on the superposition states under an increased interaction time, $\tau$, revealing the present energy gaps (in Fig. 2c) as a coherent evolution. By mapping the coherences to populations with a consecutive $\pi/2$ pulse (with $\Omega_{SL}$), spin state dependent fluorescence is observed upon the application of a laser. Hence, the readout signal contains frequency components proportional to the energy gaps (between the states in Fig. 2c). The robust state becomes perceptible by the longest coherence time which is displayed in Fig. 4.

The measured coherence times are extracted by fitting a sinusoidal exponential decay to the curves. The obtained coherence time, $T_2^{\text{det}}$, is presented as a function of detuning, $\Delta$, in Fig. 4.

**DISCUSSIONS**

With the presented scheme, it is truly possible to prolong the coherence time of the NV sensor as predicted by the theory. By suppressing the drive noise, $\delta \Omega$, introduces by the four fields, only second order contributions of $\delta B$ limit the improvement in coherence time shown in Fig. 4. However, the results as well as the adapted theory for the coherence demand for a thorough discussion.

The asymmetric shape of the curve in Fig. 4 provides insights about the appearing dynamics. Starting from the limit of a very large detuning, $\Delta$, only the dressed states $|u\rangle$ and $|d\rangle$ are present in the scheme and the AC-Stark shifts are negligible (see Fig. 2b). Thus, the coherence time of the sensor is then predominantly determined by the noise, $\delta \Omega$, as $\delta B$ has a smaller impact at this drive field strength, $\Omega$ (cf. Suppl. S2.B).

Decreasing the detuning, $\Delta$, introduces Stark shifts on the $|u\rangle$, $|d\rangle$ and $|B\rangle$ states, which effectively reduces the fluctuations of the doubly-dressed state energy gap and thereby prolonging the coherence time of the sensor. Hence, the height of the peak is now solely limited by second order noise contributions from $\delta B$ (see the obtained peak in Fig. 4).

Further decrease of the detuning starts to drive $|0\rangle \leftrightarrow |B\rangle$ and introduces thereby again more drive noise, which impacts strongly on the doubly dressed states. Approaching zero detuning will eventually eliminate the doubly-dressed states, $|u\rangle$ and $|d\rangle$, and at $\Delta = 0$ only the two-level system transition, $\nu_1$, remains to be addressed by $2\Omega$. The same behavior as described above is expected for negative a detuning, $\Delta$.

The theoretical description allows us to introduce three parameters in order to mimic the experimental situation. These are the drive noise, $\delta \Omega_B$ and $\delta \Omega_D$, on the dressed states (cf. Fig. 1b), the magnetic noise, $\delta B$, and a possible one photon detuning, $\Delta_{ph}$. One crucial remark has to be made right at the beginning. The mentioned parameters do not change the asymmetric shape of the curve as it is fully determined by the model. However, an imbalance between the drive noise and a one-photon detuning change the position of the peak, whereas the magnetic noise impacts the peak height.

The measured free induction decay time, $T_2^* = (1.78 \pm 24)$ $\mu$s, sets the boundary for the peak height and the theory confirms a coherence time of about $\sim 190$ $\mu$s, where the drive noise is eliminated. By selecting a three times higher noise, $\delta \Omega_B = 3 \delta \Omega_D$, the theory curve as illustrated in Fig. 4 is obtained. Alternatively, we could have assumed a one photon detuning of $\Delta_{ph}/2 \pi = 2$ MHz and a reduced noise difference of $\delta \Omega_B = 2.5 \delta \Omega_D$ leading to the same result. In the following, it has to be clarified how these assumptions are grasped in the experiment.

First of all, it appears that the DC and AC components of the magnetic noise, $\delta B$, have equal contributions to both transitions, $\omega_1$ and $\omega_2$, as we obtain (within the error bar) the same values for $T_2^*$ and $T_2 = (215 \pm 31)$ $\mu$s (see Suppl. S2.C and S2.D). However, by comparing the coherence time of the Rabi drives, we obtain a drastic difference, $T_2^{\text{det}} = (62 \pm 12)$ $\mu$s and $T_2^{\text{det}} = (159 \pm 24)$ $\mu$s, which hints at a drive frequency dependent noise spectrum. As noise in the real world has a small but finite correlation time [45] and all the fields are produced by the same signal generator, it is valid to assume correlations in the drive noise. The combination of both effects can truly cause the noise imbalance between $\delta \Omega_B$ and $\delta \Omega_D$, which affects directly the position of the peak with respect to the detuning, $\Delta$. As this is a setup specific setting, the AC-Stark shifts have to be adjusted to compensate for that value. However, the coherence time improving effect, as predicted by the theory, is expected to be within the range of 50 MHz at the utilized drive field, $\Omega$, as larger detunings have a negligible energy shift on the states. A further and more detailed investigation of
the drive noise is performed in Suppl. S3.

CONCLUSION

In this work we demonstrate a protocol for a three-level system, which creates doubly-dressed states by means of multiple drive fields. The realization of the protocol relies on the combination of on-resonant and off-resonant drive fields. Moreover, the mere manipulation of only a three-level system simplifies the creation of the protected qubit and its integration into a potential target application. The special drive configuration creates a drive noise free qubit subspace and its robustness to environmental noise is tunable by increasing the drive of the system, $\Omega$, thus creating a fully robust qubit.

In addition, this scheme allows the task of high frequency sensing in the GHz regime, where the sensitivity would be solely limited by the coherence time of the robust qubit. The AC-Stark shifted energy gaps make it possible to implement fast qubit operations, which are only limited by the applied drive fields, $\Omega$.

All in all, the presented concept is applicable to both, optical and microwave configurations. Therefore, we believe that the scheme has potential applications in a wide range of tasks in the fields of quantum information science and quantum technologies, and in particular, quantum sensing.

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6.3 Supplementary Information

Supplementary information for:
Concatenated continuous dynamical decoupling of a three-level system

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S1. SETUP CONFIGURATION

A standard home-build confocal microscope configuration was used to address individual NV centers. The sample, hosting the NVs, is a 98.9% $^{12}$C electronic grade diamond substrate, with a 40 $\mu$m 99.8% $^{12}$C low-N grade layer, grown by means of CVD. The top side of the diamond is attached to a glass slide with a lithographically deposed microwave structure, to enable close vicinity of the microwave field for an efficient microwave drive. The glass structure with the diamond was attached to a printed circuit board (PSB) sample holder with SMA connectors for microwave field delivery. The PSB board is mounted on a xyz home-build micrometer stage to position the sample relatively to the confocal scanner. The actual nanometer precise scanning operation in the confocal setup was realized by a xyz piezo scanner, P-527.3CD from Physik Instrumente with a E-725.3CD Digital Piezo Controller, which controls the position of an Olympus UPlanFL N 100X/1.30 $\times/0.17$/FN 26.5 oil iris microscope objective.

The laser light is provided by a 2W Verdi G2-SLM OPS 532nm Laser diode, which is pulsed by an AOM from Brimrose. The red fluorescence is separated from the green illumination by a dichroic mirror FF552-Di02-25x36 from Semrock. The fluorescent response of the NV is converted with an avalanche photon diode (APD), SPCM-AQRH from Excelitas Technologies, into an electrical signal. The digitalized signal is either recorded by a custom configured XEM6310 LX45 FPGA from Opal Kelly, with a 950MHz detection bandwidth (for fast counting), or with a PCI-6232 X-series card from National Instruments (for slow counting). The hardware triggered confocal scanning operation as well as the ODMR measurements are controlled with the NI PCI-6232 card.

The creation of the microwave field is either realized with a Rohde&Schwarz SMR20 microwave signal generator (for ODMR measurements) or with an arbitrary waveform generator AWG70002A from Tektronix (which was borrowed for a small period). In both cases, the microwave field was amplified by the ZHL-16W-43 S+ high power amplifier from Mini-Circuits.

All the instruments are remote controlled by a PC, which handles the measurement procedure and data acquisition. The powerful software suite Qudi [1] serves as the operational tool to establish the control over all hardware devices and perform all measurements.

The selected NV for this experiment was about one micrometer deep in the diamond and about 10 $\mu$m away from the microwave structure. The experiment was performed in the vicinity of the excited state level anti-crossing [2–4], where the nitrogen nuclear spin becomes polarized. Hence a static magnetic field of 358G was applied to the NV center, aligned to its quantization axis.

Two neodymium magnet created the required quadrupole magnetic field. The magnets are attached (with a 3D printed holder) to a three axes NRT150/M 150mm motorized linear translation stage with a BDC103 three channel controller from Thorlabs. In addition, a Thorlabs motorized rotation state with a TST001 driver controls the angular movement of the magnet.

A laser power of about 250 $\mu$W, measured before the objective, yields fluorescence counts of about 200kcounts/s at zero magnetic bias fields.
S2. SYSTEM PARAMETERS

A. Pulsed ODMR measurements

A high resolution pulsed ODMR measurement was performed to identify potential coupling to adjacent $^{13}$C. Within the full width half maximum of the Lorentzian fit, no additional couplings to $^{13}$C are visible (cf. Fig. S1).

The average of both transition frequencies $\nu_1$ and $\nu_2$ should correspond to the zero-field splitting parameter $D = 2.87 \text{ GHz}$. In the present case the average yields $(\nu_1 + \nu_2)/2 = (2870.7005 \pm 0.008) \text{ GHz}$ indicating a well aligned magnetic field.

![Figure S1](image)

Figure S1. Pulsed ODMR measurement. (a) For the lower transition, a 3.3\,µs long $\pi$-pulse was applied to invert the population. A Lorentzian fit yields an FWHM of $(306 \pm 23)\,\text{kHz}$ at $\nu_1 = (1865.955 \pm 0.006)\,\text{MHz}$. (b) For the upper transition, a 2.7\,µs long $\pi$-pulse was applied, with an obtained FWHM of $(387 \pm 24)\,\text{kHz}$ at $\nu_2 = (3875.446 \pm 0.006)\,\text{MHz}$. The error bar in both plots corresponds to the normalized Poissonian error of the photon count signal.
B. Coherence times of the Rabi drives

The drive between the transitions, $\nu_1$ and $\nu_2$, has to be adjusted to ensure the same Rabi frequency, $\Omega$. The inset of Fig. S2 shows the dependency of the Rabi frequency with respect to the applied amplitude in the signal generator. It becomes apparent that the lower transition with $\nu_1$ requires a larger amplitude to perform equal to $\nu_2$. The reason for the imbalances can be manifold and are discussed in Sec. S3.

The largest coherence time at $\nu_1$ was achieved for a drive of $\Omega/2\pi = (1.366 \pm 0.006) \text{ MHz}$ with $T^2_{\Omega(\nu_1)} = (62 \pm 12) \mu \text{s}$. For the upper transition at $\nu_2$ a drive of $\Omega/2\pi = (2.816 \pm 0.006) \text{ MHz}$ yields $T^2_{\Omega(\nu_2)} = (159 \pm 24) \mu \text{s}$.

An even larger different can be seen in the coherence times for both transitions. In order to record the coherence times, a first measurement determined the correct Rabi frequency for a given amplitude. Then the coherence time was measured by deliberately undersampling the obtained Rabi frequency to reduce considerable the measurement time.

There are interesting characteristics which can be extracted from the coherence time measurement in Fig. S2. Each driving field, $\Omega_i$, posses a noise component $\delta \Omega_i$. The amplitude of the driving noise is dependent on the strength of the drive and its scaling with amplitude is dependent on the measurement configuration. The coherence time curves coincide for small Rabi frequencies, as the external magnetic noise, $\delta B$, constitutes to be the main contribution. By increasing the drive a larger dressed state energy gap can be created protecting the system from the magnetic noise, $\delta B$. At the same time drive fluctuations become more prominent and dominate for a certain drive value the noise contribution. The sweet spot in the coherence time curve indicates a balanced situation, where the dressed states of the drive experiences similar noise contributions from the drive and the environment. Increasing the drive further leads to a non linear increase of the drive noise and the coherence time begins to decrease.

The drastic difference in the appearance of the coherence time curves indicates a frequency dependent noise of the drive fields for $\nu_1$ and $\nu_2$. In contrast to this, the magnetic noise seems to have an equal impact on both transitions.

The main figure shows coherence times of Rabi oscillations, $T^2_{\Omega}$, as a function of Rabi drive, $\Omega$. The difference in $T^2_{\Omega}$ between both transitions originates mainly from a different drive noise characteristic at $\nu_1$ and $\nu_2$. The error bars are standard deviations, $\Delta T^2_{\Omega}$, from the fit $S(t) = A\cos(\Omega t) \exp(-t/T^2_{\Omega})$ to the decaying Rabi signal. Inset: measurement of the Rabi frequency for different amplitudes of the signal generator. A linear fit to the slope yields $m_{\nu_1} = (89.68 \pm 0.10) \text{ kHz/mV}$ and $m_{\nu_2} = (69.72 \pm 0.05) \text{ kHz/mV}$. The error bars correspond to $\Delta \Omega$.

Figure S2. Rabi measurements at two transitions, $\nu_1 = (1864.942 \pm 0.019) \text{ MHz}$ and at $\nu_2 = (3876.467 \pm 0.016) \text{ MHz}$. The main figure shows coherence times of Rabi oscillations, $T^2_{\Omega}$, as a function of Rabi drive, $\Omega$. The difference in $T^2_{\Omega}$ between both transitions originates mainly from a different drive noise characteristic at $\nu_1$ and $\nu_2$. The error bars are standard deviations, $\Delta T^2_{\Omega}$, from the fit $S(t) = A\cos(\Omega t) \exp(-t/T^2_{\Omega})$ to the decaying Rabi signal. Inset: measurement of the Rabi frequency for different amplitudes of the signal generator. A linear fit to the slope yields $m_{\nu_1} = (89.68 \pm 0.10) \text{ kHz/mV}$ and $m_{\nu_2} = (69.72 \pm 0.05) \text{ kHz/mV}$. The error bars correspond to $\Delta \Omega$. 

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... which will be presented in the following sections.

C. Ramsey measurements

A free induction decay or Ramsey measurement was performed on both transitions to identify that external and internal magnetic noise has the same impact on the $m_s = -1$ and $m_s = +1$ spin state. Fig. S3 illustrates that the average pure dephasing time, $T_2^* = (1.78 \pm 0.24) \mu$s, corresponds well with the individual measurement.

Figure S3. Ramsey coherence time measurement. The $\pi/2$ pulses in the scheme were deliberately detuned by 2MHz to determine the coherence time, $T_2^*$, in the decreasing oscillating signal more accurately. The curves were fitted to the function $\alpha \sin(\omega t + \phi) \exp\left\{-\left(t/T_2^*\right)^\beta\right\}$, where $\beta = 2.0 \pm 0.1$ was obtained for both curves. Hence, Gaussian uncorrelated noise can be assumed. (a) shows the lower transition and (b) the upper one, where both were measured at the ODMR frequency (+2MHz) given in Fig. S1. The error bar in both plots corresponds to the normalized Poissonian error of the photon count signal.
D. Hahn Echo measurements

A coherence time measurement with a standard Hahn Echo scheme was performed (cf. Fig. S4). Within the error bar, both transitions yield the same coherence time results. An averaged coherence time of $T_2 = (215 \pm 31)$ µs can be stated for the system, which coincides well with the individual measurement. Hence, the magnetic noise acting on the $m_s = -1$ and $m_s = +1$ can be considered as equal.

![Graph](image)

Figure S4. Hahn Echo coherence time measurement. The last pulse in the Hahn Echo measurement was either projecting the measurement to the bright ($\pi/2$-pulse, top curve) or to the dark ($3\pi/2$-pulse, lower curve) spin state of the NV center. Identifying the decoherence level to which both curves decaying to, increases the accuracy of the fit. The coherence time was obtained by fitting to $\propto \exp\left\{-\left(t/T_2\right)^2\right\}$ for (a) the lower and (b) the upper transition, corresponding to the ODMR measurement in Fig. S1. The error bar in both plots represent the normalized Poissonian error of the photon count signal.

S3. ESTIMATED NOISE CHARACTERISTICS

In this section, we try to identify the components contributing to drive noise of the fields. From the Ramsey and the Hahn Echo measurements we can assume the same external magnetic noise at both transition frequencies, $\nu_1$ and $\nu_2$. In continuous dynamical decoupling the NV is permanently subjected to strong drive fields. Therefore, drive noise will be the strongest noise contribution in the scheme. As we did not had the possibility to measure the fast noise components directly with a fast oscilloscope, we would like to state the various noise contributions in our system, based on conservative estimates.

The drivings field in this work are created by the Tektronix AWG70002A. The maximal output amplitude for this device is 0.5Vpp (or 1Vpp by including the inverted channel) with a DAC accuracy of 8bit, yielding a maximum precision of 0.5Vpp/2^8 = 2mVpp or 1mV for the amplitude of an oscillating signal. Consequently, a sinusoidal field with amplitude of 0.25V (0.5Vpp) has an accuracy of (250 ± 1) mV, which gives a relative error of 0.78%. The vendor specifies the amplitude accuracy to ±10 mVpp [5], which increases the relative error to 4%, indicating that besides the DAC inaccuracy, other noise sources exist. Therefore, the application of 4 sinusoidal fields at different frequencies can have at maximum 1/4 of the total amplitude noise, if it is equally distributed on all of them (which is not necessarily the case). Thus, one frequency component is accurate up to (62.5 ± 1.3) mV, yielding an relative amplitude error of 2%.

A field of 62.5 mV (≈ 125 mVpp) would create (see the inset of Fig.1 in the main text) a Rabi drive of the order of 10 MHz. Increasing the drive field above that value caused heating problems, as a lot of power is dissipated in the microwave structure causing a drastic decrease in ODMR contrast and fluorescence signal. To prevent that from
happening the amplitude has to be lowered so that such effects are not dominating. We found a balance between the thermal effects and the maximal obtainable Rabi frequency for fields of the order of 40 mVpp, if 4 fields are used simultaneously (which result in a total amplitude of 160 mVpp). Given these limitation in the drive fields, the relative noise for each frequency components is on average 6.25%.

Moreover, the amplitude noise of the AWG is correlated to its base clock, which introduces correlated (amplitude) noise on µs timescale and may very well be responsible for frequency dependent (fast) noise imbalances, translated to the dressed states of the considered three-level system (see main text Fig. 4). The origin of this noise character can be grasp by investigating how data become sampled in an AWG.

Modern, very fast AWGs (with sample rate exceeding 1GS/s) do not process each sample directly as the rate the data is read exceeds the speed of common semiconductor memories [6]. To circumvent the problem, data is processed in parallel and then multiplexed inside the high-speed digital logic in the AWG that drives the DAC. However, the multiplexing operation is directly bound to the internal clock of the AWG, which is usually of the order of 10MHz and thus, creates correlation on that time base. Any errors in the timing of the internal clock edge translate directly into timing errors in the analog output, which reflects to phase noise and is immediately detectable by amplitude fluctuations. This is a noise source, which adds to the DAC inaccuracy and creates (among other effects) the total noise floor of the device. Eventually, a high-speed oscilloscope would be the preferential choice to identify fast noise on the frequency components.

On top of this, we assume also a frequency dependent noise, since the output of the amplifier is not flat over a range of $\nu_2 - \nu_1 \approx 2$ GHz. Moreover, the microwave structure experiences frequency dependent attenuation of the applied drive fields leading (among other contributions) to different Rabi frequencies for $\nu_1$ and $\nu_2$ at the same amplitude output (cf. the inset of Fig.1 in the main text).

All these various noise contributions have a strong impact on the drive of the NV system as they cause the dressed states to fluctuate. Therefore it becomes a crucial task in CCD to reduce the impact of noise, introduced by multi-frequency drive fields, by creating robust states, which suffer less from drive noise contributions. By demonstrating robustness to all these noise sources underlines the importance of the present work.

6.4 Appendix

The present section summarizes the initial steps and ideas of how the experiment in this chapter started. The main goal was the experimental realization of the theoretical proposal for a fully robust qubit in Ref. [118]. It turned out that the implementation would require much stronger off-resonant drive fields, which could not be realized in the present configuration. However, upon the investigations new phenomena have been discovered, which led to the results presented in Sec. 6.2.

Hence, the following section can be seen as a motivation and gives a pictorial view of the situation. For more detailed calculations and derivations, it should be referred to Ref. [118].

6.4.1 Schematics of detuned double lambda drive

Power fluctuations in the drive fields constitute to be the major noise source in continuous dynamical decoupling protocols. The concatenation of drives is one idea how to decrease the impact of drive noise on the resulting state (cf. Chapter 5). The on-resonant drives in this approach, however, does not allow fast gate operations between the created state as their rate, $\Omega_g$, is set by the strength of the drive fields. Every consecutive concatenation reduces the rate by a factor of 2.

An alternative approach is the application of off-resonant fields as depicted in Fig. 6.1. This scheme allows to obtain a fully robust qubit by utilizing a three-level system and four driving fields. In addition, it offers an alternative approach to perform the task of high frequency sensing and predicts much faster gate operations, limited solely by the utilized detuning, $\Omega_g/2\pi \ll \Delta$. It can also be regarded as a continuing effort to improve further the ideas described in Chapter 5 by pursuing a slightly different approach.

The protocol in Fig. 6.1a requires four drive fields in order to address the three level states in a detuned way:

$$B_{\text{tot}}(t) = B_1 \cos \left(2\pi (\nu_1 + \Delta_1)t\right) + B_2 \cos \left(2\pi (\nu_2 + \Delta_1)t - \pi\right) + B_3 \cos \left(2\pi (\nu_1 + \Delta_2)t\right) + B_4 \cos \left(2\pi (\nu_2 + \Delta_2)t\right).$$ (6.1)

The drives represent two inverted lambda configurations. It appears that $B_1 = B_3$ and $B_2 = B_4$, as they drive almost the same transitions. ($\nu_1, \nu_2 \gg \Delta_1, \Delta_2$).

However, the control fields, $B_1$ and $B_2$, have significantly different transition frequencies, $\nu_1$ and $\nu_2$, respectively, and the same field amplitude, $B_i$, does not lead to the same Rabi drive, $\Omega$, for $\nu_1$ and $\nu_2$. The reasons for this behavior are diverse. For instance, the amplifier has not a flat response over the whole frequency range and amplifies some frequency components stronger than other. Moreover, the microwave structure is
6 Concatenated continuous dynamical decoupling of a three-level system

Fig. 6.1: Schematics of the detuned double lambda drive. (a) In the bare state basis $\nu_1$ and $\nu_2$ denote the energy difference between the states. Four drive fields are applied and adjusted to have the same Rabi frequency, $\Omega$. The (inverted) lambda drive detuned by $\Delta_1$ ($\Delta_2$) creates the dressed state $|B\rangle$ ($|D\rangle$). (b) The dressed states, $|B\rangle$ and $|D\rangle$, in the interaction picture with an energy separation of $\Omega$. The population transfer happens here at a rate $\Omega_{\text{SL}}$, which is by the factor $\sqrt{2}$ faster.

not designed to be impedance matched, therefore frequency dependent attenuations or resonances might occur.

In general, it has to be ensured, that the on-resonant drives on $\nu_1$ and $\nu_2$ yield the same Rabi frequency, $\Omega$. Hence, the field amplitudes, $B_1$ and $B_2$, have to be adjusted to fulfill this requirement and the total Rabi drive (with $\gamma_{\text{NV}} B_i = \Omega$) becomes

$$\Omega_{\text{tot}}(t) = \Omega \cdot \left( \cos \left( 2\pi (\nu_1 + \Delta_1) t \right) - \cos \left( 2\pi (\nu_2 + \Delta_1) t \right) + \cos \left( 2\pi (\nu_1 + \Delta_2) t \right) + \cos \left( 2\pi (\nu_2 + \Delta_2) t \right) \right).$$

Driving the three states $\{-1, 0, +1\}$ in the denoted configuration creates dressed states $\{|B\rangle, |0\rangle, |D\rangle\}$, as illustrated in Fig. 6.1b. It has to be noted, that the dark state drive $|0\rangle \leftrightarrow |D\rangle$ can only be realized by off-resonant fields. Setting $\Delta_1 = \Delta_2 = 0$ would create destructive interference, where a drive of a two-level system with $2\Omega$ would remain.

The population transfer in the dressed state basis happens at a rate, $\Omega_{\text{SL}}^{\Delta_i}$, which denotes the Rabi frequency in the single lambda (SL) drive, with a detuning $\Delta_i$. By adjusting the values for the detunings, $\Delta_1$ and $\Delta_2$, a protected qubit can be realized.
Fig. 6.2a illustrates how the protected qubit appears. Under an off-resonant drive of $\Omega_{\Delta,\text{SL}}^\Delta$, doubly dressed states are created due to AC-Stark shift of the dressed states. Their energy separation is $\propto (\Omega/2\pi)^2/\Delta_i$, which is an approximation of $\Delta_i - \sqrt{\left(\Omega/2\pi\right)^2 + \Delta_i^2}$. Therefore, a drive $\Omega_{\Delta,\text{SL}}^\Delta$ with a detuning, $|\Delta_1| > |\Delta_2|$, results in a smaller energy gap in the doubly dressed states. By choosing a detuning with $\Delta_2 = -\Delta_1/2$ two eigenstates, $|B\rangle$ and $|0\rangle$, will have identical energies, $\Omega_r$. Hence, fluctuations in the drive fields will affect both states at the same time and alter them always in the same direction, by maintaining the robust qubit.

A crucial requirement for the creation of the robust qubit in this scheme is $\gamma_{\text{NV}} B_z \gg \Delta \gg \Omega_r/2\pi$, where $B_z$ represents the static magnetic field, separating the NV electron spin states to obtain the transition frequencies, $\nu_1$ and $\nu_2$.

To quantify the robust qubit, its energy gap and coherence time has to be investigated in order to show the coherence time prolonging effect of the scheme. This can be done by means of a Ramsey measurement, performed on the dressed states (cf. Sec. 2.5.3). A superposition state $|\psi\rangle = (|B\rangle + |0\rangle)/\sqrt{2}$ will be affected by the energy gap of the robust qubit and the coherences of the superposition state will oscillate with $\Omega_{\text{RQ}}$. Recording

$$\Delta_1 = 0 \quad \Delta_1, \Delta_2 \neq 0 \quad \Delta_1 = 0$$

$$\frac{\pi}{2\sqrt{2}} \quad \Omega_{\Delta_1} \quad \frac{\pi}{2\sqrt{2}}$$

$$\frac{\pi}{2\sqrt{2}} \quad \Omega_{\Delta_1} \quad \frac{\pi}{2\sqrt{2}}$$

$$\Omega_{\Delta_1} \quad \Omega_{\Delta_1} \quad \Omega_{\Delta_1}$$

$$\Omega_{\Delta_1} \quad \Omega_{\Delta_1}$$

$$\Omega_{\Delta_1}$$

The number $N$ indicates that this sequence have to be repeated many times.
the persistence of the oscillations yields the coherence time of the robust qubit.

The pulse sequence for the coherence time measurement is depicted in Fig. 6.2b, displayed in the bare state basis (cf. Fig. 6.1a). The resonant fields couples the $|0\rangle$ to the $|B\rangle$ state and by applying a $\pi/2$-pulse with $\Omega_{SL}$, the superposition state can be created. On the superposition state the drive in eq. (6.2) is applied, where the duration $\tau$ is varied. After the drive the coherences are transformed to populations in a consecutive $\pi/2$-pulse with $\Omega_{SL}$ and are read out by a laser. The results are summarized in the next section.

6.4.2 Results

The contribution of the intrinsic nitrogen nuclear spin of the NV center to the scheme was not completely clear. To eliminate this uncertainty the measurements were performed in the vicinity of the excited state level anti-crossing (ESLAC) [84], where the nitrogen spin becomes polarized.

A very condensed version of the findings is shown in the measurement matrices of Fig. 6.3, displaying the energy gap and the coherence times. From the theoretical considerations, an improvement is expected at about $\Delta_2 = -\Delta_1/2$ (see [118]). The theory indicates also that the moderate energy separation by $\nu_1$ and $\nu_2$ of the bare states near the ESLAC might change the relation between $\Delta_1$ and $\Delta_2$. Thus, for every pair of detunings

Fig. 6.3: Energy gap (a) and coherence time (b) measurements of the robust detuned qubit with respect to a variation of $\{\Delta_1, \Delta_{rel}\}$. $\Delta_{rel}$ changes the value of $\Delta_2$ according to $\Delta_2 = -\Delta_1/2 + \Delta_{rel}$. A moderate Rabi frequency of $\Omega/2\pi = (2.72 \pm 0.01)$ MHz was used for all four drive fields. The green dashed line indicates the scenario, where $|D\rangle \leftrightarrow |0\rangle$ are driven on resonance ($\Delta_2$ becomes zero). The combination of an on-resonant drive, and an off-resonant drive generates a large enough energy gap, in order to protect the robust qubit from magnetic noise.
\{\Delta_1, \Delta_{\text{rel}}\}$ the coherence time and the oscillations (introduced by the energy gap) are recorded for an increasing value of $\tau$. With the introduced relative detuning, $\Delta_{\text{rel}}$, the region around the optimal detuning could be mapped out by $\Delta_2 = -\Delta_1/2 + \Delta_{\text{rel}}$.

The measurement of the energy gap in Fig. 6.3a show an expected behavior. For very large detunings the energy gap of the robust qubit decreases. If one of the detunings approaches zero, then the energy gap increases again as one of the drives, $|0\rangle \rightleftharpoons |B\rangle$, or $|0\rangle \rightleftharpoons |D\rangle$, become resonant. Only the case for $\Delta_1 = \Delta_2 = \Delta_{\text{rel}} = 0$ was not measured correctly as the obtained energy gap frequency becomes undersampled. Anyway, this point is not of interest, since it represents solely a drive of the transition $\nu_1$ with the Rabi frequency $2\Omega$.

It becomes apparent, that the energy gap of the resonant bright state drive (for $\Delta_1=0$) is larger in comparison to the case when the dark state (for $\Delta_2=0$) is on resonance. The resonant dark state drive is indicated by a green dashed line in the matrix plots of Fig. 6.3.

An interesting scenario shows the measurements of the coherence times in Fig. 6.3b. No improvement in coherence time at all can be identified for the case $\Delta_2 = -\Delta_1/2$, which corresponds to the horizontal line at $\Delta_{\text{rel}} = 0$. The coherence times become larger if the resonant energy gap is approached. However, the coherence times for the resonant dark state drive ($\Delta_2=0$) and the off-resonant bright state drive shows a considerable improvement in coherence times. These findings were the motivation for further investigations and lead to the measurements presented in Sec. 6.2.

The reason, why this scheme did not improve the coherence time as anticipated in the theory might be connected to the small energy gap of the robust state. Off-resonant drives require a much larger field amplitude to create the same energy gaps as resonant drives. As mentioned before, higher field amplitudes introduced strong heating effects and prevents a further increase of the drive frequency $\Omega$.

In addition, free induction decay measurements for the present NV yield a coherence time of about $T_2^* \approx 2\mu s$ (cf. Sec. 6.3). Hence, if an improvement in this configuration should be realized and the drive fields cannot be increase any further, then a NV with a longer $T_2^*$ time (of the order of $10\mu s$) would be needed. This would ensure a smaller magnetic field noise component and the small energy gap, created by this sequence, could be sufficient to decouple the robust qubit from the environmental noise.
Conclusion and Outlook

*Thoughts without content are empty, intuitions without concepts are blind.*

— Immanuel Kant

*Critique of pure reason (1781)*
This thesis covers and combines three projects, which are all devoted to the task of developing new measurement protocols for quantum magnetometry. At the heart of this approach stands the aim to increase the capabilities of the nitrogen-vacancy (NV) center in diamond by employing and protecting its quantum character to improve its sensitivity for the task of magnetic field sensing.

In order to achieve these goals a sophisticated control and manipulation of the NV center is required, which eventually demands for the implementation of complex protocols. This has led to the development of a very powerful software suite, termed Qudi, and constitutes the first project within this thesis.

The project started with a small group and evolved to a collaborative work between both institutes, University of Ulm and the Technical University of Denmark. This software suite has not only facilitated the control and implementation of microwave protocols for the present work, but has also created an organized way to transfer the physical idea to the experiment by maintaining flexibility and transparency in the development process. Meanwhile, the abilities of the software framework have grown significantly and the open-source character has invited many groups to participate in the development process and establish on this basis further collaborative interactions. As the concept of the suite is not limited to the manipulation of color centers in diamond, ongoing development processes expand already the applicability of Qudi in different research fields (like quantum cryptography or quantum communication). Hence, this software framework exhibits a broad scope of possibilities for any laboratory environment and offers industrial applications.

The task of high-frequency sensing in the GHz regime was reserved to relaxometry measurements possessing a sensitivity proportional to the pure dephasing time, $T^*_2$, of the sensor. With this work the capability of the NV center was extended towards high-frequency fields by overcoming the $T^*_2$ bandwidth limitation. Furthermore, it was demonstrated, that continuous dynamical decoupling could be integrated into the sensing task and thereby prolonging the coherence time of the sensor to its transverse relaxation time, $T_2$, while being susceptible to high frequency fields. It was possible to expand the coherence time of the sensor towards its lifetime limit and to show the measurement of a smallest detectable magnetic field strength of 4 nT at 1.6 GHz. Moreover, the narrow-bandwidth character of this scheme ensures a very selective frequency probing. Thus, with this approach the NV center has the potential to be used as a high-sensitive, narrow-bandwidth, high-frequency sensor, which paves the way specifically for radar applications.

Although the developed protocol exhibits great sensing performance, it still faces limitations as the small protected energy gap results in slow qubit operations, which restricts its application in quantum information science. In addition, the necessity for many concatenated microwave fields might set an impediment to the applicability of the protocol, as further concatenation of controls fields are required to increase the sensitivity of the sensor.
The third project tried eventually to circumvent the posed limitations by a different approach, while still maintaining the possibility for high-frequency sensing. By utilizing the three-level character of the NV center under a combination of on-resonant and off-resonant drives, a fully robust qubit system could be realized. The on-resonant fields are responsible for the large energy gap, protecting the quantum state from environmental noise, whereas the off-resonant fields produce AC-Stark shifted energy levels and decouple the sensor thereby from noise of the drive fields. This combination results in doubly-dressed states, whose robustness to environmental noise can be directly controlled solely by the strength of the applied microwave field, while maintaining a protection to drive noise.

The theoretical model, developed in this approach, predicated an improvement of the coherence time by at least 2 orders of magnitude. An implementation of the scheme demonstrated its performance and showed that for a correct choice of the detuning for the off-resonant drive, robustness to drive noise could be achieved. It is expected that further increase of the microwave drive would lead to a significant enhancement of the coherence time of the sensor.

The potential susceptibility of the protocol to high frequency sensing tasks, its simplified experimental realization and the predicted fast qubit operations extend further the capabilities of the NV sensor. This opens eventually the doors for applications not only in quantum sensing, but also quantum information science and quantum technology.
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