Study of Nitrogen-Vacancy Centers in Diamond

Thesis submitted in partial fulfilment of the requirement for the degree of
“Doctor of Philosophy”

by

Yechezkel Schlussel

Submitted to the Senate of Ben-Gurion University of the Negev

July 2017

Beer-Sheva
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Approved by the advisor: Prof. Ron Folman
Approved by the Dean of the Kreitman School of Advanced Graduate Studies: Prof. Michal Shapira

July 2017

Beer-Sheva
This work was carried out under the supervision of Prof. Ron Folman
In the Department of Physics
Faculty of Natural Sciences
Research-Student’s Affidavit when Submitting the Doctoral Thesis for Judgment

I, Yechezkel Schlussel, whose signature appears below, hereby declare that:

I have written this Thesis by myself, except for the help and guidance offered by my Thesis Advisors.

The scientific materials included in this Thesis are products of my own research, culled from the period during which I was a research student.

This Thesis incorporates research materials produced in cooperation with others, excluding the technical help commonly received during experimental work. Therefore, I am attaching another affidavit stating the contributions made by myself and the other participants in this research, which has been approved by them and submitted with their approval.

Date: 5/7/17  Student’s name: Yechezkel Schlussel  Signature:______________
Affidavit stating the contributions made by myself and by other participants in this research

I, Yechezkel Schlussel, whose signature appears below, hereby declare that the scientific materials included in this Thesis are products of my own research, except for the following parts, which were produced in cooperation with others: the Doppler-free like spectroscopy of the NV center (chapter 4) as well as theoretical calculations for the conditions required for NVs to behave adiabatically (chapter 5) were developed in cooperation with Yossi Rosenzweig; The characterization of thin-film high-$T_c$ superconductors (chapter 7) and parts of section 6.3 were developed in cooperation with Dr. Amir Waxman.

Date: 5/7/2017 Name: Yechezkel Schlussel Signature: 

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Date: 5/7/2017 Name: Amir Waxman Signature: 


ABSTRACT

The Nitrogen-Vacancy (NV) center in diamond is an atom-like system (defect) inside a carbon solid lattice, with the possibility of sharp spectroscopy lines, relatively long coherence times, and optical pumping, even at room temperature. This opens possibilities for utilizing this system for fundamental studies as well as technological applications. In this work we experimentally and theoretically study the physical properties of the NV system, and also utilize it as a magnetic probe in order to characterize high-$T_c$ superconductors, an important field in its own right.

Specifically, we study the physical properties of the NV center by:

2. Analyzing the spectroscopic broadening mechanisms related to the local environment of the NV in the lattice via novel two-frequency Doppler-like spectroscopy.
3. Analyzing the adiabatic criterion for the NV center under rapidly changing magnetic fields.

In the second half of the thesis we describe utilizing the NV as a magnetic probe:

1. We present a novel method for probing magnetic fields around zero field, a situation in which the high natural strain of a diamond hinders effective probing.
2. We describe a first characterization of a superconductor through its magnetic field. We observe the Meissner state as well as vortex pinning.
3. We describe spatial imaging (CCD) enabling maps of the magnetic field and comparing the supercurrent to an updated theory of superconductivity.
Publishable work:

Only one paper was published so far (PRB, 2014, probing superconductivity), and Y. Schlus-sel is the second author.

However, we believe that several of the topics described above are novel and warrant a publi-cation, and this will be done in the near future. Here is a list of the publications we aim to submit:

1. Excitation of NVs with polarized microwave radiation (including a difference of opinion with a previously published paper).
2. Doppler-like spectroscopy of the NV center (including a difference of opinion with a previously published paper).
3. Adiabaticity in the NV center.
4. Utilizing the NV center as a magnetic probe of near-zero fields.
5. Mapping the magnetic fields of a superconductor.
ACKNOWLEDGMENTS

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5.2 A map of the probability of finding the system in its original eigenstate $m_s = +1$ (in the field frame) after the magnetic field is changed from $B_z$ to $-B_z$. In this figure we consider the situation at time $T/2$ for the case where $B_z = B_0 \cos(2\pi ft), B_x = 0, B_y = 0$. The map was generated using a similar calculation to the one depicted in Fig. 5.1b. Note that in the following experiments we utilize the $m_s = -1$ state, but this does not alter the results. In order to generate this map we utilized the fact that D (the spin-spin interaction) is much larger than the other elements and hence the $m_s = 0$ level is well separated from the $m_s = \pm 1$ levels. Using the rotating-wave approximation we reduced our Hamiltonian into an effective 2D Hamiltonian. A detailed explanation of this approximation is given in Appendix A.
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5.4 (a) The chip for the adiabaticity experiment. The silicon wafer, with 2 \(\mu m\) thick gold wires, is mounted on a copper base. The chip pads (visible) are connected to an ac current supply. The two crossing wires connected to the pads are just 10 \(\mu m\) wide and therefore hardly noticeable. (b) A zoom-in on the crossing wires. A \{110\} diamond is placed above the cross wires. The chip right edge is attached to a white ABS structure (visible is (a) and cutting the figure here) which is used for aligning the [-11-1] orientation parallel to one wire and perpendicular to the other wire. By controlling the currents in the wires we can manipulate the effective magnetic field in 1D along \(z\) or to rotate it in the \(x-z\) plane, where the coordinate system is that of the [-11-1] NV orientations (\(z\) being the NV axis parallel to the [-11-1] direction).
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5.8 (a) Experimental sequence for the linear case in which $B_y = 0$ and $B_z = B_{z0} \cos \omega t$. (b) In blue, the normalized fluorescence after a single $\pi$ pulse (dark state reference). The $\pi$ pulse was applied before applying changes to $B_z$. In this sequence $B_z = B_{z0} \cos \omega t$ where $\omega = 2\pi f$ and $f = 150$ kHz. In red, the normalized fluorescence after a $2\pi$ pulse (bright state reference), again before the magnetic field was changed. In black (our signal), the normalized fluorescence measured for the case in which one $\pi$ pulse is applied before changing the magnetic field and one $\pi$ pulse is applied after changing the magnetic field. As expected from Fig. 5.2 the level of adiabaticity goes down as the magnetic field becomes stronger. Explanation of how the normalized fluorescence is measured is given in Fig. 5.6b. The time separation between the two $\pi$ pulses is $20 \mu s$. 

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5.12 Calculation of the time for which the magnetic field $\vec{B}$ is oriented at an angle less than 30° from the y axis as a function of $B_{y0}$ for the case in which $B_y = B_{y0} \sin \omega t$ and $B_z = B_{z0}$. The four curves are calculated for different frequencies where $\omega = 2 \pi f$. It is easy to see that the higher the frequency, the shorter the time for which the total magnetic field is oriented by less than 30° from the y axis. On each curve we mark the value of $B_{y0}$ for which the system shows a transition from adiabatic to non-adiabatic behavior. The markers are taken from Fig. 5.9 by taking the amplitude $B_{y0}$ for which the blue curve and the red curve join each other.
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By orienting the MW wire antenna perpendicular to the [-11-1] lattice vector
the emitted magnetic field will be aligned along the [-11-1] NV orientation.
Since the MW field will have no components in the plane perpendicular to
the [-11-1] orientation it will not interact with the NVs g.s. levels. (d) The
proposed setup. The laser beam hits the diamond along the [-110] lattice
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ABBREVIATIONS

NV  nitrogen vacancy
ODMR  optically detected magnetic resonance
g.s.  ground state
MW  microwave
HPHT  high-pressure high-temperature
CVD  chemical vapor deposition
ER  electrons radiation
IR  irreducible representation
HF  hyperfine
CW  continuous wave
ABS  acrylonitrile butadiene styrene
SC  superconductor or superconducting
SQUID  superconducting quantum interference device
YBCO  YBa$_2$Cu$_3$O$_7$
FM  frequency modulation
ZFC  zero field cooling
SPCM  single photon counter
SNR  single-to-noise ratio
EMCCD  electron-multiplier CCD
1. INTRODUCTION

The negatively charged nitrogen vacancy (NV) center in diamond is formed by a missing carbon adjacent to a nitrogen ion impurity in the diamond lattice [1, 2]. The spatial electronic distribution of the NV center can be represented as a single wavefunction with discreet energy levels [3, 4]; these are responsible for the atom-like properties of the NV center. In recent years NV centers have generated immense scientific and technological interest. The simple protocols for manipulating and reading the system [5] together with its room temperature long coherence times [6, 7] make the NV center a leading candidate for several applications. The potential relevance for applications such as quantum communication [8], quantum computing [9, 10], and quantum memory [11, 12] has been demonstrated. The potential relevance as a sensor was also demonstrated, e.g., as a biomarker [13, 14], electric field probe [15], and for high-spatial-resolution magnetic imaging [13, 16, 17, 18, 19, 20, 21].

The origin of the NV atom–like behavior is the high level of isolation from the crystal electronic wavefunction (responsible for the diamond band structure). Large band-gap materials can contain many defects with ground-state energy levels that are far from the conduction band minimum. This energy gap is responsible for these defects having a ground state that is stable at high temperatures. However, the fact that the defect ground state is inside the energy gap is not enough for having atom-like features. To have optically-accessible electronic transitions, the defect excited state should also be inside the energy gap. The diamond band gap is 5.5 eV, and the NV ground state (g.s.) is located 2.6 eV under the diamond conduction band while the NV’s optically excited states are 1.95 eV above the NVs g.s. levels [22]. These unique features of a highly localized quantum system with atom-like properties, such as narrow energy levels with sharp spectroscopic lines, make this system interesting from both scientific [2] and technological aspects [23].

In order to define a physical system as a good quantum two–level system several basic
requirements should be fulfilled. The first requirement is the ability to pump the system to a defined quantum state. The second is the ability to manipulate the system. The third is a long coherence time and the fourth is a readout protocol. In the case of the NV center all these requirements are fulfilled. It is possible to initialize the NV g.s. level with a short green light pulse. The NV can be transferred to a different energy level using MW radiation, and the NV quantum state can be read by exciting the NV and measuring its fluorescence [5]. In addition, the room temperature coherence time of a single NV is $T_2^* = 2.7\mu s$ while $T_2$ of a single NV is $\geq 1.6\, ms$ [7]. The $T_2$ coherence time of an NV ensemble ranges from several $\mu s$ at room temperature to hundreds of milliseconds at liquid nitrogen temperature [6, 24]. A long coherence time is crucial for applications such as quantum computation and quantum memory as well as for high-precision measurements. In quantum computation the coherence time limits the number of actions that can be done on the quantum system. In high-precision measurements the sensitivity is inversely proportional to the square root of the coherence time [25].

Though studied for more than 50 years, NV centers in diamonds became the subject of intensive research only quite recently. A critical point in the evolution of this field was reached when the initialization, manipulation and readout of a single NV center was demonstrated [26]. Following the extensive study of NV centers it became clear that for some applications there is an advantage in using NV ensembles over the use of single NVs [23]. Studying ensembles gives rise to interesting new physical and technological questions. In the first part of this thesis we will focus on the physics and behavior of an NV ensemble. The second part of this thesis will be dedicated to NV-based magnetometry.

1.1 Thesis outline

This thesis is constructed as follows: In chapter 2 we briefly describe basic theoretical and physical aspects of the NV center. First we describe the formation of the center and the special photoluminescence behavior of the center. In the second part of this chapter we describe the Hamiltonian of the system and its selection rules. In order to explain the notations of the energy levels and the selection rules we give a short introduction to group theory. Here we also present a contrasting point of view we have developed concerning some recently
published results [27]. In chapter 3 we describe the Optical Detected Magnetic Resonance (ODMR) protocol. The ODMR protocol is the basic method used to retrieve the spectrum of the NV g.s. levels. We detail how this protocol is used for magnetometry. In chapter 4 we present a novel protocol to better understand inhomogeneous broadening and to improve the NV ensemble-based magnetometer. In addition, we present an extended and more detailed analysis of recently published results [28]. In chapter 5 we present a theoretical and experimental study of the g.s. spin behavior in an alternating magnetic field. This examines the conditions for adiabaticity in this system. In addition to the fundamental importance, we also explain its applicability. Several protocols have been suggested in order to optimize the magnetic [29], spatial [30, 31] and temporal [32] sensitivity of an NV-based magnetometer. When dealing with an NV ensemble additional effort is required in order to optimize the NV concentration [33, 34]. In chapter 6 we discuss how strain limits the magnetic sensitivity of the NV center. We then present methods we investigated in order to overcome this limitation. The optical nature of the NV center readout protocol together with the NV magnetic sensitivity make it an optimal magnetometer for studying superconductivity [35, 36, 37]. In chapter 7 we present the investigation of the SC phase transition using an NV sensor. In chapter 8 we present magnetic maps imaged with our 2D NV-based magnetometer and study current distribution in a meander-shaped superconducting wire. We summarize this thesis in chapter 9.

Finally, we end this thesis outline by providing a table of the diamond samples we used in this work. The names noted here will be used as acronyms throughout the thesis.
<table>
<thead>
<tr>
<th>Name in thesis</th>
<th>Type and orientation</th>
<th>Dimensions</th>
<th>NV layer thickness</th>
<th>distance to surface</th>
<th>NV concentration</th>
<th>Implantation</th>
<th>source</th>
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Table 1.1: A list of the samples that were used in this thesis.
2. THEORY

2.1 NV physical structure

The negatively charged NV center is formed by a missing carbon adjacent to a nitrogen ion impurity in the diamond lattice (Fig. 2.1). The physical structure of the NV center and its symmetries may be analyzed by understanding two aspects. The first aspect is the nature of the NV electronic states and its allowed transitions. The NV selection rules are a direct result of its $C_{3v}$ symmetry. This aspect will be discussed in section 2.5. The second aspect which helps in understanding the system is that of transformations from the lab frame coordinate system to the NV coordinate system. At low magnetic fields the quantum axis of the NV center is oriented along the virtual line that connects the nitrogen ion and the vacancy. From symmetry considerations a magnetic field along the quantum axis will split two degenerate energy levels while a magnetic field in the xy plane will be responsible for mixing the electronic states. The NVs themselves may appear in 4 different orientations [111],[1-1-1],[-11-1], and [-1-11] (Fig. 2.1). The angle between any two NV axes is 109.5°.

![Fig. 2.1: The four possible orientations of NV centers in the diamond lattice. The gray spheres represent carbon atoms, the blue ones are for nitrogen atoms and the red spheres denote vacancies. Each orientation is defined by the virtual line that connects the nitrogen ion and the vacancy.](image)
The diamond crystal structure is typically cut along three different planes perpendicular to the \{110\}, \{111\} or \{100\} lattice vector (Fig 2.2). The linear transformation from the lab frame coordinate system to the diamond coordinate system should be taken into account when choosing the preferred orientation or orientations for manipulating the NV center or utilizing it as a sensor.

Fig. 2.2: (a-c) The three possible crystallographic planes. (d-f) A view of the crystal and the different NV orientations when the crystallographic plane is in the image plane.

2.2 NV formation

The formation of NV centers requires the presence of nitrogen ions and vacancies in the lattice. Synthetic diamonds are fabricated either by chemical-vapor deposition (CVD) or by high-pressure high-temperature (HPHT) synthesis. These synthetic diamonds have nitrogen impurities. The classification of diamonds according to their nitrogen impurity is as follows: Type I has a nitrogen density of \( n > 5 \) ppm (typically 100 to 3000 ppm). Type Ia contains aggregated nitrogen and hence is usually not used for the fabrication of NV centers. Type Ib contains single substitutional nitrogen. Type II is defined as diamonds with very low nitrogen concentrations, \( n < 5 \) ppm, where type IIa means the major impurity in the diamond is
nitrogen and the diamond is an insulator, while in type IIb diamonds the major impurity is boron. This classification holds for both natural and synthetic diamonds. Typically most of the HPHT diamonds are classified as type Ib and most of the CVD diamonds are classified as type IIa. Another way to classify synthetic diamonds is according to their main use. It is worth mentioning the Electronic Grade (EL-grade) samples. The EL-grade is a high purity CVD-grown diamond originally designed for active detector electronics. The EL-grade contains less than 5 ppb (often below 1 ppb) nitrogen concentration and typically has an NV concentration less than 0.03 ppb. One way to create vacancies in the diamond is by irradiating the diamond with electrons. This method is good when the initial concentration of nitrogen is high enough for creating the desired concentration of NV centers [33]. Usually electron irradiation is used to generate samples with a high density of NVs from HPHT diamonds, although this method may also be used for enhancement of single NV sites. Another way to generate vacancies in the lattice is by bombarding the sample with ions. When the sample doesn’t contain the needed concentration of nitrogen or alternatively when \(^{15}N\)-based NV centers are desired, the sample can be bombarded with nitrogen ions \((^{14}N \text{ or } ^{15}N)\); this method is usually called implantation. For NV-based magnetometry with high spatial resolution it is often favorable to have a thin layer of NV centers. The penetration of nitrogen ions in diamond as a function of the ion energy calculated by the SRIM program is depicted in Fig. 2.3. The diamonds DDK111, DDK100A, DDK100B, and E6110 were implanted with \(^{14}N\) at 10 keV, 35 keV, and 60 keV (all three for each diamond) to generate a 100 nm-thick layer of NVs beginning 20 nm from the diamond surface. A detailed study of different ways to radiate NVs may be found in [33]. The next essential step in the NV formation is the annealing of the sample. During the annealing the nitrogen impurities move towards the vacancies and generate the NV centers’ chemical bonding. In our diamonds the annealing was done in an inert atmosphere at 850°C for three hours.

2.3 Franck-Condon effect

The NVs exhibit a fluorescence spectrum which is significantly different from its absorption spectrum. This difference is explained according to the Franck-Condon principle. The Franck-Condon principle states that electronic transitions occur without changes of the nuclei
Fig. 2.3: (a) The distance $^{14}$N ions penetrate into the diamond as a function of their energies. (b) The longitudinal straggling of the $^{14}$N as a function of their energy. The longitudinal straggling is in principle the standard deviation of the ion distribution. Our diamonds were irradiated with 10 keV, 35 keV, and 60 keV $^{14}$N in order to generate a thin layer of NVs near the diamond surface. At these energies the ions penetrate on average to depths of 22 nm, 70 nm, and 114 nm with a longitudinal straggling of 8 nm, 18 nm, and 25 nm.

position because the timescales of the two processes are significantly different. The energy minima for ground and excited state are not at the same nuclear configuration but are shifted by $\delta Q$ in the normal coordinate space. The Franck-Condon principle states that the most likely transitions are at the same nuclear configuration, i.e. the vertical ones in Fig. 2.5, the other transitions are also allowed as the overlap of the vibrational wave functions is non-zero and the probability is given by the Franck-Condon factors. Due to the fact that in the vertical transition the g.s. overlaps a vibrational excited state it is easier to excite the NV center to the phonon sideband than to the zero phonon level. Since the decay of the vibronic levels is much faster (picosecond) than the electronic decay (nanosecond) the radiative decay will be from the ground vibronic level emitting a photon with lower energy [2]. The energy levels are described in Fig. 2.4. An illustration of the Franck-Condon rule, and the NV center optical spectrum are depicted in Fig. 2.5.
Fig. 2.4: Nitrogen-vacancy center energy-level structure. Radiative transitions indicated by solid arrows and non-radiative transitions by dashed arrows.

Fig. 2.5: (a) Illustration of the Franck-Condon rule via a schematic of the NV system excitation and emission cycle. The most probable transition is indicated by a green arrow. The non-radiative decay is denoted by a dashed arrow, while the radiative decay is indicated by a red arrow. (b-c) Photoluminescence spectra of NV centers at room temperature (b) and at 4K (c). Figure taken from [41].

2.4 Ground state fine and hyperfine structure

When defects involve vacancies, the absence of an ion breaks bonds in the crystal, producing unpaired electrons or dangling bonds which, to leading order, can be used to represent the
single-electron orbitals around the defect. In the case of a diamond there are four dangling bonds. Three bonds come from the carbons and one bond comes from the nitrogen. Since four spatial wave functions can be occupied by eight electrons the center will have a full chemical shell if it is populated by eight electrons. In the case of the negatively charged NV center the population of its outer shell is as follows: Three electrons come from the carbons, two electrons come from the nitrogen, and one electron comes from the lattice, totaling six electrons. It is just as accurate to think of the NV center as a two-hole spin-1 system rather than as a six-electron system. The NV optical ground state (g.s.) is a spin triplet and spatial singlet that transforms as $A_2$. The Hamiltonian of the g.s. is [38]:

$$
\begin{align*}
H_S &= (D_{gs} + d \parallel \Pi_z) S_z^2 + g_s \mu_B (\vec{S} \cdot \vec{B}) + d \perp \Pi_x (S_x^2 - S_y^2) + d \perp \Pi_y (S_x S_y + S_y S_x) \\
H_I &= P \vec{I}^2 - g_I \mu_N (\vec{I} \cdot \vec{B}) \\
H_{SI} &= A \parallel S_z I_z + A \perp (S_x I_x + S_y I_y)
\end{align*}
$$

$$\vec{\Pi} = \vec{\sigma} + \vec{E},$$

where $E$ is an electric field and $\vec{\sigma}$ stands for a strain field, $d$ is the g.s. electric dipole moment, $\vec{S}$ is the electronic spin, $\vec{B}$ is the external magnetic field, $\vec{I}$ is the nuclear spin, $g_s$ and $g_I$ are the electronic and nuclear g-factors respectively, and $\mu_N$ is the nuclear magneton. $H_S$ stands for the spin Hamiltonian and includes the spin-spin interaction and the interactions of the NV center with strain, magnetic, and electric fields. $H_I$ stands for the nuclear Hamiltonian and $H_{SI}$ describes the interaction of the nuclear spins with the electronic spins. As noted, the electronic spin in the NV center is $S = 1$ (two holes). The NV g.s. spatial wave function is a singlet that can be written as $e_x e_y - e_y e_x$ [4], where $e_x$ and $e_y$ are single-electron wave functions which are a linear combination of the dangling bonds and conserve the $C_{3v}$ symmetry of the center. The spin triplet g.s. energy levels can then be written as $(e_x e_y - e_y e_x) \otimes |11\rangle$, $(e_x e_y - e_y e_x) \otimes |-1-1\rangle$ and $(e_x e_y - e_y e_x) \otimes (|1-1\rangle + |1+1\rangle)$. The axial spin-spin interaction, described by the $D_{gs} S_z^2$ term, lifts the triplet degeneracy, where $D_{gs} \approx 2.87$ GHz, so both $m_s = \pm 1$ states differ equally from the $m_s = 0$ state. The axial spin-spin interaction makes the Hamiltonian no longer invariant under rotation, but the Hamiltonian is still invariant under reflection. This symmetry is responsible for the degeneracy of the $m_s = \pm 1$ levels. Applying a magnetic field along the $z$ axis will break this symmetry and will
lift the degeneracy of the \( m_s = \pm 1 \) levels. The effective matrix representation of the g.s. interaction with an electric field is the same as the matrix representation of the strain potential. For this reason we use \( \vec{\Pi} = \vec{\sigma} + \vec{E} \). The term \( S_x^2 - S_y^2 \) is responsible for the zero-field splitting of the g.s. levels and its coefficient is usually denoted by \( E \). In practice, except for very high pressure experiments (using a diamond anvil), \( D \) is governed by the spin-spin interaction. For this reason this coefficient is almost the same for different types of diamonds with differing internal strain. The transverse component that lifts the degeneracy of the \( m_s = \pm 1 \) states (even in the absence of an external magnetic field) is usually the strain [38]. The strain breaks the degeneracy of the \( m_s = \pm 1 \) state and yields a \( 2E \) separation between the new states. The amplitude of the strain splitting ranges from several hundred kHz (in CVD diamonds) up to a few MHz (in HPHT diamonds) [40]. As can be seen from \( \mathcal{H}_S \), an additional external magnetic field along the \( z \) quantum axis will further sperate the spin levels by a factor of 2.8MHz/G, where \( g_s = 2.003 \) is the Landé factor and \( \mu_B = 1.4 \text{MHz/G} \) is the Bohr magneton. Note, as described in Fig. 2.1, that there are 4 possible NV orientations, and therefore 4 different quantum axes, since the \( z \) quantum axis is defined by the nitrogen vacancy axis, and an external field that is aligned with one orientation is at 109.5° relative to the other 3 \( z \) quantum axes. Thus, the magnetic field that the other three orientations feel along their local \( z \) axis is \( B \cos(109^\circ) \). The effect of four NV orientations on the atomic spectrum in the presence of an external magnetic field will be discussed in chapter. 3.

The nuclear spin system generates a spin-spin interaction which can be described by \( P I_z^2 \) where \( P = -4.95 \text{MHz} \). The NV center can interact with its own nitrogen spin which is 1 for \(^{14}N\) and \( \frac{1}{2} \) for \(^{15}N\) and with the nuclear spin of one of its carbons or a nearby \(^{13}C\) carbon. The nuclear spin also interacts with the external magnetic field. However this interaction is very weak, almost 2000 times weaker than the interaction of the electronic spin with the magnetic field, since the nuclear magnetic moment is \( \mu_N = 0.76 \text{kHz/G} \) and \( g_I = -0.416 \). In this thesis, where we use magnetic fields of no more than a few dozen Gauss and a resolution not better than 1kHz, the above interaction is not observable and therefore we neglect it.

Due to the symmetry of the system the hyperfine Hamiltonian \( \mathcal{H}_{SI} \), which describes the interaction of the nuclear spin with the electronic spin, was also rearranged into axial and non-axial terms, where \( A_\perp \approx 2.7 \text{MHz} \) and \( A_\parallel \approx 2.16 \text{MHz} \). The \(^{14}N\) nuclei with its \( I = 1 \)
causes a 3-fold splitting in each of the Zeeman levels giving rise to a 9-state system. Since the allowed MW transitions ($\Delta m_s = \pm 1$), as depicted in Fig. 2.6, conserve the nuclear angular momentum projection ($\Delta m_I = 0$), the spectroscopy of each allowed transition shows a 3-fold splitting.

![Fig. 2.6: Schematic of the $^{14}N$ NV energy ground state levels arising from interactions splitting the ground state in the presence of an axial magnetic field. The energy splittings are expressed in terms of the hyperfine constants. Allowed transitions are indicated by double arrows. Electronic and nuclear spin projections are denoted by $m_s$ and $m_I$, respectively. The color code will be used in the next chapters. Taken from [39].](image)

2.5 Group theory and selection rules

From the spatial configuration of the NV center it is easy to see that the observable of this system will be invariant under specific symmetry operations, e.g. rotations of $0^\circ$, $120^\circ$ and $-120^\circ$ around the NV axis. These rotations will be denoted by $E$, $C^+$ and $C^-$. The system will also be invariant under reflections along the planes that contains the nitrogen the vacancy and one of the carbons; these reflections will be denoted by $R_1$, $R_2$ and $R_3$. This symmetry group is known as the $C_{3v}$ symmetry group. Based on the system symmetry alone (even without knowing the exact wave functions) we can already predict the system selection rules. For doing this we will use group theory. According to group theory, the wave functions of a system can be characterized by the way that they transform under symmetry operations.
We assume that there is a set of symmetry operations which are also unitary transformations \( \{U_k\} \), and that the Hamiltonian is invariant under all these transformations and that all the symmetry operations of the Hamiltonian are included in that group. We then find that

\[
\hat{U}_k \hat{H} \hat{U}^*_k = \hat{H},
\]

for every \( \hat{U}_k \). We can rewrite the Schrödinger equation

\[
\hat{H} |\Psi_n\rangle = E_n |\Psi_n\rangle,
\]

as

\[
\hat{U}_k \hat{H} \hat{U}^*_k |\Psi_n\rangle = E_n |\Psi_n\rangle,
\]

and if we apply one of the symmetry operations, i.e., multiply by \( U^*_k \) from the left, we obtain:

\[
\hat{H} U^*_k |\Psi_n\rangle = E_n U^*_k |\Psi_n\rangle.
\]

The existence of a symmetry group for the system raises the possibility of degeneracy. We can see that the new eigenstates, \( U^*_k |\Psi_n\rangle \), must have the same energy after applying the \( C_{3v} \) symmetry operations on \( |\Psi_n\rangle \). Unless \( U^*_k |\Psi_n\rangle = C |\Psi_n\rangle \) for all \( U^*_k \), the level is degenerate [42]. The new eigenfunctions \( U^*_k |\Psi_n\rangle \), or just \( |\Psi_n\rangle \), could have duplicates. The distinct degenerate eigenstates are related by \( |\Psi_n^k\rangle = \sum_i M_{k,i} |\Psi_i^n\rangle \) where the \( M \) matrices are the irreducible representations (IR) of the symmetry group. We can match each eigenfunction of the Hamiltonian to a set of IR \( M \) matrices, thus classifying each eigenfunction by its IR.

We start by finding the reducible representation. We can represent each atom using a vector, where \( N = \begin{pmatrix} 1 \\ 0 \\ 0 \\ 0 \end{pmatrix} \), \( C_1 = \begin{pmatrix} 0 \\ 1 \\ 0 \\ 0 \end{pmatrix} \), \( C_2 = \begin{pmatrix} 0 \\ 0 \\ 1 \\ 0 \end{pmatrix} \), and \( C_3 = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix} \). The representations matrices for the symmetry operations are depicted in table 2.1. If for example, we apply the matrix \( \Gamma(C^+) \) on the vector, we will switch the carbons and leave the nitrogen in its place. The \( 4 \times 4 \)
matrices in table 2.1 are not block-diagonal and are therefore reducible. The criteria for an irreducible representation is \( \sum |\chi_i|^2 = g \) where \( \chi_i \) is the character of the matrix \( i \) and \( g \) is the number of elements in the group [42]. The matrices of the IR representation of the \( C_{3v} \) point group are given in table 2.2. With the IR, we can find the eigenfunction that transforms as

\[
\phi_r = \frac{l_r}{g} \sum_{e} \chi_e^r \Gamma_e \sigma_i,
\]

(2.4)

where \( l_r \) is the order of the IR \( r \), \( g \) is the number of group elements, \( \chi_e^r \) is the character of the operation \( \Gamma_e \) in the IR \( r \) and we sum over all the elements of a certain IR. The projection operator gives 4 single electron orbitals which transform according to the symmetry operation (more precisely- IR) of the \( C_{3v} \) group [4]:

\[
\begin{align*}
a_N &= \sigma_1 \\
a_e &= \frac{\sigma_1 + \sigma_2 + \sigma_3}{3} \\
e_x &= \frac{2\sigma_2 - \sigma_1 - \sigma_3}{\sqrt{6}} \\
e_y &= \frac{\sigma_1 - \sigma_2}{\sqrt{2}}
\end{align*}
\]

(2.5)
Table 2.3: Character table of the $C_{3v}$ point group [42].

<table>
<thead>
<tr>
<th></th>
<th>$E$</th>
<th>$2C_3$</th>
<th>$3R$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A1$</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$A2$</td>
<td>1</td>
<td>1</td>
<td>-1</td>
</tr>
<tr>
<td>$E$</td>
<td>2</td>
<td>-1</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 2.4: Direct product multiplication table of the $C_{3v}$ point group [42].

<table>
<thead>
<tr>
<th>$C_{3v} \otimes C_{3v}$</th>
<th>$A1$</th>
<th>$A2$</th>
<th>$E$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A1$</td>
<td>$A1$</td>
<td>$A2$</td>
<td>$E$</td>
</tr>
<tr>
<td>$A2$</td>
<td>$A2$</td>
<td>$A1$</td>
<td>$E$</td>
</tr>
<tr>
<td>$E$</td>
<td>$E$</td>
<td>$E$</td>
<td>$A1 \oplus A2 \oplus E$</td>
</tr>
</tbody>
</table>

The first two orbitals are totally symmetric and we say that they transform as the $A_1$ IR. The $e_{x,y}$ orbitals transform as the $E$ IR to give a spatial doublet. The totally symmetric orbitals are mixed via Coulomb interaction [44] and we obtain a final basis \{\(a'_1, a_1, e_x, e_y\)\} such that

\[
\begin{align*}
    a'_1 &= \sqrt{1 - \alpha^2} \sigma_1 - \frac{\alpha}{\sqrt{3}} (\sigma_2 + \sigma_3 + \sigma_4) \\
    a_1 &= \alpha \sigma_1 - \sqrt{\frac{2}{3}} (\sigma_2 + \sigma_3 + \sigma_4)
\end{align*}
\]  

where $\alpha$ describes the amount of mixing with $0 \leq \alpha \leq 1$ and the $e_x$ and $e_y$ stay the same.

As mentioned above the NV center has two holes in its outer shell and therefore its orbitals are a linear combination of two single-electron functions. The representation of the two-hole spatial wavefunction is combined together with another representation of the same group, but a reducible one. The reduction of this combined representation into a direct sum of irreducible representations can easily be done using the orthogonal relations of its characters. The characters of the $C_{3v}$ group are depicted in table 2.3 and the direct product multiplication table is given in table 2.4. The two-electron wavefunctions are found using the Clebsch-Gordan coefficients. In the case of the g.s. the representation of the two-electron wavefunction is $A2$. As one can see from the direct product multiplication table this could be generated in only one way. According to the multiplication table it could be generated by the direct product of a single-electron wavefunction that transforms like $A1$ and a single-electron wavefunction that transforms like $A2$ (which we don’t have in our system). Alternatively, a two-electron wavefunction that transforms like $A2$ could be generated by taking the direct product of wavefunctions that belong to the $E$ representation. The expansion of the spatial
g.s. wavefunction with single-electron wavefunctions is depicted in eq. 2.7 [3]

\[ \Psi_{A2} = \sum_i \sum_j \left( \begin{array}{cc} E & E \\ i & j \end{array} \right) \left( \begin{array}{c} A2 \\ 1 \end{array} \right) e_i e_j = \frac{1}{\sqrt{2}} (e_x e_y - e_y e_x) \] (2.7)

<table>
<thead>
<tr>
<th>(A1 A1 A1)</th>
<th>= 1</th>
<th>(E E E A1)</th>
<th>= \frac{1}{\sqrt{2}} \left( \begin{array}{c} 1 \ 0 \end{array} \right)</th>
</tr>
</thead>
<tbody>
<tr>
<td>(A1 A2 A2)</td>
<td>= 1</td>
<td>(E E E A2)</td>
<td>= \frac{1}{\sqrt{2}} \left( \begin{array}{c} 0 \ 1 \end{array} \right)</td>
</tr>
<tr>
<td>(A2 A2 A1)</td>
<td>= 1</td>
<td>(E E E E)</td>
<td>= \frac{1}{\sqrt{2}} \left( \begin{array}{c} 1 \ 0 \end{array} \right)</td>
</tr>
<tr>
<td>(1 j k)</td>
<td>= \left( \begin{array}{c} 1 \ 0 \end{array} \right)</td>
<td>(E E E E)</td>
<td>= \frac{1}{\sqrt{2}} \left( \begin{array}{c} 0 \ -1 \end{array} \right)</td>
</tr>
<tr>
<td>(A2 E E)</td>
<td>= \left( \begin{array}{c} 0 \ 1 \end{array} \right)</td>
<td>(E E E y)</td>
<td>= \frac{1}{\sqrt{2}} \left( \begin{array}{c} -1 \ 0 \end{array} \right)</td>
</tr>
<tr>
<td>(1 j k)</td>
<td>= \left( \begin{array}{c} -1 \ 0 \end{array} \right)</td>
<td>(E E E y)</td>
<td>= \frac{1}{\sqrt{2}} \left( \begin{array}{c} -1 \ 0 \end{array} \right)</td>
</tr>
</tbody>
</table>

Table 2.5: The non-zero Clebsch-Gordan coefficients corresponding to the cartesian representations of the C3v irreducible representation [2, 3].

Getting the new wavefunctions out of the single-electron functions including space and spin degrees of freedom, is done by taking the direct product of the representation for each hole and its spin \( \Gamma \Psi = \Pi_n (\Gamma_{hn} \otimes D_{1/2}) \) where \( D_{1/2} \) is the representation of the spin 1/2 in the corresponding point group.

### 2.6 Electric dipole transitions

The symmetry of the g.s. spatial wave function is \( A_2 \). The components of the electric dipole moment operator transform like \( E_1, E_2 \) and \( A_1 \). It's clear that the electric dipole moment aligned along the NV axis transforms like the identity representation \( A_1 \). The electric dipole moments along the \( x \) and \( y \) axes transform like the first and second row of the \( E \) representation; in the following we will denote them as \( E_{x(y)} \). Suppose we have a wavefunction \( | \phi \rangle \) that transforms like the \( E_x \) representation of the \( C_{3v} \) symmetry and an operator \( \hat{O} \) that transforms like the \( E \) representation; we can then build four linear combinations that transform like the different representations of the \( C_{3v} \) symmetry using the Clebsch-Gordan coefficients [3] (see also [42] page 169 for a full derivation of these linear combinations and the Clebsch-Gordan
coefficients).
\( \hat{O}_x |\phi_x\rangle + \hat{O}_y |\phi_y\rangle \) belongs to the identity representation \( A_1 \),
\( \hat{O}_x |\phi_y\rangle - \hat{O}_y |\phi_x\rangle \) belongs to \( A_2 \),
\( \hat{O}_x |\phi_x\rangle - \hat{O}_y |\phi_y\rangle \) and \(-\hat{O}_x |\phi_y\rangle - \hat{O}_y |\phi_x\rangle \) belongs to \( E_x \) and \( E_y \) respectively.

The dipole transition between the ground state and the excited state is allowed if the matrix element \( \langle A_2 | e \cdot D | \phi \rangle \) is not zero. The overlap \( \langle \phi_1 | \phi_2 \rangle \neq 0 \) only if \( \phi_1 \) and \( \phi_2 \) belong to the same row of the same representation. \( \hat{O}_y |\phi_x\rangle \) and \( \hat{O}_x |\phi_y\rangle \) belong to a linear combination of \( E_y \) and \( A_2 \) and hence the matrix elements are non-zero, \( \langle A_2 | \hat{O}_y |E_x\rangle \neq 0 \langle A_2 | \hat{O}_y |E_y\rangle \neq 0 \).

\( \hat{O}_x |\phi_x\rangle \) and \( \hat{O}_y |\phi_y\rangle \) can be written as \( \frac{E_x + A_1}{\sqrt{2}} \) and \( -\frac{E_x + A_1}{\sqrt{2}} \) respectively, and hence \( \langle A_2 | \hat{O}_x |E_x\rangle = \langle A_2 | \hat{O}_y |E_y\rangle = 0 \).

2.7 Review of published work on excitation of NV centers with polarized microwave radiation

Our theoretical understanding of the NV center has also led us to disagree with several published works. Here we bring an example of such criticism. Under an external magnetic field of several Gauss and above, the NV eigenenergies \( (m_s = \pm 1) \) are well separated and a selective transition can be applied using on-resonance MW. In addition to selecting a transition to a specific sublevel by utilizing the correct frequency, the right polarization should be used; due to angular momentum conservation of MW transitions in the g.s., only circularly polarized radiation, \( \sigma^\pm \), can enable transitions from the \( m_s = 0 \) to the \( m_s = \pm 1 \) Zeeman sublevels.

Most of the MW antennae in NV experiments generate linearly-polarized MW radiation (i.e. a linear combination of \( \sigma^\pm \)) and consequently can induce transitions to both Zeeman states just by frequency selection. At small external magnetic fields, the picture is more complicated; not only are the eigenenergies only a few MHz apart (due to strain) but the eigenstates of the system are no longer the Zeeman states, but rather linear combinations of the Zeeman states because other terms in the Hamiltonian (e.g. the strain term) are becoming dominant.

Following the derivation of [38] we define the field spin states \( \{|0\rangle,|−\rangle,|+\rangle\} \) in terms of the \( S_z \) eigenstates \( \{|S,m_s\}\) as
\[
|0\rangle = |1,0\rangle, \\
|\rangle = e^{i\frac{\phi_{\epsilon}}{2}} \sin \frac{\theta}{2} |1,1\rangle + e^{-i\frac{\phi_{\epsilon}}{2}} \cos \frac{\theta}{2} |1,-1\rangle, \\
|+\rangle = e^{i\frac{\phi_{\epsilon}}{2}} \cos \frac{\theta}{2} |1,1\rangle - e^{-i\frac{\phi_{\epsilon}}{2}} \sin \frac{\theta}{2} |1,-1\rangle,
\]

(2.8)

where \( \tan \phi_{\epsilon} = \epsilon_y / \epsilon_x \), \( \tan \theta = \epsilon_\perp / B_z \), and \( \epsilon_\perp = \sqrt{\epsilon_x^2 - \epsilon_y^2} \) (with \( \epsilon_k \) being \( \Pi_k \cdot d_\perp \) from the g.s. Hamiltonian, Eq. 2.1). For \( B \gg \epsilon \), \( \theta = 0 \) and circular polarization will indeed produce a selective transition. Assuming low external magnetic field (compared to strain), \( \theta = \pi/2 \) and substituting into Eq. 2.8 we get the same amplitude for the \( |1,1\rangle \), and \( |1,-1\rangle \) states. Accordingly, using circularly-polarized MW radiation in the low magnetic field regime will yield a transition from \( |0\rangle \) to both states \( |\pm\rangle \) (assuming angular momentum conservation), and it seems impossible to achieve selective transitions using circular polarization. This seems to contradict the result published in [27]. The results of [27] are depicted in Fig. 2.7.

Fig. 2.7: Selective ODMR spectra at zero magnetic field measured by Mrózek et al. Taken from [27].

we assume happened in the experiment at low magnetic fields is the following: when taking into account the angles of the MW magnetic field in the NV frame, for an oscillating MW magnetic field \( \vec{M} \), the transition rates are [38]

\[
W_{0\rightarrow\pm} \propto \frac{1}{2} M_\perp^2 (1 \mp \sin \theta \cos \phi_{\tilde{M}}),
\]

(2.9)

where \( M_\perp = \sqrt{M_x^2 + M_y^2} \), \( \tan \phi_{\tilde{M}} = \frac{M_y}{M_x} \), and \( \phi_{\tilde{M}} = 2 \cdot \phi_M + \phi_{\epsilon} \). At a fixed direction of the strain, say \( \phi_{\epsilon} = 0 \), it is possible to align the MW field projections on the \( xy \) plane so that the MW
magnetic field will oscillate along the x axis (linear polarization, $M_y = 0$, $\phi_M = 0$ and thus $\phi_m = 0$), and consequently a selective transition to only $| - \rangle$ will be possible (see Eq. 2.9 with $\theta = \pi/2$ and $\phi_m = 0$). If, on the other hand, the MW radiation is polarized along the y axis ($M_x = 0$, $\phi_M = \pi/2$, and $\phi_m = \pi$), only a transition to the $| + \rangle$ state will occur. During their experiment a phase difference between the two antennae was used to generate circularly-polarized MW radiation. Since the electrical path from the signal generator to antenna a and antenna b is a function of the frequency, the phase difference was adjusted until a selective MW transition was obtained. The exact phase difference between the two antennae was not measured directly. What we assume happened is that the phase difference was adjusted in a way that generated linearly-polarized MW radiation and hence induced a selective transition to the $| + \rangle$ or $| - \rangle$ state. In the following we will demonstrate how it is possible to generate a linearly-polarized radiation by controlling the phase difference between the antennae that were in used in [27].

![Diagram](image)

**Fig. 2.8:** (a) The two antennae used by Mróz et al. to generate the polarized MW field. The NV center is positioned above at the same distance from antenna a and antenna b; taken from [27]. The generated magnetic field direction is $45^\circ$ and $-45^\circ$ to the vector perpendicular to the diamond surface. (b) $\vec{B}_a$ and $\vec{B}_b$ are the projections of the magnetic field produced by antenna a and antenna b on the NV xy plane. According to Eq. 2.9, linearly polarized MW radiation along the x axis will generate a transition to the $| - \rangle$ g.s.; Linearly polarized MW radiation along the $\hat{y}$ axis will generate a transition to the $| + \rangle$ g.s.; see text for details. In the configuration depicted here, the magnetic field is canceled along the x axis and lies along the y axis if the relative phase between the two antennae is zero. If the relative phase between the two antennae is $\pi$ the field will be canceled along the y axis and lies along the x axis.

Consider the case depicted in Fig. 2.8 and assume equal amplitudes for both antennae so that $B_a = B_b$. In this configuration, when the phase difference is zero, we will have linearly-
polarized radiation along the x axis. We can then switch from linearly-polarized radiation along the x axis to a linearly-polarized radiation along the y axis by having a relative phase of $\pi$. On the other hand a relative phase of $\pi/2$ will generate $\sigma^+$ polarization and a $3\pi/2$ will generate a $\sigma^-$ polarization.
3. STANDARD ODMR AND MAGNETIC SENSITIVITY

3.1 ODMR

The ODMR protocol is a basic procedure which is used for resolving the NV center g.s. energy level structure. At the heart of this protocol is the intersystem crossing. Due to the NV’s weak spin-orbit coupling, optical excitation and decay conserve the projection of the NV spin along the \( z \) axis. However when the NV decays through non-radiative channels the NV wavefunction goes through a singlet (in which the total spin is zero) and hence does not conserve its spin projection. The transition to the singlet level is facilitated by the spin-orbit interaction. The spin-orbit interaction can be written in terms of the angular momentum \( l_i \) and in this case it takes the following form [4]:

\[
H_{so} = \sum_k \lambda_{xy} (l^x_k s^x_k + l^y_k s^y_k) + \lambda_z l_z s_z,
\]

(3.1)

where \( \lambda_{xy} (\lambda_z) \) denotes the non-axial (axial) strength of the interaction. The non-axial part of the spin-orbit interaction links states with non-zero spin projections with singlets among different electronic configurations [4]. This interaction is responsible for the non-radiative decay channel of the \( m_s = \pm 1 \) excited state levels. The g.s. spin is usually measured by excitation to higher electronic levels and measuring the fluorescence due to spontaneous emission. If the g.s. spin projection on the NV axis is zero (i.e. \( m_s = 0 \)), the NV center will be optically excited to the \( m_s = 0 \) excited-state and will then decay back to the \( m_s = 0 \) g.s. emitting a 637-800 nm photon (this range is due to vibration levels, i.e. phonons, where the zero-phonon line is 637 nm). If the NV center is in an \( m_s = \pm 1 \) state it will be excited to an \( m_s = \pm 1 \) excited-state and will then have a \( \sim 30\% \) probability to decay to the \( m_s = 0 \) g.s. via the above noted non-radiative decay. This enables optical pumping of the NVs to the \( m_s = 0 \) g.s. and measuring the NVs spin-state by measuring its fluorescence,
since the fluorescence from the excited $m_s = 0$ state is higher (as it has a much weaker non-radiative decay channel). The optical excitation is most efficiently done with a green laser. The NV center then undergoes a fast vibronic decay (ps time scale) which preserves the spin projection. After pumping the NV to the $m_s = 0$ g.s. it is possible to excite the center to the $m_s = \pm 1$ g.s. with microwave (MW) radiation. The MW resonance frequency is a function of the external magnetic field (the energy levels are Zeeman-shifted). Excitation and emission wavelengths and observed spectra are depicted in Fig. 2.5.

The experimental setup for measuring ODMR is relatively simple. A green laser is used to excite the NV center, a dichroic mirror is used to resolved the excitation beam from the fluorescence and a MW source is scanned to detect the resonance frequencies. The fluorescence can be collected by photodiodes, cameras, or single-photon counters (SPCM). A spectrum is generated by plotting the fluorescence as a function of the MW frequency. A sketch of such a setup is depicted in Fig. 3.1. Several experimental techniques are commonly used to increase the signal-to-noise ratio (SNR). For example, in several instances during our work we used a lock-in amplifier with AM or FM modulation to increase the SNR.

The picture gets more complicated when we introduce all 4 different NV orientations in the diamond crystal. For example, applying an external magnetic field along one NV axis (which is also the $z$ quantum axis of that orientation) will yield four spectroscopic transitions.
Two of them are the $m_s = \pm 1$ transition frequencies of the NV orientation that is aligned with the magnetic field, and the other two are associated with the $m_s = \pm 1$ levels of the other 3 orientations because they all sense the same magnetic field at the same angle relative to their local $z$ axis ($109.5^\circ$). In this simple case we can retrieve the magnetic field amplitude by dividing the frequency difference of the magnetic levels of the aligned orientation from the g.s. spin-spin splitting of 2.87GHz by 2.8MHz/G. The typical situation is, however, that all 4 orientations have slightly different magnetic field projections and we thus have as many as 8 different transitions. For this general case, we need to insert the non-axial terms in $\mathcal{H}_S$, $B_x S_x$ and $B_y S_y$ with respect to each orientation’s local quantum axis. This complexity has also an upside as it enables us to sense not only the field’s magnitude but also its orientation [45] thus yielding a full description of the magnetic field. The most common cases are when the magnetic field is applied along the $z$ axis. An example of the NV spectrum with and without a magnetic field along the $\{111\}$ orientation axis is depicted in Fig. 3.2. When the external magnetic field is strong, the dot product between $\vec{B}$ and $\vec{S}$ in the Hamiltonian forces us to take into consideration both axial and transverse magnetic fields. While the axial magnetic field splits the $m_s = \pm 1$ levels, a strong off-axis magnetic field ($> 5$ mT) [32] changes the natural quantization axis of the NV center. The new quantization axis is then defined by the background magnetic field rather than by the intrinsic crystal field, and the spin sublevels $m_s = 0$ and $m_s = \pm 1$ are no longer eigenstates of the NV system. In this case, mixing of the spin-state populations causes a reduction in the spin polarization efficiency by optical pumping, and thereby the maximal achievable fluorescence intensity when the NV is under optical pumping [46]. This spin mixing disturbs ODMR-based imaging protocols at high magnetic field strengths, but also provides an alternate approach to magnetic imaging via the optical response of the NV center. In this thesis we did not reach high transverse magnetic fields and hence the mixing of the states as well as non-symmetric splitting of the $m_s = \pm 1$ levels were not taken into account.

3.2 Magnetic sensitivity

The ODMR protocol is widely used for NV-center based DC magnetic sensing. In the most naive way the magnetic field is calculated using the two resonances in the spectrum $\nu_+$ and
Fig. 3.2: NV center spectroscopy measured without (a) and with (b) an external magnetic field. The magnetic field is applied along the [111] crystal axis resulting in a strong splitting for the NVs with the [111] orientation and small splittings for the three other orientations. The linewidth in (a) is much larger than in (b) due not to the small Zeeman splitting, but mainly to the strain zero-field splitting.

The magnetic field is then calculated by the following formula

\[ B_z = \frac{\nu_+ - \nu_-}{2g_s\mu_B}. \]  \hspace{1cm} (3.2)

The fact that this protocol is based on changes in frequency (and not in the fluorescence) filters out noise sources such as laser instability and inhomogeneity of the NV’s concentration. When measuring small and fast changes of the magnetic field it is often easier to use a dif-
Fig. 3.3: Magnetic sensitivity. At the highest slope of a Lorentzian, a small frequency change $\delta \nu$ (in the driving field frequency or more relevantly in the transition frequency) causes a large corresponding change in the signal, $\delta S$. If the small change in frequency comes from an additional magnetic field, we can evaluate that additional field according to the relative change in the signal. Figure taken from [39].

Different method. The MW is tuned to be nearly on-resonance with a bias field or the zero-field transition. When the magnetic field is changed the fluorescence of the NV center is changed. By analyzing the fluorescence level it is possible to determine the change in the magnetic field. The optimal working point of the MW field occurs when $\frac{\partial S}{\partial B}$ is maximal, where $S$ is the collected fluorescence, namely, on the slope of the spectroscopic dip. In general the ODMR spectrum takes the form $S(\nu)$. When the magnetic field is changed the spectrum will be shifted and become $S(\nu) = S(\nu - \gamma B)$ where $\gamma = g_s \mu_B$. For small fields the change of $S$ depends on the local gradient of the spectrum $\Delta S = \frac{\partial S}{\partial \nu} \cdot \gamma B$ (Fig. 3.3). Assuming a Lorentzian shape for the ODMR (i.e. the reduction in signal as a function $\nu$), we obtain a general description of the ODMR signal [47]:

$$ S(\nu) = N_0 \left( 1 - \frac{C(\Gamma)^2}{(\gamma(B - B_0))^2 + (\Gamma)^2} \right), \quad (3.3) $$

where $N_0$ is the unperturbed photon count in regions of the spectrum far detuned from $B_0$ and $\Gamma$ and $C$ are the transition FWHM and contrast respectively.

As noted, the optimal working point occurs for the largest change in signal with respect
to a change in $B$, i.e., where the slope of the Lorentzian is highest. The maximum gradient occurs at $\nu = \pm \gamma B_0 \pm \Gamma / \sqrt{3}$ [47] and

$$\max \left| \frac{\partial S}{\partial \nu} \right| = \frac{N_0 C 3 \sqrt{3}}{\Gamma} \frac{8}{\sqrt{3}}.$$  

(3.4)

To ensure a signal-to-noise ratio greater than 1, we require $\Delta S > 1 / \sqrt{N} = 1 / \sqrt{Rt}$ (i.e. $\Delta S_{\text{min}} = 1 / \sqrt{Rt}$), where $N$ is the total number of photons collected in time $t$, and $R$ is the photon count rate. Realizing that $\delta \nu_{\text{min}} = \left( \max \left| \frac{\partial S}{\partial \nu} \right| \right)^{-1} \Delta S_{\text{min}}$, the minimum detectable field strength (or in other words the sensitivity) is

$$\delta B_{\text{min}} = \frac{\delta \nu_{\text{min}}}{\gamma} = \frac{1}{C N_0 \gamma \sqrt{R \Gamma}} \frac{8\Gamma}{3 \sqrt{3}}.$$  

(3.5)

It is customary to describe the sensitivity not as $\delta B_{\text{min}}$ but as:

$$\eta = \delta B_{\text{min}} \sqrt{t_m},$$  

(3.6)

where $t_m$ is the measurement time. The sensitivity will then be [47]

$$\eta = \frac{1}{C N_0 \gamma \sqrt{R} \frac{8\Gamma}{3 \sqrt{3}}}.$$  

(3.7)

The count rate $R$ is a monotonically increasing function of the laser power until it reaches the saturation intensity. Both $C$ and $\Gamma$ are influenced also by the microwave and laser power. Maximal sensitivity is achieved by optimising these parameters. This sensitivity calculation is a phenomenological calculation whereby the specific features of the NV center are hidden in $\Gamma$ which depends for example on the coherence time of the two-level system.

### 3.3 Hyperfine interaction with the nitrogen atom

The nuclear NV spin often plays an important role in NV magnetometry. As can be seen from the Hamiltonian (Eq. 2.1) each of the $m_s = \pm 1$ levels is split by the hyperfine interaction. In our samples the nitrogen ion is $^{14}N$ with spin 1 and the splitting it induces is on the order of 2.16 MHz. In accordance with Fig. 2.6, each transition is split into 3 sub-transitions.
The two batches of three peaks represent the transitions from the $m_s = 0$ to the $m_s = \pm 1$ states and each color of arrow represents a transition for a different $m_I$ state. The color code is the same as in Fig. 2.6: the blue arrows represent transitions with $m_I = -1$ nuclear spin. Green and orange lines are the transitions with $m_I = 0$ and $m_I = 1$ respectively.

depending on $m_I$. This is depicted in Fig. 3.4 where we use the same color code as in Fig. 2.6.
4. HOLE-BURNING SATURATION SPECTROSCOPY AND DOPPLER-FREE LIKE SPECTROSCOPY OF THE NV CENTER

NV-based magnetometer sensitivity is limited by the linewidth of the ODMR transition. The transitions are inhomogeneously broadened due to differences in the NV local environments, limiting the ensemble sensitivity. Diamond samples with more paramagnetic impurities have more inhomogeneous broadening. Hole-burning MW saturation procedure was suggested as a way to determine the origin of the inhomogeneous broadening [48]. In the first part of this chapter we will introduce the hole-burning saturation spectroscopy and show that this method is not accurate enough in order to determine the origin of the NV inhomogeneous broadening. In the second part of this chapter we will introduce a Doppler-free like spectroscopy protocol and show how this protocol can determine the origin of the inhomogeneous broadening (or at the very least identify which are its dominant contributions).

4.1 Hole-burning saturation spectroscopy

In a recent publication [48] the hole burning saturation spectroscopy method was suggested for studying the role of local magnetic fields in the inhomogeneous line-broadening process. In order to do so, a local magnetic field $\delta B$ and a local strain field $\delta \varepsilon$ were added to the Hamiltonian in Eq. (2.1). After neglecting the nuclear spin and the global strain terms the Hamiltonian was rewritten as [48]

$$\mathcal{H} = (D_{gs} + d || \delta \varepsilon) S_z^2 + g_s \mu_B (B_z + \delta B_z) S_z.$$  \hspace{1cm} (4.1)

The transition frequencies will then be

$$f_\pm = (D_{gs} + d || \delta \varepsilon) \pm g_s \mu_B (B_z + \delta B_z),$$  \hspace{1cm} (4.2)
where \( f_\pm \) are the transition frequencies from state \( m_s = 0 \) to state \( m_s = \pm 1 \). If we assume that the main contribution to the inhomogeneous line broadening originates from local strain fields, \( d \parallel \delta \epsilon \), and neglect the local magnetic field, \( \delta B_z \simeq 0 \), we find the relation

\[
f_+ = f_- + 2 g_s \mu_B B_z.
\]

(4.3)

On the other hand, if the main contribution to the line broadening is \( \delta B \), either from local fields or an external field gradient, we can set \( \delta \epsilon \simeq 0 \) and obtain

\[
f_+ = 2D g_s - f_-.
\]

(4.4)

Besides of their orientations, NVs differ by the projection of the nuclear spin \( m_I \) of the \(^{14}\text{N}\), for which \( I=1 \). In a hole burning saturation experiment we label the central transition frequencies (of a certain \( m_I \) population) by \( f_0^+ \) and \( f_0^- \). If we set the MW field to the resonance of, say, the \( |m_s = 0; m_I = +1\rangle \) to \( |m_s = +1; m_I = +1\rangle \) transition, it will “burn a hole” in the \( |m_s = 0; m_I = +1\rangle \) to \( |m_s = -1; m_I = +1\rangle \) transition, since the population in the state \( |m_s = 0; m_I = +1\rangle \) will already be excited. Namely, if a strong (pump) driving field is tuned to the first transition, a weak (probe) field tuned to the second transition will show reduced absorption or excitation. Here, we can describe \( f_+ \) as a pump field and \( f_- \) as a probe, and expect that if we apply a pump at \( f_+ \), we will cause a spectroscopic hole at \( f_- \). This hole will affect only NVs that are experiencing the same \( \delta B \) (or \( d \parallel \delta \epsilon \)) field.

### 4.1.1 Review of published work studying the origin of the inhomogeneous broadening [48]

The authors of [48] measured the pump-probe relations depicted in Fig. 4.1. They stated that since the pump-probe relations satisfy Eq. 4.4 this spectrum supports the claim that the origin of the inhomogeneous broadening is the local magnetic field. Looking carefully in the pump-probe relations of Fig. 4.1, it seems that what happened is a bit different, and therefore this measurement cannot reliably determine the source of the inhomogeneous broadening. Specifically, the pump and probe indeed interacted with the same populations but this population did not necessarily share the same local magnetic field but rather the same nuclear spin.
Fig. 4.1: “Pump-Probe” relations measured in [48]. The effect of changing the pump frequency ($f_+ = 2934.2$ MHz + $\Delta f$) on the hole center frequency $f$. The lock-in signal when the probe is off resonance is due to the NV fluorescence being modulated by the pump MW at the lock-in frequency and is a measure of the pump absorption. When the CW probe is resonant with the same population that is interacting with the pump, the lock-in signal due to the pump becomes smaller. See text for more details. Figure taken from [48].

Let us elaborate. In order to validate Eq. 4.4 where $f_+ + f_-$ is constant, one needs to move the pump a little to the right (higher frequency) and see the probe spectroscopic dip moving to the left (lower frequency) by the same amount. If we look at the ±2.0 peaks in Fig. 4.1 they indeed behave as required. However, if we look at Fig. 3.4 we realize that this behavior is simply due to a jump of the pump (and therefore the probe dip) to a new $m_I$ population which has a resonance about 2 MHz away. Indeed, according to this figure the $m_I$ transitions for the $f_+$ and $f_-$ frequencies are in reverse order relative to one another and this would give exactly the effect expected from Eq. 4.4. To really validate Eq. 4.4 one would have to narrow the scan so that it is always within the same $m_I$. This has been done at BGU as described in the following. Let us also briefly comment regarding the lower graph in Fig. 4.1: According to Eq. 2 in [48] if the pump ($f_+$) moves to the right (higher frequency) by some value X, then the hole ($f_-$) should go to the left by the same value X. However from the bottom graph of Fig. 4.1 you see that when the pump moves from +2.0 MHz to +3.2 MHz (i.e. by 1.2 MHz) the hole moves by just 0.6 MHz.

4.1.2 Experiment

As a first step, we repeated the experiment performed in [48]. In practice we carried out the experiment in the following way. We used an amplitude-modulated MW pump field with a
fixed frequency that corresponds to a specific HF transition \( f_0 \), and scanned around \( f_0 \) with a CW probe field. We set the scan wide enough to go over all three HF transitions. During the scan we measured the modulated fluorescence coming out of the diamond as a function of the probe frequency using a lock-in amplifier, where the on/off modulation of the pump was at the lock-in frequency (Fig. 4.2). When the pump beam is exactly on resonance \( f_0 \) but the probe beam is far from resonance, the probe will not affect the lock-in signal and we will get a high contrast in the lock-in due to the modulated pump beam. As we tune the probe frequency closer to resonance, the probe frequency and the pump frequency are now addressing NVs of the same population (same \( m_f \) and \( \delta B \) or \( \delta \epsilon \)) and since the probe is CW there is less modulated fluorescence light and we get a reduction in the lock-in signal. The net result of this fixed-pump scanned-probe process is the creation of a spectroscopic hole at \( f_0 \). A demonstration of the hole burning technique is depicted in Fig. 4.3. We can clearly see the jumps from one \( m_f \) transition to the next.
Fig. 4.3: Spectroscopic holes measured using a lock-in amplifier. The lock-in frequency is 36.6kHz. The color code is the same as in Fig. 2.6 and Fig. 3.4: dots (data) and lines (fits) are for the pump laser set to the $m_I = +1$ (red), $m_I = 0$ (green), and $m_I = -1$ (blue) transitions. In each spectrum we can clearly see the other two transitions as small side dips. A discussion regarding the possible origins of the side dips can be found in [39]. The average linewidth of the holes is 0.9MHz, which is $\sim 200$kHz less than an ODMR width. Figure taken from [39].

Next, in order to avoid the wide scan problem described previously, we limit our scans to be within a specific $m_I$ transition. This requires a scan with a step size much smaller than previously employed. In order to find the main contributor to the inhomogeneous broadening we plotted the frequency of the hole as a function of the pump frequency. If indeed the main contributor to the line broadening is $\delta B$ then according to Eq. (4.4) we expect the slope of this plot to be $-1$. We made a series of tests where we changed the pump frequency with a 100kHz step size, starting at the central frequency of the $m_I = 0$ transition, and scanned with the probe to observe the hole. Each test yielded a hole that was later fitted to a Lorentzian and the hole location was extracted. We plotted the hole location vs. the pump frequency and made a linear fit to extract the slope. For a modulation frequency of 36.6kHz we found a slope of $-0.59$ (Fig. 4.4), and for a modulation frequency of 426Hz we found a slope of $-0.99$ (Fig. 4.5). For a different magnetic field and the same modulation frequency (426Hz) we found a slope of $-0.83$ (Fig. 4.6). In all experiments we expected a slope of $-1$ regardless of the different magnetic field or modulation frequency. We believe that one possible reason for the inconsistency in the slope value, could be thermal fluctuations: if we look again at Eq (4.4) we can see that not only $f_+$ can generate a change in $f_-$ but so can $D_{gs}$. $D_{gs}$ is not constant and can have thermal fluctuations as high as $-75$kHz/K at room temperature [49]. Thus, even small temperature changes such as $\pm 0.5$K can appear as a 75kHz change in the hole location.
At this point, although we did show a negative correlation between \( f_+ \) and \( f_- \), it seems that we have not clearly proven that the main inhomogeneous broadening mechanism is due to the local magnetic field environment. Similar uncertainties were observed in the work done in [48] as conveyed to us in private communications [50].

Figure 4.4: Migration of hole \((f_-)\) using a modulation frequency of 36.6kHz. Data in blue, linear fit in red. The pump frequency was changed in small steps of 100kHz. The fitted slope is \(-0.59 \pm 0.21\) and an intercept of \(4567 \pm 615\)MHz (which according to Eq. (4.4) is expected to be \(2D_{gs} = 5740\)MHz). Figure taken from [39].

Figure 4.5: Migration of the hole \((f_-)\) using a modulation frequency of 426Hz. Data in blue, linear fit in red. The pump frequency was changed in small steps of 100kHz. The fitted slope is \(-0.99 \pm 0.13\) and an intercept of \(5809 \pm 396\)MHz (which according to Eq. (4.4) is expected to be \(2D_{gs} = 5740\)MHz). The slight change in the pump frequencies relative to Fig. 4.4 is probably due to a re-alignment of the setup together with a strong gradient of the external magnetic field. Figure taken from [39].

4.2 Doppler-free like spectroscopy of the NV center

In the search for a more robust method we introduced a novel spectroscopic method which is analogous to Doppler-free spectroscopy in vapor. In Doppler-free spectroscopy we use
two counter-propagating laser beams with the same frequency and observe the interaction of the laser light with atoms having different velocities. The atoms can be classified according to their velocities, where $P(v)$ is their velocity probability distribution. Due to the Doppler effect, one laser beam will interact with atoms of class $P(v)$ while the other interacts with $P(-v)$. Therefore, the two fields will always interact with different populations (due to the Doppler shift) except for the population with zero velocity (component along the optical axis). In such a case, the Doppler shift is zero and if the radiation field is resonant with the atomic transition, both fields will address the same population. If the pump field has higher amplitude than the probe, then it will “burn a hole” in the ground level of the $P(v = 0)$ population, which can be detected by the probe since the pumped atoms will barely interact with the probe and we expect to observe a reduction in probe absorption (or increased probe transmission). Since the width of this hole is only limited by the atomic natural linewidth and the laser linewidth, the observed linewidth is narrower than the Doppler-broadened linewidth. In our case the NVs natural linewidth is broadened mainly by the fact that each of the NVs in our ensemble can have a slightly different environment, and consequently a different transition frequency causing inhomogeneous broadening. As before we will work with the $m_s = 0 \rightarrow m_s = +1$ and $m_s = 0 \rightarrow m_s = -1$ transitions. Our method is analogous to Doppler-free spectroscopy in vapor in the sense that both cases use two radiation fields that always address different populations except when both fields are on
resonance with the center of the inhomogeneously broadened distribution. We replace the counter-propagating beams in the vapor experiment with \( f_+ \) (pump) and \( f_- \) (probe). A main feature of the new method is that we fix the frequency gap between the two fields; namely, we scan at different pump frequencies while keeping the frequency gap constant, so that the probe frequency change follows exactly the change of the pump frequency. Focusing on the \( m_I = 0 \) transition, we fix the frequency gap to be \( \Delta f = f_{m_s=+1; m_I=0} - f_{m_s=-1; m_I=0} \), where the latter are the resonance frequencies. We scan with a CW probe while the pump frequency is pulse modulated as before. The signal is detected using a lock-in amplifier locked on the modulation frequency of the pump. When the probe is on the \( m_I = \pm 1 \) transition the pump is on the \( m_I = \mp 1 \) transition and the probe has no impact as it always addresses \( m_I \) states that are different than those addressed by the modulated pump, and we expect to see a regular ODMR signal generated by the pump. When we scan within the \( m_I = 0 \) linewidth, both radiation fields address the same \( m_I \). However, when they are not directly on resonance, the negative sign in Eq. (4.4) dictates that they are still addressing different populations; when the probe and pump fields are “red-detuned” with respect to the transition frequency, the hole is “blue-detuned” and the probe is not at the hole frequency and has no impact on the lock-in signal. Only if both fields are on resonance (\( f_{0}^{0} \) and \( f_{0}^{i} \)) will they address the same population and we should observe a narrow “Doppler-free” type hole at the center of the ODMR peak. The setup is similar to the hole burning saturation spectroscopy setup (Fig. 4.2).

We modeled our expected experimental line shape as the sum of four Lorentzians. We started by assuming that the HF transitions have the same amplitude and derived the general form:

\[
S(f_+) = \sum_{i=-1}^{+1} a_{1} \frac{\gamma_{1}}{2} \frac{1}{(f_+ - f_{0}^{i})^2 + \left(\frac{\gamma_{1}}{2}\right)^2},
\]

where \( f_+ \) is the pump frequency (+ represents transition to \( m_s = +1 \)), \( f_{0}^{i} \) is the transition frequency to \( m_s = +1 \) with \( m_I = i \) and \( a_{1} \) and \( \gamma_{1} \) are the amplitude and FWHM respectively. Thus, Eq. (4.5) simply describes a regular ODMR signal of the HF transitions using a lock-in (i.e., three Lorentzians with equal positive amplitude and width). As explained above, the CW probe affects the pump lock-in signal only when both fields are near \( m_I = 0 \). The hole
in the pump signal should be described by a single Lorentzian. Since the hole location is not constant we describe the hole using the following Lorentzian:

\[ h(f_-, t) = a_2 \frac{\gamma_2^2}{2} \frac{1}{(f_- - t)^2 + \frac{\gamma_2^2}{2}}. \]  \( (4.6) \)

where \( f_- \) is the probe frequency (\( - \) represents transition to \( m_s = -1 \)) and the location of the center of the Lorentzian \( t \) changes with \( f_+ \). We would like to write \( h(f_-, t) \) as a function of \( f_+ \). Consider the case where \( f_+ \) (pump) is 1 MHz below \( f_{+i}^0 \), then \( f_- \) (probe) is 1 MHz below \( f_{-i}^0 \), but \( t \) (hole) will be 1 MHz above \( f_{-i}^0 \). The relation between \( t \) (hole) and \( f_{-i}^0 \) can be written as

\[ f_{-i}^0 = \frac{t + f_-}{2}. \]  \( (4.7) \)

Eq. 4.7 allows us to write \( h(f_-, t) \) as \( h(f_-) \). In order to write \( h(f_-) \) as \( h(f_+) \) we will use the relation \( f_+ - f_- = \Delta f \). We can replace \( f_- - t \) in Eq. 4.6 using the following relation

\[ f_- - t = 2f_- - 2f_{-i}^0 = 2(f_+ - \Delta f) - 2(f_{+i}^0 - \Delta f) = 2f_+ - 2f_{+i}^0. \]  \( (4.8) \)

We can therefore write Eq. 4.6 as

\[ h(f_+) = a_2 \frac{\gamma_2^2}{2} \frac{1}{(2f_+ - 2f_{+i}^0)^2 + \frac{\gamma_2^2}{2}}. \]  \( (4.9) \)

The total signal will be \( h(f_+) + S(f_+) \). A simulation of the signal for the case where \( f_{+i}^0 = f_{+0}^0 \) is depicted in Fig. 4.7a. The simulation predicts that using the current settings we should observe a small “Doppler-free hole” at the center of the \( m_I = 0 \) transition with half the width of the hole, and that the other two \( m_I \) transitions (\( m_I = \pm 1 \)) will not be affected. In order to test the simulation we used the setup described in Fig. 4.2. The frequency gap was tuned to be \( \Delta = f_+ - f_- \) where \( f_+ \) is on resonance with the \( |m_s = 0; m_I = 0 \rangle \) to \( |m_s = +1; m_I = 0 \rangle \) transition, and \( f_- \) is on resonance with the \( |m_s = 0; m_I = 0 \rangle \) to \( |m_s = -1; m_I = 0 \rangle \) transition. We then scanned around the \( m_I = 0 \) transition frequency (to \( m_s = -1 \) for probe and \( m_s = +1 \) for pump). We observed a pseudo Doppler-free hole inside the transition lineshape. The observed signal was fitted to \( h(f_+) + S(f_+) \) and is found to be in a good agreement with
theory. The experimental results are depicted in Fig. 4.7b.

In conclusion, we based our model on Eq. 4.4 which states that the main contribution to the inhomogeneous broadening comes from the local magnetic field, thus substantiating our assumption.

Finally, one may be tempted to think of the new technique as being able to improve the magnetic sensing sensitivity, but this is not so. Although the width of the hole in our simulation and measurements is smaller than the regular ODMR signal, the depth of the hole is much weaker. As was shown in chapter 3 a better magnetic sensitivity is achieved when $\frac{\partial S}{\partial B}$ is larger. In the data presented in Fig. 4.7b the Doppler-free spectroscopy does not exhibit a larger $\frac{\partial S}{\partial B}$ and hence in this case this protocol will not improve the magnetic sensitivity of the system.
(a) Phenomenological simulation of “Doppler-free” type spectroscopy. We plotted the three Lorentzians of the ODMR [Eq. (4.5)] together with Eq. (4.6). The parameters were taken from the results of ODMR and hole-burning experiments. The $x$ axis represents the detuning of the $f_+$ (pump) from $f_{x_0}^0$ (i.e., $\delta f$). The small hole at center is due to the fact that only when both fields are addressing the same population can the probe “steal” population from the pump. The two Lorentzians at the sides are the ODMR lineshapes generated by the probe-pump interplay. Figure taken from [39].

(b) Experimental results for “Doppler-free” type spectroscopy. The data, shown in blue, is fitted to four Lorentzians (red curve). The $x$ axis represents detuning of the $f_+$ (pump) from $f_{x_0}^0$ (i.e., $\delta f$) as measured in an ODMR experiment. The width of the hole is $338 \pm 53$kHz. The model gives an excellent fit to the data with an R-square value of 0.999. Figure taken from [39].

Fig. 4.7
5. ADIABATIC BEHAVIOR OF THE NV CENTER

5.1 Theoretical background and motivation

5.1.1 Motivation

Characterizing theoretically and experimentally the adiabaticity of a quantum system is an important part of understanding its underlying physics. By adiabaticity we mean its ability to stay in the same quantum state even though external conditions may vary. Aside from being an interesting topic on a fundamental level, such a study may also have practical implications. Let us give an example.

A modification of quantum mechanics, or some self-gravitational effects, are hypothesized to appear when one reaches a spatial quantum superposition having an extent of 100 nm or more for objects as heavy as $10^9$ AMU. One of the candidates for such an experiment is a single NV in a nano-crystal [51]. A suggested experiment is to place a nano-crystal in a magnetic field gradient and then put it into a superposition of its g.s. spin projections. The diamond trajectory will then be depend on its internal quantum state as in a Stern-Gerlach experiment. By recombining the trajectories and measuring the quantum state it would be possible to study the properties of weak fields such as gravitation [51]. This kind of large-mass interferometry requires the ability to spatially trap the NV nano-diamonds. Currently it is possible to trap nano-diamonds using ion traps [52] or optical tweezers [53, 54, 55]. Recently, magnetic fields were also used to trap nano-diamonds [56]. These traps, however, do not depend on the NV’s internal degrees of freedom and have some technical limitations, such as heating the diamonds by laser light. An alternative way is to use a magnetic trap, similar to what is done with ultra-cold atoms, whereby the interaction between the spin and the magnetic field provides the trapping force.

Although NV centers exhibit several atom-like features they differ from atoms in several ways. One of the main differences between the NV centers and alkali-like atoms is the
direction of the quantum axis. While the quantum axis of atoms is defined by the direction of the external magnetic field, the NV quantum axis is defined mainly by the nitrogen-vacancy axis. In atom traps the trapping is enabled by the weak-field seeking state of the atoms. When the atom moves across the magnetic trap, its spin follows the varying direction of the magnetic field. This ability to stay in the same quantum state (e.g. anti-parallel to the external magnetic field), even as its direction and amplitude varies, is termed adiabaticity. In order to understand whether NVs could be trapped in an atom-like magnetic trap it is important to understand their behavior under rapid magnetic changes.

Even if the diamagnetic levitation [56] may be found to be superior, levitation and trapping a macroscopic body by its quantum state, where techniques like optical pumping are used, has to the best of our knowledge never been done, and it remains an important and attractive goal. In any case, currently our main motivation is to better understand the fundamental physics of the NV center, and how its Hamiltonian behaves under rapidly changing magnetic fields.

In this chapter we study both theoretically and experimentally the behavior of the NV spin under rapid changes of the external magnetic field, namely, we examine the question of adiabatic behavior of the NV center. This may form a significant step towards the first-ever trapping of a macroscopic object by way of its quantized spin.

### 5.1.2 Theoretical background

We start by taking an NV center and positioning it in an external magnetic field $B_z$ (aligned along the NV axis). The g.s. $m_s = \pm 1$ energy levels are then split by $2g_s \mu_B B$. Next we apply a 532 nm laser pulse and polarize the NV into the $m_s = 0$ level. Using an on-resonance MW $\pi$ pulse we transfer the population to the $m_s = 1$ state. We now change the external magnetic field as a function of time until $B_z = -B_z$. In order to find the adiabatic (non-adiabatic) regime, we will utilize the $m_s = 1$ and $m_s = -1$ states and solve the time-dependent Schrödinger equation $i\hbar \frac{\partial}{\partial t} \Psi = \hat{H} \Psi$. The state vector can be written as $\Psi(t) = \begin{pmatrix} \psi_1(t) \\ \psi_0(t) \\ \psi_{-1}(t) \end{pmatrix}$.

Taking into account the spin-spin interaction $D_{gs}S_z^2$, the interaction with an external magnetic
field $g_s \cdot \vec{\mu}_B \vec{S} \cdot \vec{B}$, and the interaction with strain $E(S_x^2 - S_y^2)$ we can write the Hamiltonian as

$$H = g_s \cdot \vec{\mu}_B \cdot \begin{pmatrix} \frac{D_{gs}}{\hbar g_s \mu_B} + B_z & \frac{B_x - iB_y}{\sqrt{2}} & \frac{E}{\hbar g_s \mu_B} \\ \frac{B_x + iB_y}{\sqrt{2}} & 0 & \frac{B_x - iB_y}{\sqrt{2}} \\ \frac{E}{\hbar g_s \mu_B} & \frac{B_x + iB_y}{\sqrt{2}} & \frac{D_{gs}}{\hbar g_s \mu_B} - B_z \end{pmatrix}.$$  

(5.1)

We solved these equations for two cases. In the first case we considered changes in the magnetic field along the $z$ axis alone, with $B_x = 0$, $B_y = 0$ and $B_z = B_0 \cos(\omega t)$. In the second case we took the magnitude of $\vec{B}$ to be constant but the direction of $\vec{B}$ was changed as a function of time $B_x = B_0 \sin(\omega t)$, $B_y = 0$ and $B_z = B_0 \cos(\omega t)$, so that it rotates in the $xz$ plane. The initial conditions were $\Psi(t = 0) = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix}$. The solution of the Hamiltonian for the case where $B_z = B_0 \cos(2\pi ft)$, $B_x = 0$, and $B_y = 0$, with $B_0 = 100$ G, $f = \frac{1}{2}$=100 kHz, and $E = 2.8$ MHz, as a function of time $t$, is depicted in Fig. 5.1. In order to better understand the behavior of the system and to compare it with our experiments, we generated maps of the $m_s = 1$ population (after the change of the magnetic field). Maps of the population changes as a function of the magnetic field amplitude and frequency for the case $B_z = B_0 \cos(2\pi ft)$, $B_x = 0$, $B_y = 0$ and the case $B_z = B_0 \sin(2\pi ft)$, $B_x = 0$, and $B_z = B_0 \cos(2\pi ft)$ are depicted in Fig. 5.2 and Fig. 5.3 respectively.

5.2 Experimental apparatus

5.2.1 Sample and magnetic fields

In order to generate and control fast changes of high magnetic fields, a chip with two crossing wires was fabricated. By locating the NVs close to the wires it is possible to generate high magnetic fields with relatively low currents, while at the same time the low inductance of the wire enables fast changes of the field. For this experiment we use an HPHT \{110\} diamond with a 100 nm layer of NVs (E6110). The [-11-1] and [1-1-1] orientations are found in the plane of the diamond surface with an angle of 109.5° between them (Fig. 5.4b). By measuring the ODMR signal of the diamond for magnetic fields aligned along the $x$ axis (perpendicular
Fig. 5.1: (a) A simulation of the $m_s = \pm 1$ states eigen-energies as a function of time in the presence of a magnetic field $B_z = B_0 \cos(2\pi ft)$, $B_x = 0$, $B_y = 0$, with $B_0 = 100$ G, $f = \frac{T}{2} = 100$ kHz, and $E = 2.8$ MHz. (b) The probability of finding the system in the original eigenstate ($m_s = +1$ in the field frame), using the same parameters as in (a), drops to 0.58 at $t = T/2$ where $T$ is the period of the magnetic field change (10 $\mu$s). Note that an abrupt change of the population happens at $t = 2.5 \mu$s when the rate of change of the magnetic field is highest.

Fig. 5.2: A map of the probability of finding the system in its original eigenstate $m_s = +1$ (in the field frame) after the magnetic field is changed from $B_z$ to $-B_z$. In this figure we consider the situation at time $T/2$ for the case where $B_z = B_0 \cos(2\pi ft)$, $B_x = 0$, $B_y = 0$. The map was generated using a similar calculation to the one depicted in Fig. 5.1b. Note that in the following experiments we utilize the $m_s = -1$ state, but this does not alter the results. In order to generate this map we utilized the fact that $D$ (the spin-spin interaction) is much larger than the other elements and hence the $m_s = 0$ level is well separated from the $m_s = \pm 1$ levels. Using the rotating-wave approximation we reduced our Hamiltonian into an effective 2D Hamiltonian. A detailed explanation of this approximation is given in Appendix A.
Fig. 5.3: The probability of finding the system in the original eigenstate ($m_s = +1$ in the field frame), after time $T/2$ for a magnetic field of $B_x = B_0 \sin(2\pi f t)$, $B_y = 0$, and $B_z = B_0 \cos(2\pi f t)$, for different magnetic field magnitudes and frequencies. In this configuration the absolute value of the magnetic field is kept constant and its direction is changed from $z$ to $-z$. For low frequencies the population remains unchanged as the NV electron spin follows the magnetic field. For higher frequencies the change is no longer adiabatic and the system may be found in different eigenstates. Note that unlike the 1D situation depicted in Fig. 5.2, in this 2D case, a high magnetic field preserves adiabaticity. This simulation was carried out by solving the full 3D Hamiltonian using cluster computing. Note that in the following experiments we utilize the $m_s = -1$ state, but this does not alter the results. Similarly, in the following we exchange the definitions of the x and y directions, but again this does not alter the results.

to the long diamond edge) and y axis (perpendicular to the short diamond edge), we verified the directions of the orientations with respect to the diamonds edges. Since the ODMR for magnetic fields aligned along the x and y axes showed a \{100\} and \{110\}-like spectrum respectively, we concluded that the [-11-1] and [1-1-1] orientations are oriented $\sim 35.5^\circ$ from the short diamond edge (Fig. 5.4b). The chip edge was attached to a white ABS structure for keeping the [-11-1] orientation parallel to one wire and perpendicular to the other wire (Fig. 5.4). Cancellation of the earth’s magnetic field can be done using Helmholtz coils (Fig. 5.5d), but we did not use them in the measurements presented in this work.
Fig. 5.4: (a) The chip for the adiabaticity experiment. The silicon wafer, with 2\,\mu m thick gold wires, is mounted on a copper base. The chip pads (visible) are connected to an ac current supply. The two crossing wires connected to the pads are just 10\,\mu m wide and therefore hardly noticeable. (b) A zoom-in on the crossing wires. A \{110\} diamond is placed above the cross wires. The chip right edge is attached to a white ABS structure (visible in (a) and cutting the figure here) which is used for aligning the [-11-1] orientation parallel to one wire and perpendicular to the other wire. By controlling the currents in the wires we can manipulate the effective magnetic field in 1D along \(z\) or to rotate it in the \(x-z\) plane, where the coordinate system is that of the [-11-1] NV orientations (\(z\) being the NV axis parallel to the [-11-1] direction).

5.2.2 MW chain

The MW radiation is produced by a signal generator (SRS384), and the signal is modulated using a MW switch (Minicircuits-ZASW-2-50DR). The modulated signal is then sent to a MW amplifier (ZHL-16W-43+). In order to generate a strong and relatively homogeneous MW field, an omega-loop antenna ($r \sim 0.5\,\text{mm}$) is used. The omega loop is placed on the upper side of the diamond (Fig. 5.5a). The complete experimental setup is presented in Fig. 5.5.

5.2.3 Optical setup

Confocal microscopy is performed by excitation with green light (532 nm) supplied by an optically pumped semiconductor laser (Coherent V18). The output beam is focused onto an AOM to achieve short rise and fall times. The beam is then recollimated and injected into a single-mode fiber to ensure a TEM00 gaussian beam. The diameter of the beam outside the fiber is larger than the diameter of the objective pupil so the objective is exposed to an
approximately plane wave (this is necessary for focusing the beam to a focal point which is as small as possible). The objective (Olympus, Pro Plan) has a numerical aperture of N.A.=0.6 and a maximal working distance of d=4.2 mm. The laser power outside the fiber was measured to be $\sim 12$ mW. The relatively high power needed (for achieving saturation intensity) is due to the use of an air objective. The red fluorescence emitted by the NV centers is collected with the same objective used to focus the incident green light, and is transmitted through a dichroic mirror (Semrock) mounted in a cube and then through a long-pass filter. The cube is mounted on a 1D translation stage which is controlled by an actuator while the sample itself is mounted on a 2D translation stage. Two lenses and a 15 $\mu$m pinhole are used to filter out fluorescence originating from outside the focal plane. The fluorescence is then aligned and focused on an Excelitas SPCM-AQRH14 single-photon counter module. A neutral density filter is used in cases of saturation of the SPCM.
Fig. 5.5: (a) The MW omega-loop antenna placed on the diamond. The loop antenna is connected on one side to the MW signal and on its other side to a 50 Ω terminator. (b) Optical excitation is provided by an optically pumped semiconductor laser (Coherent). The excitation laser is pulsed by focusing it through an AOM (Crystal Technology). (c) A sketch of the optical setup. The beam is focused onto the sample using a , NA = 0.65, air objective (Olympus). The NV fluorescence is collected through the same objective and separated from the excitation beam using a dichroic filter (Semrock). The light is additionally filtered and focused onto a single-photon counting module. (d) A photograph of the system inside the Helmholtz coils.

5.3 Experimental sequence

In order to check our simulations we need to carry out the following experimental sequence:

(a) After optical pumping to $m_s = 0$, we prepare the system in the $m_s = -1$ state with a first $\pi$ pulse.

(b) We change the magnetic field from $B_z$ to $-B_z$ either along the $z$ axis or by rotating it in the $zy$ plane.

(c) We measure the population of the $m_s = -1$ state using a second $\pi$ pulse which should transfer the population back to $m_s = 0$ if the evolution was adiabatic.
Preparing the system in the $m_s = -1$ state is done in two steps. The first step is to pump the system into $m_s = 0$ with green light for 4 µs. The next step is to transfer the population to $m_s = -1$ by applying a MW $\pi$ pulse. The determination of the $\pi$ pulse duration is done by finding the Rabi frequency of the system. An example of a Rabi sequence and a typical Rabi curve are depicted in Fig. 5.6. Schematics of the experimental sequences are depicted in Fig. 5.8a, Fig. 5.9a, and Fig. 5.10a.

The next step is to change the external magnetic field. Current is supplied to the chip by a two-channel current source modulated by arbitrary waveform function generators (Kesight 33250 and Kesight 33220). For generating the magnetic fields a crossed-wire configuration is fabricated on a single-layer chip (2x10 µm$^2$ wire cross section). For injecting currents in both wires simultaneously, a custom-made two-channel floating current source is used. Off-axis magnetic fields may cause mixing of the $m_s = 0$ and $m_s = \pm 1$ levels. Since in our setup, the three orientations are not aligned along the magnetic field direction, the operation of the magnetic field involves mixing of their g.s. levels and therefore a reduction of the total fluorescence (Fig. 5.7b). ODMR measurements for currents in the $B_z$ wire (responsible for the magnetic field aligned along the NV axis) and the $B_y$ wire (responsible for the transverse magnetic field) are used for calibrating the currents into magnetic fields, finding the on-resonance frequencies, and verifying the orientation of the magnetic field (Fig. 5.7). A magnetic field aligned along the NV axis ($B_z$) is expected to generate a $\{111\}$-like spectrum with one orientation shifted $\sqrt{3}$ times more than the three other orientations, while the three other orientations’ transition frequencies are expected to overlap (Fig. 5.7c). The ODMR caused by a transverse magnetic field ($B_y = [1 - 10]$) is expected to generate a $\{110\}$-like spectrum with two orientation shifted by $\Delta \nu = \frac{1}{\sqrt{3}} g_s B_z \mu_B$ and the other two orientations suffering much smaller shift (Fig. 5.7d).

In our sequences, adiabatic behavior means the following: first the system is exposed to a magnetic field $B_z = B_{z0}$ and then the first MW $\pi$ pulse transfers the system into $m_s = -1$ (dark state), where we define $m_s$ relative to the local quantum axis which is defined by the magnetic field direction. The magnetic field is then changed to $B_z = -B_{z0}$; if the system is adiabatic, the projection of the NV spin on the NV $z$ axis in the magnetic field frame is conserved and the transition frequency of the populated $m_s = -1$ state is the same as the
frequency that was used for the first $\pi$ pulse. In this case the system will end up in $m_s = 0$ (bright state). Non-adiabatic behavior means that while the first MW $\pi$ pulse transfers the system into $m_s = -1$ (dark state), when the second MW $\pi$ pulse is applied, some of the population has already flipped to the $m_s = +1$ (dark state) due to the change of the magnetic field. Since the second MW $\pi$ pulse is at the same frequency as the first $\pi$ pulse, it is off-resonance with respect to the $m_s = +1$ state. Some of the population will therefore remain in a dark state and the fluorescence will be lower.
Fig. 5.6: (a) Pulse sequence for measuring Rabi oscillations. (b) The fluorescence response for the system that is initially in $m_s = 0$ (red line) and $m_s = -1$ (blue line) immediately after turning the laser beam on. The normalized fluorescence in all our measurements is calculated as $[(A - B) / (A + B)]$, where $A$ is the number of photons detected by the SPCM during 300 ns immediately after turning the laser beam on, and $B$ is the number of photons detected by the SPCM for 300 ns after optical pumping (3 $\mu$m later). The fluorescence as a function of time always starts with a peak and then decreases sharply for the first 500 ns, and more moderately for several $\mu$s afterwards. This response is responsible for the positive values of the normalized fluorescence in all our measurements. A similar fluorescence response is observed by other groups for single NVs, for example see [58]. (c) A typical Rabi curve for our sample when the magnetic field is constant ($B_z = 28.6$ G). The normalized fluorescence is calculated as explained in (b).
Fig. 5.7: (a) The sequence used to probe the changes of the total fluorescence from the diamond due to the changes of \(B_z\). The green laser was on during the whole measurement. (b) The fluorescence as a function of time for the sequence depicted in (a). The sequence starts at \(t = 5\, \mu s\) and ends at \(t = 15\, \mu s\). \(B = 0\) at \(t < 4\, \mu s, t > 16.5\, \mu s\) and at \(t = 10.1\, \mu s\). Since the magnetic field is aligned along the [-11-1] lattice axis it has an off-axis projection on the three other orientations. The off-axis component of the magnetic field mixes the \(m_s = 0\) and \(m_s = \pm 1\) levels and reduces the total fluorescence. (b) The ODMR spectrum for \(B_z\) (blue) and for \(-B_z\) (red). \(B_z\) is aligned along the [-11-1] axis and therefore its projection on the three other orientations is \(|B|/\sqrt{3}\). (c) The ODMR spectrum for \(B_y\). \(B_y\) is aligned along the [1-10] lattice vector. The generated ODMR is a \{110\}-like spectrum with two orientations shifted by \(\Delta \nu = \frac{1}{\sqrt{3}} g \mu_B B_z \mu_B\) and two other orientations almost not shifted.
5.3.1 Limitations of our measurements

Several limitations in our current setup (including the diamond) need to be considered when choosing the magnetic field frequencies and amplitudes to be used in our sequences. The lower limit of the frequency of the magnetic field change is set by $T_1$ which is typically several milliseconds for HPHT samples. To be faster than $T_1$ and to avoid long cycle times (since averaging many cycles is needed) we use frequencies higher than 50 kHz. Another limitation is set by $T_2^*$. In order to measure the electronic state of the NVs we apply a MW $\pi$ pulse. This pulse must be shorter than $T_2^*$ which (in our sample) is $< 0.5 \mu$s. A short $\pi$ pulse requires a strong MW field and such a field may excite other orientations (due to power broadening). In practice we use $\pi$ pulses shorter than 100 ns. To avoid excitation of other orientations, we use magnetic fields higher than 40 Gauss and have verified spectroscopically that the ODMR of the different transitions are completely separated. In the rotating-field experiments for which we need to apply a current to both wires, we are limited to frequencies lower than 150 kHz by our floating current driver (since the two wires are connected). This could be solved in the future by using a two-layer chip and two different current drivers.

5.4 Experimental results

5.4.1 Linear model

In this experiment, we change the magnetic field along a single direction so that the total field must go through zero. Our simulation (Fig. 5.2) shows that for a frequency change of $f=150$ KHz ($\omega = 2\pi f$) the system behavior changes as a function of $B_z^0$ from adiabatic to non-adiabatic. As can be seen, even at our maximal magnetic field of about 110 G, the non-adiabaticity is at the 50% level. The normalized fluorescence as a function of $B_z^0$ for experiments in which we change the magnetic field from $B_z = -B_z^0$ to $B_z = +B_z^0$ at $f=150$ kHz is depicted in Fig. 5.8. As a reference we show both the bright-contrast limit which is expected from full adiabaticity (where a continuous $2\pi$ pulse is applied) and the dark limit which is expected from full non-adiabaticity (where a single $\pi$ pulse is applied). It may be seen that, as expected, our signal (due to one $\pi$ pulse before the magnetic change and one after) shows less adiabaticity as the magnetic field is increased. In the following, we will use
the dark limit as a reference for all our results.

Fig. 5.8: (a) Experimental sequence for the linear case in which $B_y = 0$ and $B_z = B_{z0}\cos \omega t$. (b) In blue, the normalized fluorescence after a single $\pi$ pulse (dark state reference). The $\pi$ pulse was applied before applying changes to $B_z$. In this sequence $B_z = B_{z0}\cos \omega t$ where $\omega = 2\pi f$ and $f = 150$ kHz. In red, the normalized fluorescence after a $2\pi$ pulse (bright state reference), again before the magnetic field was changed. In black (our signal), the normalized fluorescence measured for the case in which one $\pi$ pulse is applied before changing the magnetic field and one $\pi$ pulse is applied after changing the magnetic field. As expected from Fig. 5.2 the level of adiabaticity goes down as the magnetic field becomes stronger. Explanation of how the normalized fluorescence is measured is given in Fig. 5.6b. The time separation between the two $\pi$ pulses is $20\mu$s.
5.4.2 Rotating-field model

The rotation of the magnetic field around the $x$ axis is done using $B_z = B_{z0}\cos\omega t$ and $B_y = B_{y0}\sin\omega t$. Theoretical expectations are given in Fig. 5.3. In order to better understand the behavior of the system under rotations we start by examining the case of a constant $B_z$ ($B_z = B_{z0}$) while changing only $B_y$ ($B_y = B_{y0}\sin\omega t$). The normalized fluorescence as a function of $B_{y0}$ for frequencies of 50 kHz, 75 kHz, 100 kHz, and 150 kHz is depicted in Fig. 5.9. We can see in Fig. 5.9 that for each frequency above a certain amplitude the fluorescence is reduced, indicating a non-adiabatic population change from $m_s = -1$ to $m_s = +1$. This reduction of the fluorescence seems to depend on the duration for which the magnetic field is pointing toward the direction transverse to the NV axis (i.e. towards $y$). This duration depends both on $B_{z0}$ and $B_{y0}$, and on the frequency of the alternating currents. This indicates that a magnetic field in the transverse direction harms the adiabaticity.

The next step is to measure the population when both $B_z$ and $B_y$ are changed so that the magnetic field does a full 180 degree rotation. In order to better observe the adiabatic to non-adiabatic transition, we scan as before the values of $B_{y0}$. Results for $B_{z0} = 49\, G$ and $B_{z0} = 109\, G$ for various frequencies are depicted in Fig. 5.10 and Fig. 5.11.

Finally, for the case in which the magnetic field along the NV $z$ axis never goes to zero (Fig. 5.9), the time for which the field is perpendicular to the NV axis, $t_T$, is calculated and presented in Fig. 5.12 (more precisely, we measure the time for which the magnetic field is oriented inside a 30 degree cone around the transverse axis). It seems to be experimentally determined that there is a universal time of $t_T = 4\, \mu s$ under which adiabaticity is maintained. Conversely, we present in Fig. 5.13 the case for which the magnetic field along the NV axis $z$ goes to zero (Fig. 5.11). Here there does not seem to be such a universal time, and the duration for which the field may point in the transverse direction, while adiabaticity is still maintained, is inversely proportional to the rate at which the field along $z$ changes when it is close to zero (i.e. $t_T$ is proportional to $1/\omega$). Indeed, this is the same behavior we observed in Fig. 5.8, where $\omega$ remained constant but the rate of change was determined by the value of $B_{z0}$. While these observations may be qualitatively intuitively understood, a detailed theory is required to explain them.
Fig. 5.9: (a) Experimental sequence for the partially rotating field case (the field vector never reaches the transverse direction): $B_z$ is constant during the sequence. A 4 $\mu$s 532 nm laser pulse is used to pump the NVs into $m_s = 0$; a MW $\pi$ (75 ns) pulse is then used to populate the $m_s = -1$ level; when the system is in $m_s = -1$ the transverse magnetic field $B_y = B_{y0} \sin \omega t$ is turned on; after $t = \frac{\pi}{\omega}$ another $\pi$ pulse is applied (with the same MW frequency). The time interval between the two $\pi$ pulses is 20 $\mu$s. A second 4 $\mu$s 532 nm laser pulse is used to probe the $m_s = -1$ population. (b-e) The normalized fluorescence as a function of $B_{y0}$ for $f=50$ kHz, $f=75$ kHz, $f=100$ kHz, and $f=150$ kHz. The normalized fluorescence for the sequence with two $\pi$ pulses is depicted in red. The normalized fluorescence for the case of only one MW $\pi$ pulse is depicted in blue for reference. It is easy to see that the higher the frequency, the larger $B_{y0}$ should be in order to transfer the population from $m_s = -1$ to $m_s = 1$, namely, in order to cross from the adiabatic regime to the non-adiabatic regime.
**Fig. 5.10:** (a) Experimental sequence for the fully rotating field case: $B_z = B_{z0}\cos(\omega t)$, $B_{z0} = 49 \text{ G}$. A 4 µs 532 nm laser pulse is used to pump the NVs into $m_s = 0$ a following MW π (70 ns) pulse is used to populate the $m_s = -1$ level. At $t < 0$ $B_z = B_{z0}$ and at $t > \frac{\pi}{\omega}$ $B_z = -B_{z0}$. Before $t = 0$ the system is pumped to $m_s = -1$. At $t = 0$ the axial magnetic field $B_z = B_{z0}\cos(\omega t)$ and the transverse magnetic field $B_y = B_{y0}\sin(\omega t)$ are turned on. After $t = \frac{\pi}{\omega}$ another π pulse is applied (with the same MW frequency). The time interval between the two π pulses is 20 µs. A second 4 µs 532 nm laser pulse is used to probe the $m_s = -1$ population. (b-d) The normalized fluorescence as a function of $B_{y0}$ for $f=50 \text{ kHz}$, $f=100 \text{ kHz}$, and $f=150 \text{ kHz}$. The normalized fluorescence for the sequence with two π pulses is depicted in red. The normalized fluorescence for the case of only one MW π pulse is depicted in blue for reference.
Fig. 5.11: Experimental sequence for the fully rotating field case (with a high transverse magnetic field): (a) Experimental sequence: $B_z = B_{z0}\cos(\omega t)$, $B_{z0} = 109$ G. A 4 $\mu$s 532 nm laser pulse is used to pump the NVs into $m_s = 0$, followed by a MW $\pi$ (70 ns) pulse used to populate the $m_s = -1$ level. At $t < t = 0$ $B_z = B_{z0}$ and at $t > \frac{\pi}{\omega}$ $B_z = -B_{z0}$. Before $t = 0$ the system is pumped to $m_s = -1$. At $t = 0$ the axial magnetic field $B_z = B_{z0}\cos(\omega t)$ and the transverse magnetic field $B_y = B_{y0}\sin(\omega t)$ are turned on. After $t = \frac{\pi}{\omega}$ another $\pi$ pulse is applied (with the same MW frequency). The time interval between the two $\pi$ pulses is 20 $\mu$s. A second 4 $\mu$s 532 nm laser pulse is used for probing the $m_s = -1$ population. (b-d) The normalized fluorescence as a function of $B_{y0}$ for $f=50$ kHz, $f=100$ kHz, and $f=150$ kHz. The normalized fluorescence for the sequence with two $\pi$ pulses is depicted in red. The normalized fluorescence for the case of only one MW $\pi$ pulse is depicted in blue for reference. Black arrows indicate the points where $B_{z0} = B_{y0}$ which according to Fig. 5.3 should be adiabatic (and indeed they are).
Fig. 5.12: Calculation of the time for which the magnetic field $\vec{B}$ is oriented at an angle less than $30^\circ$ from the $y$ axis as a function of $B_{y0}$ for the case in which $B_x = B_{y0}\sin\omega t$ and $B_z = B_{z0}$. The four curves are calculated for different frequencies where $\omega = 2\pi f$. It is easy to see that the higher the frequency, the shorter the time for which the total magnetic field is oriented by less than $30^\circ$ from the $y$ axis.

On each curve we mark the value of $B_{y0}$ for which the system shows a transition from adiabatic to non-adiabatic behavior. The markers are taken from Fig. 5.9 by taking the amplitude $B_{y0}$ for which the blue curve and the red curve join each other.
5.5 Conclusions and outlook

We qualitatively verified our simulations for adiabatic behavior in different regimes. It is quite remarkable, and beyond what is commonly expected, that even for full rotations of the magnetic field adiabaticity may still be maintained. More specifically, from Fig. 5.8 we see that the difference in normalized fluorescence between maximally adiabatic (red curve) and maximally non-adiabatic (blue curve) is about $3 \cdot 10^{-3}$. Indeed, this is what we find in the case of small rotations (Fig. 5.9) and also in the case of full rotations for high magnetic fields and frequencies (Fig. 5.11d). It thus seems that for certain parameters adiabaticity may be maintained.

In addition, as long as the magnetic field along the NV axis $z$ never goes to zero, we have shown that the transition to non-adiabaticity depends on the duration $t_T$ for which the magnetic field is transverse to the NV axis. There seems to be a universal time $t_T$ for which adiabaticity is maintained independent of the frequency of the field or its amplitude. When
the magnetic field along the NV axis z goes to zero adiabaticity may still be maintained but a
universal time is not apparent. While the results are consistent with our simulations and with our intuitive understanding, more complex theoretical models are required to explain these observations. In any case, these observations open the door to new and more detailed theoretical analyses which could explain the exact results and lead to better understanding of the underlying physics of the NV center.

These measurements, and specifically the robustness of adiabaticity under certain conditions, also open the road for levitation of NVs in magnetic traps similar to cold atom traps. Obviously, a detailed feasibility study is required in order to carefully simulate the trajectories in such a trap.

Finally, as an outlook let us also note that improvements remain to be done in the next generation of experiments. For example, in our current setup we used a 15 µm pinhole to filter out fluorescence from centers which are exposed to different MW and magnetic fields. The magnetic field gradients near the wire are extremely high and we hypothesize that this is the reason for the fact that in Fig. 5.8 our signal (black line) does not reach the red line for low magnetic fields, which represents the expected signal for full adiabaticity. In the near future we will check if a smaller pinhole improves the signal.

Unfortunately, as this was the last experiment done as part of this PhD project, there was not enough time to examine further experimental improvements, or to develop a detailed theoretical model.
In this chapter we introduce three protocols for magnetic sensing with improved sensitivity. Our first two methods deal with overcoming reduction of sensitivity caused by strain for small magnetic fields (for a previous suggested method to combat strain see [59]). The third method is based on improving the sensitivity by reducing the ODMR linewidth, achieved by probing only one orientation.

6.1 Orthogonal bias field

While HPHT samples have high concentrations of NV centers, these samples suffer from the effects of strong crystal strain [33]. This strain, which reduces sensitivity for low magnetic fields, increases with the NV concentration [57]. Here we describe methods to bypass the hindering effect of strain and thereby allowing for magnetic sensing at lower fields.

A common technique for measuring magnetic fields using NV centers is based on measuring the splitting between the \( m_s = \pm 1 \) resonant transitions from \( m_s = 0 \) (chapter 3). It is clear that the sensitivity of the system depends on \( \frac{\partial \nu_{\pm}}{\partial B} \) where \( \nu_{\pm} \) are the on resonance transition frequencies. By solving the Hamiltonian it is easy to see that the strain term \( E(S_x^2 - S_y^2) \) is responsible for reducing \( \frac{\partial \nu_{\pm}}{\partial B} \) for magnetic field magnitudes below the order of \( \frac{E}{\gamma g \mu_B} \).

A simulation of the transition energies as a function of the external magnetic field is depicted in Fig. 6.1.
Fig. 6.1: A simulation of the transition frequencies $v_+$ and $v_-$ from $m_s = 0$ to the $m_s = \pm 1$ states respectively, as a function of an external magnetic field (along the NV z axis) in the presence of strain corresponding to $E=4.5 \text{ MHz}$. Note that at small fields the Hamiltonian eigenstates are no longer the Zeeman eigenstates but rather mixed states (Eq. 2.8). In the inset we present the splitting $\Delta = v_+ - v_-$ as a function of the external magnetic field.

Straightforward solution for the presence of strain is to apply a bias field onto the sensor in order to move away from the region of low slope of the graph which in turn means low field sensitivity and resolution. Such a bias field will transfer the NV center energy levels from the mixed states, i.e. the $|\pm\rangle$ states (Eq. 2.8) to the Zeeman states with a well-defined spin projection along the NV axis. However in certain cases applying a bias field along the same axis of the detected signal is not possible. For example, when sensing superconducting phenomena the bias field is screened by the superconductor Meissner currents. It turns out that for \{100\} diamonds it is impossible to resolve only one NV orientation by applying a magnetic field in the diamond $xy$ plane (the plane perpendicular to the [100] axis). We therefore suggest the following protocol. The measured magnetic field is aligned along the [100] axis (which is the usual case when using \{100\} diamond, where the [100] is perpendicular to the large facet of the diamond plate and is typically considered the $z$ axis in the lab). We apply a bias field along the [011] axis. This bias field is orthogonal to both the [-11-1] and the [-1-11] orientations on the one hand (the convention of the vectors in this work is from N to V), and on the other hand has the same projection (up to a sign) on the [111] and [1-1-1] orientations (Fig. 6.2). Therefore the transition frequency from $v_+^{[1-1-1]}$ $m_s = 0$ to the $m_s = -1$ of the [1-1-1] orientation and the transition frequency $v_+^{[111]}$ from $m_s = 0$ to $m_s = 1$ of the
[111] orientation will be the same. Additional magnetic fields along the [100] axis will split
the resonance of the [111] and [1-1-1] orientations, since the projection of the [100] field onto
the two NV axes is aligned with the projection of the [011] field in the case of one NV and
opposite in the case of the other. The magnetic field is measured by converting the splitting
\( \Delta = \nu^{[111]}_+ - \nu^{[1-1-1]}_+ \) orientation into a magnetic field. A simulation of the transition energies
\( \nu^{[111]}_+ \) and \( \nu^{[1-1-1]}_+ \) as a function of an external magnetic field aligned along the [100] axis is
depicted in Fig. 6.3. A comparison of the quantity \( \frac{\partial \Delta}{\partial B_z} \) as a function of \( B_z \) for one orientation
and for the suggested method is depicted in Fig. 6.4.

Fig. 6.2: (a)-(d) The projections of a bias field aligned along the crystal [011] axis (gold arrow) upon the 4 NVs orientations. (a)-(b) The projection of the bias field on the [111] and [1-1-1] orientation is \( \frac{|B|}{\sqrt{3}} \). (c)-(d) The projection of the bias field on the [-1-11] and [-11-1] orientations is 0. (e) A schematic plot of all the four orientations, the bias field, and the field along the z axis (black arrow) which is the subject of the measurement. (the convention of the vectors in our plots and throughout this work is from N to V).
Fig. 6.3: A simulation of the transition energies $v_{[111]}^{[1]}$ from $m_s = 0$ to $m_s = +1$ and $v_{[1]}^{[1-1-1]}$ from $m_s = 0$ to $m_s = -1$ of the [1-1-1] orientation in the presence of an 8 G bias field along the [011] orientation as a function of the measured external magnetic field along the [100] lattice vector. The simulation assumes a strain field of $E = 4.5$ MHz. In the inset we present the splitting $\Delta = v_{[1]}^{[1-1-1]} - v_{z}^{[111]}$ as a function of the measured external magnetic field along the $z$ axis.

Fig. 6.4: The spectral splitting as a function of the external magnetic field along the $z$ axis. In blue, the splitting is between $m_s = +1$ and $m_s = -1$ of all four orientations without a bias field. The dashed black line shows, the splitting between the $m_s = -1$ of the $[1 \ 1 \ 1]$ orientation and the $m_s = +1$ of the $[111]$ orientation in the presence of a bias field of 8 Gauss along the [011] axis. Note that while the blue line (representing the sensor sensitivity) goes through zero at low fields, the black curve does not.
The method discussed in the preceding section involves the operation of a bias magnetic field. In some cases an added bias field may change the magnetic phenomena that we are trying to measure and is thus not desirable. For these cases we suggest the following protocol, which to the best of our knowledge is a completely novel idea.

The strain insensitivity regime is caused by the fact that although for high magnetic fields the g.s. triplet splits into $|0\rangle$, $|1\rangle$, and $|-1\rangle$ (the Zeeman states), for fields lower than the internal strain the g.s. splits into $|0\rangle$, $|+\rangle$, and $|-\rangle$, which are a superposition of the Zeeman states (see Eq. 2.8 and Fig. 6.1).

Transitions to the $|+\rangle$ and $|-\rangle$ are done by applying a MW pulse polarized either along the $x$ or $y$ axes (see Eq. 2.9 and text). On the other hand, the transitions from the $m_s = 0$ level to the Zeeman $|±1\rangle$ are done by applying a circularly-polarized MW pulse. In order to improve the sensitivity we will measure the values $|\langle -1|-\rangle|^2$ and $|\langle 1|+\rangle|^2$. The values of $|\langle -1|-\rangle|^2$ and $|\langle 1|+\rangle|^2$ as a function of the magnetic field are depicted in Fig. 6.5. To measure $|\langle 1|+\rangle|^2$ ($|\langle -1|-\rangle|^2$) we can use the following sequence:

a) We polarize the NVs into $m_s = 0$ using a 532 nm pulse.

b) We apply a MW $\pi$ pulse polarized along the $x$ ($y$) axis, which populates the $|+\rangle$ ($|-\rangle$) state.
c) Subsequent $\sigma^-$ ($\sigma^+$) MW $\pi$ pulse transfers the $|1\rangle$ ($|-1\rangle$) component of the $|+\rangle$ ($|-\rangle$) state to the $m_s = 0$ g.s. level.

d) Another green light pulse enables readout of the $m_s = 0$ population.

An improvement of the sensitivity could be achieved by measuring the quantity $\frac{|\langle 1|+\rangle|^2}{|\langle -1|-\rangle|^2}$. The ratio $\frac{|\langle 1|+\rangle|^2}{|\langle -1|-\rangle|^2}$ as a function of the magnetic field for the case $E = 4.5$ MHz is depicted in Fig. 6.6. A plot of the slope as a function of the strain for different strain fields is depicted in Fig. 6.7.

![Image](image.jpg)

**Fig. 6.6:** The ratio $\frac{|\langle 1|+\rangle|^2}{|\langle -1|-\rangle|^2}$. The values of $|\langle 1|+\rangle|^2$ and $|\langle -1|-\rangle|^2$ are taken from the simulation depicted in Fig. 6.5. By taking advantage of the sharper slope it is possible to achieve a better magnetic sensitivity [we remind the reader that the standard slope is zero at $B=0$ (Fig. 6.4)]. Note that while we change the direction of the $B_z$ field in this graph, we do not change the frame in which the polarization of the MW is determined. Namely as the $B$ changes sign, the MW radiation stays the same. .
Fig. 6.7: The slope $\partial \frac{|\langle 1^+ \rangle|^2}{|\langle -1^- \rangle|^2} / \partial B$ at $B=0$ for different strain fields is presented. The figure was generated by plotting the ratio $\frac{|\langle 1^+ \rangle|^2}{|\langle -1^- \rangle|^2}$ as a function of $B_z$ as was done in Fig. 6.6 and calculating the slope near $B=0$.

6.3 Orientation-selective ODMR

As was explained in section 3.2 one of the factors that influence the sensitivity is the linewidth of the ODMR signal. In this section we suggest a method to improve the sensitivity by reducing the spectral linewidth. At small magnetic fields, the four orientations are not well resolved from each other and, since they are exposed to different projections of the measured magnetic field, this induces broadening. In order to probe only one orientation we can either produce a diamond with a preferred orientation, which is not an easy task, or alternatively excite or probe only one orientation. In this section we introduce a protocol for probing the fluorescence change from a single orientation without using a bias field. As mentioned in section 2.5, the dipole interaction matrix element describing the probability of the NV center electronic excitation from the $^3A_2$ ground state to the $^3E$ excited state is nonzero only if the light is polarized perpendicular to the NV symmetry axis. In other words, the excitation probability depends on the angle between the NV symmetry axis and the polarization vector. By polarizing the light in a certain direction, we may selectively excite only certain NV orientations. This can potentially be applied to partially or fully lift the degeneracies of the ODMR spectral line even in the absence of an external magnetic field, which is usually used
to resolve the orientations. Since the matrix element that describes the absorption of a photon also describes the amplitude for emitting a photon, the symmetry selection rules are the same for excitation and emitted fluorescence.

The NV center has two allowed optical transitions \( \langle A_2 | \hat{x} | E_y \rangle \) and \( \langle A_2 | \hat{y} | E_x \rangle \). It can be excited to \( E_y \) (situated along \( y \)) by a linearly polarized light along the \( x \) axis or to \( E_x \) (situated along \( x \)) by a linearly polarized light along the \( y \) axis (\( x \) and \( y \) are in the plane perpendicular to the NV axis). A simple model for the excitation rate as a function of the laser polarization was given by Alegre [60] (this model included only the geometrical effects). The excitation rate thus depends on the projection of the light electric field onto the \( x \) and \( y \) axes. Choosing the \( y \) axis of the NV center so that in addition to being in the \( xy \) plane it is also in the light polarization plane (perpendicular to the light \( \hat{k} \) vector), the contribution of \( E_y \) to the excitation rate will be \( P_0 \sin^2 \phi \), where \( 90 - \phi \) is the angle between the \( y \) axis and the light electric field. The contribution of the second dipole moment, which is directed along the \( x \) axis, will be \( P_0 \cos^2 \theta \cos^2 \phi \) where \( \theta \) is the angle between the normal to the \( E_y \) dipole in the polarization plane and the \( E_x \) dipole. \( \theta \) is equal to the angle between the light \( \hat{k} \) vector and the NV \( z \) axis. (If \( E_y \) is perpendicular to \( E_x \) then \( E_x \) is also perpendicular to the projection of \( E_x \) on the polarization plane. Since \( E_x \) is perpendicular to the NV axis, \( 90 - \theta \) is the angle between the NV axis and the polarization plane. The \( \hat{k} \) vector is perpendicular to the polarization plane and hence the angle between the NV axis and the laser beam is also \( \theta \).) A simulation of the fluorescence as a function of the impinging light polarization is depicted in Fig. 6.8.

6.3.1 Sequence and setup

In order to study the NV polarization dependence we do the following experiment. We apply an arbitrary magnetic field onto \{100\} and \{111\} diamonds with a high density of NV centers. The orientation of the centers is then resolved by the ODMR. Next, a pulsed modulated MW signal, on resonance with the ground state transition of a specific orientation, is used to cause all the centers that belong to a specific orientation to fluoresce at the modulated frequency. This modulated signal is measured with a lock-in amplifier as a function of the green light’s polarization angle, yielding the polarization dependence of the specific orientation. In order to avoid changes in the fluorescence caused by the dichroic mirror, the diamond is ex-
Fig. 6.8: Simulation of the fluorescence as a function of the laser polarization for the four NVs orientations in {100} {110} and {111} diamonds (Alegre model). The laser beam $\hat{k}$ vector is aligned along the [100], [110], and [111] lattice axes respectively.

cited from one side and the fluorescence is collected from the other (Fig. 6.9a). An example of the fluorescence as a function of the modulated signal (in time) is depicted in Fig. 6.9b. In another experiment, in order to measure the fluorescence polarization, the diamond is illuminated with unpolarized green light and the emitted fluorescence is collected and transferred
through a $\lambda/2$ waveplate and a polarized beam splitter. The experimental setup is depicted in Fig. 6.10.
(a) Experimental setup for studying the polarization dependence of the NV centers. An arbitrary magnetic field $B_0$ is used to magnetically split the 4 NV orientations’ transition frequencies. Using a waveform function generator we modulate the MW signal. The MW frequency is on resonance with a transition for one particular orientation transition. As a result of the pulsed MW signal the specific orientation fluorescence starts flickering. The flickering frequency will be at the modulation frequency. The signal from the photodiode is monitored by a lock-in amplifier using the modulation frequency as a reference, which filters out the signal from the other three orientations. By changing the $\lambda/2$ waveplate angle before the beam goes into the diamond, the polarization dependence of the specific orientation is studied.

(b) Fluorescence when the NVs are exposed to a square-wave modulated MW radiation. The MW frequency is on resonance with one of the orientations.

Fig. 6.9

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Fig. 6.10: Experimental setup for measuring the emitted fluorescence polarization angle. Similar to what is done in the exciting beam polarization dependence measurements, we use a magnetic field to separate the ODMR spectrum. A pulsed modulated MW signal is used to modulate the fluorescence from a specific orientation. We filter out the signal of the three other NVs orientations using a lock-in amplifier. A $\lambda/2$ waveplate and a beam splitter are used for studying the emitted light polarization angle.

6.3.2 Results

We measured both the excitation rate due to a beam of polarized green light and the polarization of the emitted red light of \{111\} and \{100\} diamonds. The polarization dependencies of the \{111\} diamond are depicted in Fig. 6.11. In Fig. 6.11a we show the fluorescence as a function of the laser polarization angle. In Fig. 6.11b we show the fluorescence after a beam splitter, as a function of the polarization angle (by using a $\lambda/2$ waveplate positioned before the beam splitter). The polarization dependencies of the \{100\} diamond are depicted in Fig. 6.12.
(a) NVs in the \{111\} diamond are illuminated with a linearly polarized 532 nm beam. The red fluorescence intensity as a function of the exciting beam polarization angle is depicted. For comparison with the Alegre model, see Fig. 6.8c.

(b) A measurement of the emitted red light polarization angle for the four NVs orientations. The diamond is a \{111\} diamond. The emitted red beam $\vec{k}$ vector is perpendicular to the diamond surface and is aligned along the [111] lattice axis. The emitted red fluorescence is linearly polarized but its polarization angle is different for different orientations. The polarization angle was measured by putting a $\lambda/2$ waveplate and polarizing beam splitter before the photodiode.

Fig. 6.11
(a) The NVs in the \{100\} diamond were illuminated with a linearly polarized 532 nm beam. The red fluorescence intensity as a function of the exciting beam polarization angle is depicted. For comparison with the theoretical model see Fig. 6.8a.

(b) A measurement of the emitted red light polarization angle for the four NVs orientations. The NVs are hosted in a \{100\} diamond. The emitted red beam \( \hat{k} \) vector is perpendicular to the diamond surface and hence is aligned along the [100] lattice axis. The emitted red fluorescence is linearly polarized but its polarization angle is different for different orientations. The polarization angle was measured by putting a \( \lambda/2 \) waveplate and polarized beam splitter before the photodiode (Fig. 6.10).

*Fig. 6.12*
6.3.3 Isolating one orientation

In this subsection we suggest a method to measure the spectrum of a single orientation using the polarization selection rules, thus narrowing the linewidth at low magnetic fields, which in turn improves the sensitivity. As presented previously, we have experimentally verified the crucial building blocks of the suggested method. Consider the case of a \{110\} diamond which is cut along the planes perpendicular to [110], [001], and [-110] lattice vectors (E6110). As with any other diamond, the \{110\} diamond has the following four orientations \{111\}, \{1-1-1\}, \{-11-1\}, \{-1-11\}. A linear polarized laser beam with a \(\hat{k}\) vector aligned along the [-110] direction and an electric field oriented along the [111] direction will not excite the [111] orientation (Fig. 6.13c). The reason for not exciting the [111] orientation is that the electric field has no component in the plane perpendicular to the [111] direction; in terms of the model \(\phi = 0^\circ\) and \(\theta = 90^\circ\). (Note that these are not the angles of the polarization in the lab frame which are used in Fig. 6.8 and Fig. 6.13.). In our case (a diamond which is cut from one side along the plane perpendicular to the [-110] lattice vector) these requirement can easily be met by illuminating the diamond with the laser beam along the [-110] lattice vector (Fig. 6.14a). The projection of the field on the three other orientations will result in a relative excitation probability of \(P_0(\cos^2\theta\cos^2\phi + \sin^2\phi) = 0.89P_0\).

The aim of our protocol is to measure the signal only from the NV\(_4\) = [−1−11] orientation (\(\theta = 90^\circ, \phi = 70.5^\circ\)). The same calculation can be done for the other two orientations (\(\theta = 35.26^\circ, \phi = -54.53^\circ; \theta = 144.73^\circ, \phi = -54.53^\circ\)) and one finds that the relative excitation probability is the same, and so we will have to suppress the other two orientations later on by other means. We choose the \(y\) axis of the NV to be in the polarization plane, hence \(\hat{y} = \hat{k} \times \hat{N}V_4 = [1 1 2]/\sqrt{6}\). As we defined in the beginning of this chapter, \(90 - \phi\) is the angle between \(y\) and the electric field, which is along the [111]/\(\sqrt{3}\) lattice vector. Taking the scalar product \(\hat{y} \cdot [111]/\sqrt{3}\) and the inverse cosine we get \(\phi = 70.5^\circ\). \(\theta\) is defined as the angle between the normal to \(y\) in the polarization plane and \(x\) in the NV \(xy\) plane, which as noted is simply the angle between the light \(\hat{k}\) vector and the NV axis. Since the NV orientation [-1-11] is orthogonal to \(\hat{k} = [-110]\) we get that \(\theta = 90^\circ\). By substituting \(\theta\) and \(\phi\) into the Alegre model we get \(P_0(\cos^290\cos^270.5 + \sin^270.5) = 0.89P_0\).

Next, in order to filter out the fluorescence from the [1-1-1] orientation we will col-
lect only the fluorescence which is polarized along the [1-1-1] direction (Fig. 6.14b). Since the photons emitted by the [1-1-1] orientations cannot be polarized along the [1-1-1] lattice vector, only fluorescence from the [-1-11] and [-11-1] orientations will be collected. The distribution of the fluorescence for the four orientations as a function of their polarization is depicted in Fig. 6.13d. The direction of the polarization angle in the lab frame is depicted in Fig. 6.13d.

We suppress the excitation of the [-11-1] orientation by choosing the MW field direction. In order to excite an NV center, a MW field with a magnetic field in the plane perpendicular to the NV axis is needed. In order to prevent the [-11-1] orientation from interacting with the MW field, it is possible to use a MW field with its magnetic field aligned along the [-11-1] axis. Since for a \{110\} diamond the [-11-1] lattice vector is located parallel to diamond surface, a MW B-field parallel to the surface needs to be generated. It is easy to generate such a field by locating the diamond above a MW wire. Above the center of the wire the field will be parallel to the diamond surface, and by putting the wire perpendicular to the [-11-1] orientation, its magnetic field will be aligned along the [-11-1] orientation (Fig. 6.14c). Such a field will not excite the [-11-1] orientation. By calculating the projection of this field onto the plane perpendicular to the [-1-11] orientation and taking its power to be $P_{MW}$, we find that a power of 0.89 $P_{MW}$ contributes directly to the excitation of the [-11-1] orientation.

To conclude, by choosing the impinging light polarization we can filter out the [111] orientation, by choosing the collected fluorescence polarization we can filter out the [1-1-1] orientation, and by choosing the MW polarization direction we can prevent the [-11-1] orientation from contributing to the signal. The ODMR signal will thus be generated only by the [-1-11] orientation.
Fig. 6.13: (a) Simulation for a \{110\} diamond of the fluorescence as a function of the laser polarization for the case of a laser beam with a \(\hat{k}\) vector aligned along the [-110] lattice vector (perpendicular to the small diamond plate facet). (b) Simulation of the fluorescence polarization angle for the case of an outgoing beam with its \(\hat{k}\) vector aligned along the [110] lattice vector (perpendicular to the large facet of the diamond). (c) An illustration of the case where the \(\hat{k}\) vector of the impinging beam is aligned along the [-110] lattice vector and polarization angle of the impinging beam is 125.25°. The polarization angle in the simulation is measured from the projection of the [-11-1] orientation (dashed line) onto the plane perpendicular to the \(\hat{k}\) vector (the plane is in green and the \(\hat{k}\) vector is perpendicular to the paper). This configuration enables excitation of the [1-1-1], [11-1], and [-1-11] orientations without exciting the [111] orientation. The black arrow is aligned along the [110] lattice vector which is perpendicular to the large diamond plate facet from which we collect the fluorescence. (d) An illustration of the case considered for filtering out the fluorescence from the [1-1-1] orientation. The polarization angle in the simulation is measured from the projection of the [11-1] orientation (dashed line) onto the plane perpendicular to the \(\hat{k}\) vector (the plane is in red and the \(\hat{k}\) vector is perpendicular to the page).
Fig. 6.14: A scheme for a single orientation ODMR measurement of a \{110\} diamond. (a) The laser beam is oriented along the [-110] lattice vector. By tuning the polarization so that the electric field is aligned along the [111] lattice vector (the polarization angle is 125.25° in terms of Fig. 6.13a) the [111] orientation will not interact with the exciting laser beam. (b) The optical axis of the fluorescence detecting apparatus is aligned along the [110] lattice vector. The [-1-1-1] NV orientation cant emit photons with electric field aligned along the [-1-1-1] orientation. Filtering of the [-1-1-1] fluorescence is possible by using a PBS and a $\lambda/2$ plate. (c) The MW antenna for exciting the NV centers (not to scale). A nearby MW wire positioned under the plane perpendicular to the [-11-1] orientation. By orienting the MW wire antenna perpendicular to the [-11-1] lattice vector the emitted magnetic field will be aligned along the [110] axis can produce magnetic fields parallel to the diamond face. Since the MW field will have no components in the plane perpendicular to the [-11-1] orientation it will not interact with the NVs g.s. levels. (d) The proposed setup. The laser beam hits the diamond along the [-110] lattice vector which is (in our case) the lab x axis. A $\lambda/2$ plate is used for aligning the electric field along the [111] lattice vector. The MW wire antenna is positioned under the diamond. The fluorescence is collected using an objective with its optical axes aligned along the [110] lattice vector. A $\lambda/2$ plate and a PBS is used for filtering out the fluorescence from the [1-1-1] orientation.
7. CHARACTERIZING A HIGH-\( T_c \) SUPERCONDUCTOR WITH AN NV CENTER-BASED MAGNETOMETER

The material presented in this chapter is a result of joint work with Dr. Amir Waxman and appears in a different form also in his PhD thesis. The findings were published in [A. Waxman, Y. Schlussel, D. Groswasser, V. M. Acosta, L.-S. Bouchard, D. Budker, and R. Folman, Diamond magnetometry of superconducting thin films, Phys. Rev. B 89, 054509 (2014)].

High-\( T_c \) superconductors are the subject of intensive research due to a variety of applications such as high-speed trains or filters for cellular transmission stations. Using NV centers in diamond to study these superconductors has several advantages. Contrary to superconducting quantum interference devices (SQUIDs) that measure changes of the magnetic field, an NV center based magnetometer measures the absolute magnetic field. In addition, the acquisition time for a magnetic map with an NV magnetometer is expected to be much faster than those of SQUIDs so that SC dynamics may be studied. Another reason for using NVs is that high resolution SQUIDs are made of low-\( T_c \) superconductors and hence are not suitable for measurements of high-\( T_c \) superconductors. The NV sensor may be operated over a wide range of temperatures and does not demand cooling to liquid helium temperatures. In this chapter we demonstrate the use of the NV center for probing the phase transition of a high-\( T_c \) thin-film superconductor.

Superconductors are divided into type-I superconductors and type-II superconductors. Type-I superconductors include all superconducting elements except niobium. Niobium, superconducting alloys, and chemical compounds are type-II superconductors. The high-\( T_c \) superconductors also belong to this group. The main difference between the two groups is the different response to an external magnetic field. For type-II superconductors, the energy of an interface between a normal and a superconducting region is \( \sigma_{ns} < 0 \) [61]. This implies that under certain circumstances, it is energetically favorable for these materials, when
placed in an external magnetic field, to become subdivided into alternating normal and superconducting domains. As long as the external magnetic field is $H_0 < H_{c1}$, where $H_{c1}$ is the first critical field, the average field in the interior of the sample is $B = 0$. When the magnetic field is larger such that $H_{c1} < H_0 < H_{c2}$, the field penetrates the superconductor. At a certain critical field $H_0 = H_{c2}$, the average field in the interior becomes equal to $H_0$ and the bulk superconductivity disappears. Above $H_{c1}$ the magnetic field penetrates into the material in a very special way as quantized vortices. Each vortex has a normal core which can be approximated by a long thin cylinder with its axis parallel to the external magnetic field. The vortex supercurrent circulates within an area of radius $\lambda$, the penetration depth. $\lambda$ is different for different superconductors and in general is a function of temperature in the case of a thin film it is also a function of the film thickness. Typical values for $\lambda$ are 46 nm for Nb and 170 nm for YBa$_2$Cu$_3$O$_7$ (YBCO). Once vortices are generated they may interact with many types of defects. Since regenerating the magnetic field inside the normal core requires energy, it is energetically preferred to locate a vortex above a defect (a pinning center). When a type-II superconductor is placed in a magnetic field higher than $H_{c1}$, vortices penetrate into the sample and form a lattice of vortices. If we now reduce the magnetic field, or alternatively decrease the temperature, the superconductor ”pushes” the vortices towards the edges of the sample. However, if there are pinning centers in the sample, several vortices will remain pinned to the pinning centers. The average magnetic field generated by the vortices is $B = n \cdot \Phi_0$, where $n$ is the average vortex density and $\Phi_0$ is the magnetic flux quantum $\Phi_0 = h/2e = 20.7 \mu m^2 \cdot G$.

7.1 Setup

Confocal microscopy is performed by excitation with green light (532 nm) supplied by a diode-pumped solid-state laser. The output beam is expanded to a diameter of 1 cm, larger than the diameter of the objective lens. This enables us to scan the beam across the sample by moving the objective, without affecting the beam direction. The objective (Olympus, Pro Plan) has a numerical aperture N.A.=0.6 and a maximum working distance of 4.2 mm. The laser light power at the NV location is measured to be 10 mW. The fluorescence emitted by the NV centers is collected with the same objective used to focus the incident green light, and is transmitted through two dichroic mirrors, both mounted in cubes to facilitate optical
alignment. These cubes are mounted on a three-dimensional (3D) translation stage which is controlled by actuators. A lens is used to focus the fluorescence onto a high-sensitivity photodiode (NewFocus 2151). The superconductor sample is mounted on the copper cold finger of a cryostat (Janis model ST-500). To promote good thermal contact, we use a Teflon holder which is secured by screws and presses the superconductor sample against the cold finger. Our superconducting sample is a 300 nm YBCO layer (obtained from Ceraco), which was deposited on a MgO substrate ($5 \times 5 \times 0.5$ mm$^3$). The c axis of the superconductor is perpendicular to the plane of the film, so that the CuO$_2$ planes are parallel to the surface. Transmission of the microwaves to the diamond is achieved with two copper strips placed along either side of the diamond on top of the MgO substrate and the YBCO layer. The microwave signal is produced by a Keysight signal generator (Model E4426B ESG-AP) and amplified by a Mini-Circuit power amplifier (Mini-Circuits ZHL-16W-43+).

![Experimental setup](image)

*Fig. 7.1:* Experimental setup: The superconducting sample is mounted inside a cryostat (Janis ST-500). The MW is injected through an SMA feedthrough into the cryostat. A dichroic mirror (Semrock) is used to filter out the exciting beam reflections from the NVs fluorescence. An air objective with a working distance of 4.2 mm and NA of 0.6 (Olympus) is used to focus the exciting beam and to collect the fluorescence. The distance between the NV sensor and the outer surface of the cryostat window is 2.3 mm. The diamond is pressed onto the YBCO sample with a Teflon piece held by screws.
7.2 Detection with a lock-in amplifier using frequency modulation

For technical reasons the diamond in this experiment was an E6 HPHT \{110\} with a 10 nm NV layer. Most importantly, a very thin NV layer close to the diamond surface enables high spatial resolution, and in addition, readout from a highly localized NV ensemble enabling high field resolution. In order to filter out the relatively high background noise from the low fluorescence changes we use an avalanche photodiode (APD), frequency modulated MW, and a lock-in amplifier. The signal measured by the APD is a function of the MW frequency \( V = V_s(\omega) \). The frequency modulated MW signal is \( \omega(t) = \omega_0 + \Omega_0 \cos \Omega t \) where \( \Omega \) is the modulation frequency. Using Taylor-series expansion we can write: \( V_s(\omega) = V_s(\omega_0) + \frac{dV_s}{d\omega}\bigg|_{\omega=\omega_0}(\Omega_0 \cos \Omega t) + \ldots \). By taking the lock-in reference frequency to be \( \Omega \) the lock-in signal will be proportional to the derivative of the ODMR spectrum. Thus, when the MW frequency \( \omega \) will be on resonance with one of the transitions the lock-in signal will be 0. The frequency modulation (FM) depth is usually tuned to be on the same order as the ODMR resonance linewidth (6 MHz). The modulation frequency was tuned to 500 Hz. The magnetic field can be determined from \( B = \frac{\Delta \nu}{2 g_s \mu_B} \) where \( \Delta \nu = v_+ - v_- \) and \( v_\pm \) are the transitions frequencies to the \( m_s = \pm 1 \) levels where the lock-in signal is 0.

7.3 Sequence and Results

We use two sequences to characterize our YBCO thin layer. In the first sequence we cooled the sample at zero field (ZFC) down to 65 K and start increasing \( B_z \) in increments of 4.5 G (corresponding to increments of 1 A in the bias coil). The ODMR at each point was recorded. After crossing the penetration field \( H_p \) (above which vortices enter the sample), we decreased the current back to zero. The Zeeman splitting (proportional to the magnetic field) is depicted in Fig. 7.2 as a function of the current in the coil. The hysteresis in the curve is a clear hallmark of type-II superconductivity and the presence of pinning centers in the sample. In the second sequence, the thin-film superconductor was again cooled at zero field and then an external magnetic field was applied. The magnetic field was then held constant and the temperature was raised. After reaching \( T_c \) the temperature was decreased resulting again a typical Type-II hysteresis curve. The signal at each temperature was analyzed by fitting to the
derivative of the sum of $N$ Lorentzians ($N$ being the number of resonances in the spectrum).
The Zeeman splitting as a function of temperature is depicted in Fig. 7.3. The blue dots in the experimental data in Fig. 7.3 were fit to a sigmoid function 
\[ \Delta Z(T) = \frac{a}{1+\exp\left[-\frac{T-T_c}{\Delta T_c}\right]} + b \]
where $a$, $b$, $T_c$, and $\Delta T_c$ are fitting parameters. In our measurements we found $T_c = 70$ K. The factory-stated $T_c$ of the sample is higher than 81 K. In these experiments the temperature sensor was 4 cm from the YBCO thin layer, and this most likely explains the discrepancy, since the MW and laser beam generate a temperature gradient and hence calibration is needed for estimating the actual temperature.

**Fig. 7.2:** (a-b) ODMR spectra taken during the experimental sequence for finding the local penetration field $H_P$. The two different signals, taken with the same external field, reflect different responses of the material: (a) taken after increasing the field from zero, meaning the system at this spatial point is in the Meissner state, and the external field is screened; (b) recorded after lowering the field from 50 G, which is above $H_P$, thus measuring the field of the pinned vortices; (c) the Zeeman splitting detected with the NV-diamond sensor during the measurement sequence. The applied magnetic field is proportional to the coil current. The plot demonstrates the local transition from the Meissner state to an intermediate state wherein vortices are in the sample. The blue points correspond to increasing coil current while the red points correspond to the decreasing-current sequence. The blue and the red lines are to guide the eye. Figure taken from [36].
Fig. 7.3: (a-c) Several ODMR signals taken at different temperatures with an applied external magnetic field of 15 G. The dashed black lines denote the Zeeman splitting for each case. The up/down arrows indicate if the temperature was raised or lowered. (a) An ODMR signal taken below $T_c$, at 62 K. The system is in the Meissner state, and the external field does not penetrate the superconductor layer. (b) The signal at the phase transition. Here, we see a partial penetration of the field (a Zeeman splitting of 42 MHz corresponds to a 10 G field in the z direction). (c) This signal, at 74 K, indicates that the system is no longer in the superconducting phase. In this graph, we also demonstrate the fit to a derivative of a Lorentzian function, used to determine the zero crossing of the error signal. We use this fit on all ODMR signals to find the value of the Zeeman splitting. (d) The signal taken after reducing the temperature again, without turning the external field off. Since the critical field at $T_c$ is zero, vortices penetrate the layer, leading to the average magnetic field which we measure. (e) At 60 K the magnetic field was turned off. Defects in the superconductor layer lead to flux pinning, evidenced by the field measured by the NV centers. (f) The phase-transition curve of the superconductor layer. Plotted is the Zeeman splitting between the $m_s = 0 \rightarrow m_s = -1$ and the $m_s = 0 \rightarrow m_s = 1$ resonances of the $35^\circ$ NV-axis alignments. The blue data points belong to the ascending temperature sequence, while the red ones belong to the descending sequence. The external field during the measurements is 15 G except for the last measurement (at T = 60 K), where the field was turned off. We fit the blue data points to a sigmoid function (blue line), and extract a critical temperature of $T_c = 70.0 \pm 0.2$ K. The red line is to guide the eye. Figure taken from [36].
7.4 Conclusions

In these experiments we showed that a magnetic NV sensor is capable of sensing local superconducting phase transitions. Using FM modulation improved our sensitivity and enabled the measurement with a 10 nm layer of NV centers in an HPHT diamond. The $T_c$ that was determined may require a more careful temperature calibration due to a gradient between the cold finger and the sample. From the trapped magnetic field we can infer that there are pinning centers in the sample. It is possible to calculate the local density of vortices in the sample. This information is important for further tasks such as vortex imaging.
8. SPATIAL MAPS OF THE MAGNETIC FIELD

The results of the previous chapter, in which we probed a superconducting surface, were obtained with a photodiode, i.e., equivalent to a single-pixel CCD. The next step was to develop a setup with a multi-pixel CCD so that maps of the magnetic field in a plane just above a surface may be obtained in a single shot without the need for 2D spatial scanning. While we were successful in taking images of simple room-temperature structures, we had numerous technical difficulties in obtaining such images from superconducting surfaces as explained in the following.

8.1 Setup

Our wide-field imaging setup was designed for probing high-$T_c$ superconductivity phenomena and therefore the sample and the diamonds are positioned inside an optical cryostat (Janis ST-500). For our measurements we are interested in imaging an area of $200\,\mu\text{m} \times 200\,\mu\text{m}$. For wide-field imaging (as shown in Fig. 8.1a), we use a collimated green light beam rather than a focused one. The collimation of the exciting beam is done by focusing it into the objective lens. Since the saturation intensity of the NV center is $300\,\text{kW/cm}^2$ [64], working at the saturation intensity requires a beam of $120\,\text{W}$. Since the cooling power of the cryostat is less than $1\,\text{W}$ and the thermal contact between the sample and the cold finger is not perfect, working with a high-power laser beam is impractical. To maximize the fluorescence we use HPHT diamonds with NV concentrations higher than 1 ppm, e.g. DDK111, DDK100B, and DDK100A. We use an Andor-iXon electron-multiplying CCD (EMCCD) for probing the relatively low fluorescence. The pixel size of this EMCCD is $16\,\mu\text{m} \times 16\,\mu\text{m}$. Our objective (Olympus LUCPLFLN40XRC) has a magnification (using a 180 mm tube lens) of $\times40$ with an NA of 0.6. The same tube lens which is used to magnify the emitted fluorescence is used as the EMCCD focusing lens. Since $16\,\mu\text{m}/40 = 0.4\,\mu\text{m}$ each pixel on the EMCCD
probes an area of $0.4 \mu m \times 0.4 \mu m$, where we verified this relation using a resolution target (US1951). The MW radiation is transmitted to the diamond using a homemade dipole antenna positioned on the cryostat cover. This method does not require wiring inside the cryostat and therefore reduces the heat transfer caused by the MW power. Nevertheless, MW power greater than 1 W heats the cold finger and prohibits cryogenic measurements.

![Wide-field imaging optical setup](image1.png)

**Fig. 8.1**: (a) Wide-field imaging optical setup. Wide-field imaging requires the illumination of a relatively large area with the exciting beam laser. The green light is collimated using a 150 mm focusing lens and must be adjusted so that its focal point overlaps with the focal point of the objective. The magnification of the system is $\times 40$. (b) Our homemade cryostat window. We drilled feedthrough holes in a 2 inch optical window. After soldering an SMA connector to a homemade dipole antenna the holes were sealed using epoxy glue. An objective lens turret is used to switch between a $\times 4$ objective for adjustments and a $\times 40$ objective for imaging. (c) An image of our setup including the cryostat and the Andor IXON EMCCD camera. The cryostat is positioned inside two sets of Helmholtz coils. The outer coils are used to compensate the earth’s magnetic field while the inner coils are used to generate bias fields in different directions.

### 8.1.1 ODMR measurement sequence

In order to measure the magnetic field in the vicinity of the NV centers we use the following sequence. We scan over different frequencies and for each frequency we take 2 shots. The
first shot with MW is our signal while the second shot without MW is our reference. The ODMR curve is then generated from the normalized contrast for each pixel. From the ODMR curves we should be able to retrieve magnetic maps of the imaged area. An example of a magnetic map of the field generated by a 120 mA current in a 40\(\mu\)m diameter wire imaged using this method is depicted in Fig. 8.2. For this kind of measurement we use a 32 G bias field along the z axis (vertical axis, perpendicular to the surface) which increases the system’s sensitivity and simplifies the procedure of converting the ODMR spectra into magnetic maps. (Since superconductors screen external magnetic fields and since a bias field may change the phenomena which we are trying to observe, in the SC measurements described in the following we attempt to image the magnetic fields without a bias field).
Fig. 8.2: (a) Our test wire, a 40 \( \mu m \) wire positioned under an 80 \( \mu m \) \{111\} diamond with a 100 nm layer of NV centers. (b) An optical view of the wire through our imaging system. (c) An experimentally obtained 2D magnetic map of the \( z \) component of the magnetic field above a 40 \( \mu m \) wire with a current of 120 mA. (d) A simulation of the magnetic field 1 \( \mu m \) above a 40 \( \mu m \) wire with a current of 120 mA.

### 8.2 Theoretical background and motivation

The numerous applications of thin film type-II superconductors have caused much interest in the modeling of their nonlocal electrodynamics and hysteretic response to external magnetic fields and applied transport currents. Analytical solutions are known for the Bean critical
state model [62] in the simplest geometrical configurations, such as a thin disk in an external field or an infinite strip. Numerical methods for modeling magnetization of flat films of arbitrary shapes have also been developed. In the following we try to use our 2D NV-based magnetic imaging setup as a simple method for validating the predictions of such simulations. More specifically we try to compare our results to the predications of Barrett et al. [63]. The importance of this model is that unlike other models it simulates not only the magnetic field but also the electric field in a meander-shaped superconductor. Since NV magnetometers can in principle measure electric fields [15] we tried to compare our measurements to this specific model with the hope that eventually we will also be able to compare electric field measurements to the theoretical model. The predicted current distribution and magnetic field distribution are depicted in Figs. 8.3a and 8.3b. To better understand the differences between normal and supercurrent distributions we present simulations for the magnetic fields of a straight superconducting wire and a straight copper wire in Fig. 8.4.

Fig. 8.3: Simulations of the transport current and resulting magnetic field in a meander-shaped film: (a) the current stream lines. The current density near the inner corners is much higher than the current density near the outer corner. (b) The component of the magnetic field normal to the film. The blue lines indicate the film boundary. Figure taken from [63].
Fig. 8.4: Simulations of the magnetic fields in the vicinity of a 300 nm thick × 150 µm wide wire carrying a 0.4 A current. (a) $B_z$ above a superconducting thin film wire. The current in the superconducting wire flows near the wire edges resulting high magnetic fields near the superconducting edge and zero field inside the superconductor. (b) The magnetic field in the x direction above a superconducting wire. Unlike the field in the z direction the magnetic field along the x direction is expected to be nonzero inside the superconductor. The magnetic field distribution along the y axis is expected to be zero. (c-d) For comparison we present simulations of the magnetic field distributions along the z axis (c) and along the x axis (d) above a copper wire carrying a current of 0.4 A.
8.3 Sequence

For testing the theoretical model we used a 250 nm thin-film YBCO sample positioned on a 500 µm thick sapphire substrate (Ceraco). In order to achieve good electrical contact with the YBCO strip, a 120 nm gold layer was evaporated onto the YBCO immediately after growing the YBCO layer. A 150 µm strip with a 90° corner was then etched in a lithographic process at BGU (Fig. 8.5a). Helmholtz compensation coils are used for cancelling the earth’s magnetic field. The sample was cooled down at zero magnetic field to ≈ 30K. A 0.4 A current was then injected into the superconducting strip. Let us note two important points. First, when the YBCO strip becomes superconducting it behaves like an antenna. In some places the MW field strength is considerably amplified and hence a reduction of the MW field was needed. Second, to estimate the spatial resolution with which we can image vortices and to estimate the required field sensitivity to image vortices, one is required to estimate the distance of the NV center from the YBCO. For this purpose we fabricated a 3 µm gold wire near the YBCO (Fig. 8.5b). By measuring the magnetic field induced by the current in the gold wire near the NV centers and comparing it to magnetic simulations, we can approximate the distance between the NV centers and the YBCO layer. Typically this distance was found to be ≈ 2 µm. Since both surfaces are expected to be flat, and since both surfaces were cleaned, this gap may be due to a curvature in the samples. Because our main goal of observing vortices is not achievable with such a gap, we have postponed further work on this project until this issue is resolved. While we could not use a superconducting thin film evaporated on the diamond, such as niobium, because our cryostat cannot deliver a low enough temperature, we are working together with our Mainz partners (the Budker group) on a new method in which 1 µm-thick diamonds are placed on the YBCO and are expected to closely follow the local topography so that the gap will become much smaller. Although work on this project was postponed 1, in the following we present preliminary results that have been obtained.

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1 Few weeks after the submission of this thesis using the same experimental setup and this new method we were able to image vortices in YBCO thin-films. Images of superconducting vortices that were measured with this system are presented in Appendix B.
Fig. 8.5: Our chip: (a) contacts a and b are used for injecting current into the YBCO thin-film wire. Contacts c and d are for measuring the critical current of the wire. (b) For measuring the distance of the NV centers from the diamond, we fabricated a 3 µm gold wire near the YBCO. By simulating the magnetic field from the gold wire and comparing it to the measured influence of the currents on the NV spectrum, we were able to estimate the distance of the NV centers from the YBCO layer. In order to fabricate a wire which is as narrow as possible (a narrow wire is important for a better field gradient along the z axis enabling an enhanced spatial resolution in measuring the distance between the NVs and the wire), we fabricated several wires with different widths. From the wires that were successfully fabricated we chose the narrowest one (3 µm) and cut all the other wires with a laser.

8.4 Preliminary results

The ODMR spectra for different locations above the superconducting corner are presented in Fig. 8.6. Let us first consider Fig. 8.6g. As can be seen from Fig. 8.6e, point g lies outside the superconductor. The magnetic field outside the YBCO is expected to be along the z axis (Fig. 8.4). Since the diamond in use is {111} (with one orientation aligned along the z axis and three other orientations having an angle of 70.5° from the z axis) we expect the spectrum to have 4 resonances. The two outer resonances belong to the [111] orientation while the inner resonances belong to the three other orientations. Such a spectrum is indeed depicted in Fig. 8.6g. A parameter we use in order to understand the magnetic field from the ODMR spectrum is the depth of the on-resonance spectroscopic dips. Since the optical transitions require an electrical field perpendicular to the NV axis, most of the photons are emitted along the NV axis. In the case of a {111} diamond the [111] orientation emits photons which are perpendicular to the diamond facet and which consequently easily exit the diamond, while photons from the three other orientations are more easily undergo internal reflection from the diamond surface due to the diamond’s high index of refraction. The fluorescence from the
Fig. 8.6: ODMR spectra for different locations above a 90° corner in a superconducting thin-film wire with a current of 0.4 A. The thickness of the thin film is 250 nm and its width is 150 µm. An optical image of the corner is depicted in (e). The picture is generated by summing over the normalized fluorescence at different frequencies, a method that is used to generate Fig. 8.7 as well. This technique is explained in detail in the text. The color code can be interpreted as an indicator of the magnetic field where blue indicates high field and green–yellow indicates a lower field. The green squares indicate the locations at which the ODMR of figures (a–d) and (f–i) are taken. Each ODMR is collected from a volume of $1.2\mu m \times 1.2\mu m \times 100nm$. The ODMR spectra in (c) and (g) are taken from locations outside the superconducting wire and are therefore colored in blue. For a discussion of the spectra see the text.

[111] orientation is therefore higher, and the dips in the ODMR spectra due to resonances with the [111] orientation transitions are usually deeper. For example, in Fig. 8.6g, the inner
dips have essentially the same depth as the external ones, but since they are composed of three contributions, we may estimate that each contribution is much weaker relative to the single contribution of the outer dip. In Fig. 8.6g we also see a strong asymmetry between the left dips and the right dips. This is probably due to the MW polarization which is closer to \( \sigma^+ \) than to \( \sigma^- \). Contrasting with Fig. 8.6g, the spectrum in Fig. 8.6c, which is also outside the superconductor, indicates a very low magnetic field along the \( z \) axis. This difference between the magnetic field inside the corner and outside the corner is in accordance with the Baret et al. simulation (Fig. 8.3).

Consider now the spectrum depicted in Fig. 8.6d: we know that the component of the magnetic field along the \( z \) axis must result in a splitting of the [111] orientation. Since the two deeper dips are not split by more than 17 MHz we may estimate that \( B_z < 3 \text{ G} \) (since the local strain was not measured we cannot be more precise). After estimating the magnetic field along the \( z \) axis to be relatively low, by observing the splitting of the side dips we can conclude that the projection of the magnetic field in the \( xy \) plane is about 24 G (taken form the observed 135 MHz splitting). Let us note that the two missing dips of the fourth orientation most probably overlap two of the observed dips (e.g., if the magnetic field is mainly in the \( xy \) plane and it is close to perpendicular to one of the three non-[111] orientations, then this orientation will overlap the dip of the [111] orientation). Fitting the spectrum correctly to eight dips requires either prior knowledge of the magnetic field direction or attributing each dip to a specific orientation. Attributing each dip to a specific orientation could be done by checking the polarization dependencies of the dips as illustrated in chapter 6.3.

A superconducting current in a straight thin-film superconductor in which \( d \geq \lambda \) (where \( d \) is the thin film thickness and \( \lambda \) is the penetration depth) is expected to flow near the superconductor edges [61]. Such a current density distribution is expected to generate high magnetic fields perpendicular to the current direction (Fig. 8.4a). Indeed the broadening of the spectrum observed in Fig. 8.7d relative to the spectrum in Fig. 8.6f, and the splitting of the spectrum observed in Fig. 8.6a, show high fields near the edges of the superconductor. These high fields point to a high current density near the edges of the superconductor (aside from the outer corner – as predicted in Fig. 8.3).

Although extracting detailed magnetic maps requires more measurements (with different
MW polarizations) and a significant computational effort, we can draw a qualitative picture of the current density using a method similar to the “iso-magnetic field” method. In the regular iso-magnetic field method, the MW frequency is held constant and an image is recorded by measuring the photo luminous. In order to overcome laser fluctuations we measure the normalized fluorescence, namely, $\frac{f_{la} - f_{lb}}{f_{la} + f_{lb}}$, where $f_{la}$ is the fluorescence when the MW is on and $f_{lb}$ is the fluorescence when the MW is off. According to this definition when the MW field is off-resonance the normalized fluorescence is zero. As previously explained, the fluorescence of the NV center decreases when the center g.s. is on resonance with a MW field frequency. Consequently, when the field is on resonance the normalized fluorescence is negative. Usually the iso-field imaging method is used when the changes of the magnetic fields are small compared to the linewidth of the spectrum. In our case the linewidth is in many cases broadened due to local magnetic fields and not split and therefore we are able to use this method. Since we apply a MW frequency higher than the zero-field resonance, a negative normalized fluorescence indicates the presence of a magnetic field. Our interest is in the general shape of the current density distribution, and we thus sum over the normalized fluorescence recorded for different MW frequencies which correspond to a range of several magnetic fields. The normalized fluorescence as a function of the spatial location is depicted in Fig. 8.7. From this fluorescence map we can learn the general shape of the current density distribution in the corner. We also provide simulations of the expected magnetic field. The data is in good qualitative agreement with the simulations.

8.5 Conclusions

We demonstrated the building blocks of an NV-based 2D wide-field magnetometer for the study of superconductivity. More experimental and computational effort is required for constructing full magnetic maps without any prior knowledge of the current density and the external field direction. The acquisition time of the measurement or alternatively, the field of view, is limited by the cooling power of the cryostat. While during my PhD I managed to probe the magnetic fields of a superconductor (both with a photodiode and with a CCD) – an important milestone on the way towards novel insight into high-$T_c$ superconductors, my further goal of wide-field imaging of vortices remained beyond reach. The main obstacle
Fig. 8.7: (a) Simulation of the absolute value of the field above the superconductor in the xy plane, provided by Vladimir Sokolovsky and Leonid Prigozhin on the basis of calculations done in [J.W. Barrett, L. Prigozhin, V. Sokolovsky, Supercond. Sci. Technol. 26, 105009 (2013); V. Sokolovsky, L. Prigozhin, J.W. Barrett, Supercond. Sci. Technol. 27, 124004 (2014)]. The current used is 0.75 of the critical current. Because we have not measured our critical current we are not able to turn the units into Gauss, so the simulation is presented here as a qualitative reference. (b) Simulation of the absolute value of the field above the superconductor in the z direction. (c) Simulation of the absolute value of the total magnetic field above the superconductor. This is the most relevant plot for a comparison with the data, since the data is composed of transitions made in all NV orientations. (d) An “iso-magnetic field”–like image of the superconducting corner as measured in our experiment. This image was produced by summing over the normalized fluorescence in the presence of different MW fields. To produce this figure we used the frequencies 2929 MHz, 2931 MHz, 2933 MHz, 2935 MHz, and 2937 MHz. For a detailed explanation of how this image was produced see text. Blue regions shows low normalized fluorescence (highly negative) and indicate the presence of a high magnetic field. Yellow regions shows high normalized fluorescence (close to zero) and indicate low magnetic fields. Black lines show the edges of the superconducting wire.

that prevented us from imaging vortices was the gap between the NV centers and the superconducting layer. The high-resolution imaging required for imaging vortices (which are typically 1-2 µm apart) demands that the gap between the SC layer and the NV layer be smaller than 1 µm. As explained in section 8.3, utilizing a 3 µm gold wire placed next to the YBCO wire, we determined the gap by measuring the magnetic field produced by the
wire (the wire was kept very narrow to avoid finite-size effects which degrade the magnetic gradients and thus hinder the resolution with which the gap may be measured). Since the local roughnesses of both the YBCO and diamond were measured to be less than 80 nm we believe that the measured \( \sim 2 \mu m \) distance between the NV centers and the superconducting layer is due to the curvature of the substrates. One of the options was thus to find SC layers which may be deposited directly onto the diamond surface. After two years of consultations around the world we were not able to find adequate high-\( T_c \) SCs, and the only SCs that seem to be perhaps adequate for this task were Nb, Ti, TiNb, or TiNbN, all of which have \( T_c \)'s in the range of 9-12 K. However, due to the features of our optical cryostat, we could not get attain a chip temperature below the required 12 K. As noted, we are now working together with our collaborators (the Budker group) on a new method in which 1 \( \mu m \)-thick diamonds are placed on the YBCO and are expected to closely follow the local topography so that the gap will become much smaller.
9. SUMMARY

This thesis focused on the basic physics of ensembles of NV centers as well as the possibility of using them as a sensitive magnetometer.

Concerning basic physics, we have analyzed in detail the interaction of the NV center with polarized MW radiation, and have critically analyzed a previously published analysis (Sec. 2.7). Next, we studied the source of the observed inhomogeneous broadening (chapter 4). Because of shortcomings with previous methods, we developed an improvement to the hole-burning saturation spectroscopy method. This novel method, inspired by the Doppler-free atomic spectroscopy technique, enabled us to re-check the origin of this broadening and our results support the claim that this broadening is due to local changes of the magnetic field inside the crystal. Finally, in chapter 5 we studied theoretically and experimentally the conditions for adiabatic behavior of an NV center exposed to a changing magnetic field. By adiabatic we mean the ability of the NV ground state population to stay in the same quantum state even though external conditions vary. We studied cases in which the magnetic field is changed along the $z$ axis, along the $y$ axis, and the case in which the magnitude of the magnetic field remains constant while its direction is changed in the $yz$ plane. Our results are in general agreement with our theoretical simulations. The results are surprising and counterintuitive as faster changes in the external field give rise to increased adiabaticity, whereas increasing adiabaticity typically corresponds to slower changes. Furthermore, in some cases there seems to be a universal time below which a transverse magnetic field does not give rise to non-adiabatic behavior. The results are now being studied by BGU theoreticians to see if they coincide with those expected for the straightforward Landau-Zener theory. These measurements also open the road for “spin-levitation” of NVs in magnetic traps that are similar to cold atom traps.

Concerning sensitive magnetometry, we have first addressed the problem of the zero-
field splitting due to strain. This problem prohibits magnetic measurements at low fields. We suggested two methods for overcoming the strain limitation (chapter 6). The first method described the use of a bias field in a configuration such that the bias field is orthogonal to the measured field. This method is required when a bias field along the measured field axis damages the measurement (e.g. when sensing superconductivity). The second method takes advantage of the MW polarization dependencies to increase the sensitivity. The eigenfunctions of the NV g.s. change in the vicinity of magnetic fields larger than the strain from $|m_s = 0\rangle$, $|m_s = +\rangle$, $|m_s = -\rangle$ to $|m_s = 0\rangle$, $|m_s = 1\rangle$, $|m_s = -1\rangle$, where the former are superpositions of the latter (the latter being the Zeeman eigenstates of the Hamiltonian). We showed theoretically that an improvement of the sensitivity could be achieved by measuring the quantity $\frac{|\langle 1^+ \rangle|^2}{|\langle -1^- \rangle|^2}$. In the third method presented in this chapter we describe a scheme for probing the spectrum of one specific NV orientation without applying a bias field. Probing just one orientation will result in a narrower spectroscopy line in the case of low-field measurements. In this method we take advantage of the NV’s optical and MW selection rules to isolate the fluorescence change due to one specific orientation.

In the last two chapters of this thesis we demonstrated the use of NV-based magnetometers for studying high-$T_c$ superconductors. Unlike SQUIDS, the NV-based magnetometer is capable of working in a wide range of temperatures and hence is more suitable for studying high-$T_c$ superconductors. Furthermore, it can measure the absolute value of the magnetic field, and can simultaneously probe the field from an area above the superconductor using a CCD. In chapter 7 we demonstrated the use of an NV magnetometer based on a confocal microscope and a photodiode to study phase transitions and vortex pinning in YBCO. In chapter 8 we use an EMCCD to build an NV based 2D magnetic imager. We use the 2D magnetic imager to study the current density distribution in a $90^\circ$ corner made of a thin-film superconductor.
A. EFFECTIVE 2D HAMILTONIAN APPROXIMATION

(Written by Yonathan Japha) Consider a Hamiltonian $H$ in which one of the diagonal elements $H_{00} = E_0 + \delta E_0(t)$, where $|\delta E_0| \ll |E_0|$ and $E_0$ is much larger than the other elements of $H$. This element represents a level which is energetically well separated from the other levels. We write a general state of the system as $|\psi\rangle = \sum_n c_n(t)|n\rangle$ and then the Schrödinger equations for the coefficients read

$$i\hbar \dot{c}_0 = (E_0 + \delta E_0)c_0 + \sum_{j \neq 0} H_{0j}c_j$$  \hspace{1cm} (A.1)

and for $j \neq 0$

$$i\hbar \dot{c}_j = \sum_k H_{jk}c_k.$$  \hspace{1cm} (A.2)

We now assume that initially the level $|0\rangle$ is not populated. The formal solution for $c_0$ is

$$c_0(t) = e^{-i\omega_0 t - i \int_0^t dt' \delta \omega_0(t')} c_0(0) - \frac{i}{\hbar} \int_0^t dt' e^{-i\omega_0(t-t')} - i \int_0^t dt' \delta \omega_0(t') \sum_{j \neq 0} H_{0j}(t') c_j(t'),$$  \hspace{1cm} (A.3)

where $\omega_0 = E_0/\hbar$ and $\delta \omega_0 = \delta E_0/\hbar$. Since we assumed that $|0\rangle$ is not populated, we can set $c_0(0) = 0$. Then we integrate by parts over the first term in the integral $e^{i\omega_0 t'}$ and obtain

$$c_0(t) = -\frac{1}{E_0} \sum_{j \neq 0} \left[ H_{0j}(t)c_j(t) - H_{0j}(0)c_j(0) e^{-i[\omega_0 t + \int_0^t dt' \delta \omega_0(t')]} \right]$$

$$+ \frac{1}{E_0} \int_0^t dt' e^{-i\omega_0(t-t')} \frac{d}{dt'} \left[ e^{-i \int_0^t dt'' \delta \omega_0(t'')} \sum_{j \neq 0} H_{0j}(t') c_j(t') \right].$$  \hspace{1cm} (A.4)

The time derivative in the integral is equal to

$$-i \delta \omega_0 H_{0j}c_j + \dot{H}_{0j}c_j + H_{0j} \dot{c}_j.$$  \hspace{1cm} (A.5)
According to our assumptions that \(|H_{0j}| \ll E_0\) and \(|\delta \omega_0| \ll E_0\), in addition to the assumption that the frequency components of \(H_{0j}\) are much smaller than \(\omega_0\), we may conclude that the integral (last term) is much smaller than 1. In addition, the second term oscillates as \(e^{-i\omega_0 t}\), which is much faster than the other elements in the equation, so that this term can also be neglected (rotating-wave approximation). We are then left with the first term as an approximation. We can now substitute for \(c_0\) in the equations of motion for the other coefficients \(c_j\) and obtain

\[
i \hbar \dot{c}_j = \sum_{k \neq 0} \left[ H_{jk} c_k - \frac{H_{j0}^0 H_{0k}}{E_0} \right] c_k.
\] (A.6)

A.0.1 Specific example

In the case of a \(3 \times 3\) Hamiltonian for the states \(|-1\rangle\), \(|0\rangle\) and \(|1\rangle\), we can write the \(2 \times 2\) effective Hamiltonian as

\[
H_{\text{eff}} = \begin{pmatrix}
H_{-1,-1} & H_{-1,1} \\
H_{1,-1} & H_{1,1}
\end{pmatrix} - \frac{1}{E_0} \begin{pmatrix}
H_{-1,0} H_{0,-1} & H_{-1,0} H_{0,1} \\
H_{1,0} H_{0,-1} & H_{1,0} H_{0,1}
\end{pmatrix}.
\] (A.7)

For the spin-1 system the matrix \(S_y\) is given by

\[
\hat{S}_y = \frac{1}{\sqrt{2}} \begin{pmatrix}
0 & i & 0 \\
-i & 0 & i \\
0 & -i & 0
\end{pmatrix},
\] (A.8)

so that the effective Hamiltonian due to the effect of a magnetic field \(H = -g_S \mu_B B_y \hat{S}_y\) is given by

\[
\delta H_{\text{eff}} = \frac{g_S^2 \mu_B^2 B_y^2}{2E_0} \begin{pmatrix}
1 & -1 \\
-1 & 1
\end{pmatrix}.
\] (A.9)
Sample preparation: The diamond plates were created by a combination of plasma etching and e-beam lithography from an Element 6 CVD diamond with \{100\} surfaces and an initial nitrogen concentration below 1 ppm. The NV-rich layer close to the surface of the diamond was created using ion implantation of nitrogen ions with energies of 10, 35, and 50 keV. Afterwards the diamond was annealed at 800°C for 10 hours and at 1200°C for 2 hours to form the NV centers. The plates were broken out of an array of plates using a micromanipulator and allowed to fall onto the SC. The YBCO sample properties are similar to those described at chapter 8.

Generation of vortices: The vortices were generated by field cooling the sample in the presence of an external magnetic field. We recall that the density of the vortices as a function of the external magnetic field is given by $B = n \cdot \Phi_0$. The images were obtained by holding the frequency constant at 2877 GHz and measuring the fluorescence from each pixel with and without MW. Different densities of vortices as a function of the field strength during the cooling of the system are depicted in Fig. B.1.
Fig. B.1: (a-e) Imaging of superconducting vortices at different cooling fields. The vortices were imaged by holding the MW frequency constant at 2877 MHz (the MW field is on resonance with the zero field splitting) and measuring the contrast. Images were taken for cooling fields of 0, 0.53, 0.79, 1.2, and 1.8 G. An increase in the number of vortices can be observed when the cooling field is increased. (f) A comparison of the imaged vortex density to the expected relation $B = n \cdot \Phi_0$. 

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ท่องเที่ยว

 travail de recherche consacré à la localisation des niveaux d'énergie discrets

 particular de l'ion de Rubidium, avec une onde faible de lumière infrarouge.

 basé sur la caractérisation de la polarisation de l'ionRubidium, en utilisant

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הצהרת תלמיד המחבר ע"ע הגשת עבדת הדוקטור ללא פיצוי

אני החותם מוטה מצהיר/ה בראות: (නන ספן):)

• היבחרתי את היבחרי שעומי, להציגה ע"ע התעוררה שקובלתי槭א מנחתים.

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למקימה, שאושרה על יד מוגש בחסכמה.

האריך ש"ע התلمוד: יוחאי שליסל

חתימה

*ראה הצהיר בהאגילית לפורים תורומתי השותפים למקימה
העבדה נעשתה בחדות ור"פ: וּמִלְמֵן

במסלולクラス פיזייק

במסלולundi העבש
מכתב בוגש מרכז צבע חנקן-ריק ביהלום

מחקץ לשם מח出来る על הדרישות בככלת תואר "דוקטור לפילוסופיה"

מחת

יחזקאל שילסל

הוגש טלונאותי אוניברסיטת בן גוריון בנגב

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אישור המנחה

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אישור דיקן בית הספר ללימודי מחקר מתכדמים ע"ש קריטמן

יולי 2017

תומחتشע"ז

באור שבע
מחקם банושא מרכזי באע חנקק-ריק ביהלום

מחקם לשון דיליה חללי של הדרישות למקבלת תואר "דוקטור לפילוסופיה"

מאית

יוחכאל שליסל

הוגש לפיתאון ואוניברסיטת בן גוריון בנגב

יולי 2017