Multimodal Spin Based Sensors

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Abstract

This is where you summarise the contents of your dissertation. It should be at least 100 words, but not more than 250 words.

Need to write an abstract

Declaration

I declare that this dissertation was composed entirely by myself.

Personal Statement

The project began with developing a deeper understanding of the physics underlying spintronics. The main focus was on electron paramagnetic resonance, specifically using the continuous wave optically detected magnetic resonance technique. For this, there is a wealth of literature on the diamond nitrogen vacancy (DNV). Most popular is the application of the DNV as a very sensitive magnetometer.

I worked to

I worked to understand the intricate details with reference to diamond hoping I could apply this knowledge to SiC

When I felt comfortable with the underlying physics, I began modelling the different system Hamiltonians. I applied varying $\vec{B}$ and $\vec{E}$ as well as varied temperature. The goal was both to understand the influence of these external factors on the spin-system energy levels as well as to verify my model behaved correctly in the simple cases when compared to existing literature.

When I had the capability to dynamically model both the DNV and several SiC defects, with different spin numbers, I created ensembles of specifically chosen defects to visualise how the CW-ODMR spectra might change under the influence of varying $\vec{B}$, $\vec{E}$ and T. This, as well as existing literature allowed me to isolate specific defects which were most appropriate for the sensing of specific variables. For example, the V2 Silicon defect in SiC is very insensitive to changes in temperature so would not be the most appropriate for thermometry application.

When an ensemble of defects was selected for a specific multi-modal application and the nature of the changes to the ODMR spectra was understood, I worked to develop a method to extract and disentangle the influence of each individual influence on the spectra.

This process was repeated for several model systems and in the end I developed a theoretical framework for a multi-modal sensing application of specifically chosen defects in SiC.

During the course of the project, I met with my supervisor every week, in order to discuss my progress and the direction I would head next. Toward the end, the frequency of our meetings increased somewhat, as I began to finish my calculations.

I started writing this dissertation in mid-July, and I spent the first three weeks of August working on it full-time.

Overall, I feel that the project was a success, and I found it to be extremely enjoyable throughout.

Review before submission

Acknowledgements

I'd like to thank my supervisor Professor Cristian Bonato for making this project possible, I am particularly grateful for his patience and his ability to make complex subjects seem approachable and achievable.

Additionally, I would like to thank the Royal Air Force Director of Defence Studies and the Chief of the Air Staff for financially supporting this research and I look forward to applying what I have learned to my time in service.

Most of all I would like to thank my wife Sophie for her ongoing support and patience. I could not have completed this work without your help.

Todo list

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List of Figures

Chapter 1

Introduction

Solid-state colour centres, which exists in many materials such as diamond and silicon carbide, have been one of the leading systems in quantum technology [3, 4]. The nitrogen-vacancy (NV) centre in diamond is the most comprehensively studied solid-state spin defect. The defect spin state can be initialized by laser and controlled by microwave [5, 6, 7]. It has been used in various quantum technologies, such spin–photon entanglement, a quantum computing qubit register and high-sensitivity nanoscale quantum sensing, the focus of this work [8, 9].

The NV centre is favoured for it's for its excellent quantum properties, but drawbacks of the system are a lack of established nanotechnology and the fluorescence wavelength of the NV centre, which is in the visible range and limits its wider applications [10, 11, 12] .

Develop introduction - merge lower paragraphs into this one

The field of spectroscopy studies the way atoms and molecules interact with and exchange energy with a wider physical system - specifically through electromagnetic radiation. The electric field interacts with with the electric dipole moment and the magnetic field interacts with a magnetic dipole moment. Magnetic resonance spectroscopy focusses specifically on the interaction between the **B** field with magnetic moments which exist in a given material. This can be broken into two distinct fields:

Nuclear Magnetic Resonance (NMR) which studies the interaction with nuclear magnetic moments.

Electron Paramagnetic Resonance (EPR) which studies the interaction with magnetic moments of electrons.

Using Planck's relationship $E = h\nu$ and $c = \lambda\nu$ we may characterise the electromagnetic radiation by its energy which is, to a constant, equivalent to the frequency or the wavelength. EPR is observed in systems where the magnetic dipole of the electron is influenced by an applied, oscillating magnetic field forcing transitions between electron energy levels. In general the measurable difference in energy levels for which the transition occurs is caused by an external magnetic field via the Zeeman effect. Some systems also exhibit energy level splitting in the absence of an applied external magnetic field so called zero field splitting (ZFS).

EPR is thus a tool to manipulate electron spins in solid state materials. The transition between energy levels is quantised thus the discrete amount of energy which is lost by

the system is transferred into a photon or charge state which may be detected optically or electrically [13].

A particularly successful technique is Optically Detected Magnetic Resonance (ODMR) which uses an applied microwave frequency, an oscillating magnetic field with energy quanta equivalent to the transitions between Zeeman sub levels, to drive the repopulation of those Zeeman sub levels following a spin-dependent optical transition. In essence this boosts the sensitivity since the microwave driven repopulation induces a change in photoluminescence with a much higher and thus much more readily detectable energy. The techniques of ODMR are so effective that even a single electron spin may be detected this way [14].

Spintronics, a portmantau of **spin** and elec**tronics** is a technology which exploits the characteristics of spin akin to how charge is manipulated in electronics. Fundamentally, the smallest stable magnetic moment available in nature is generated by the spin of a single electron. If efficient read-out can be achieved, the sensitivity of the electron magnetic dipole cannot be matched. Careful construction of an appropriate system, or identification of a system with appropriate characteristics allows for the initialisation, manipulation and read-out of EPR from which we may infer the physical properties of the environment surrounding the system.

With ODMR of the NV centre in diamond the manipulation of spin states in single, atomic-sized centres at room temperature has been demonstrated despite spin polarisation being a primarily thermodynamic effect (see section **??**). This is possible since optical excitation of the energy levels decay faster via a spin-preserving transition, leading to an inverse population of spin sublevels in its ground state when the system is irradiated consistently for several excitation/decay cycles.

need a reference for the thermodynamic comment

This prompted the search for other structures with similar unique quantum properties. Silicon carbide (SiC) is a promising candidate (discussed in detail in section 2.18). A major benefit of SiC is the existence of various polytypes, which each exhibit unique spin colour centre properties. Furthermore, even within a single polytype, these centres can occupy distinct and non-equivalent lattice positions. The existence of these colour centres with similar properties but different energy quanta allows for selection of a specific defect with parameters suitable for the problem at hand.

Consider writing a paragraph on ENDOR - only if relevant later in the project.

EPR spectroscopy can be approached by different methods, relevant to this work:

Continuous Wave (CW) where the magnitude of the static magnetic field (B_0) is swept, while the amplitude of the driving field B_1 is constant with time.

Pulsed where a time-dependent driving pulse B_1 is applied in addition to a static magnetic field B_0 [15].

Electron-Electron Double Resonance (ELDOR) where two microwave frequencies participate;

1. The "pump" microwave source, irradiates a portion of the ESR spectrum.
2. The effect of this irradiation on another portion of the spectrum is monitored by an observe microwave source. [16]

This work looks to explore how the physical characteristics which influence the Hamiltonian and thus the energy of the electron spin may be inferred by measuring the

effects of those characteristics on the EPR of the specific system. Further, it will look to explore whether the compound effect of multiple influences may be disentangled and distinguished simultaneously.

Chapter 2

Background Theory

2.1 Magnetism

Where charge (**E**-field) has an intuitive elementary source unit of a point charge (or monopole) which may be positively or negatively charged. Conversely the elementary source unit of magnetism (**B**-field) is the magnetic dipole.

2.1.1 Magnetic Dipole

Classically, the magnetic dipole may be modelled as a closed loop that carries an electric current. Its magnetic dipole moment, $\vec{\mu}$, is defined as the vector which points out of the plane of the current loop,

$$\vec{\mu} = IS\vec{n} \tag{2.1}$$

where I is the current in the loop and S is the surface area enclosed by the loop.

The magnetic dipole produces a magnetic field $\vec{B}$, which for points a large distance from the dipole may be calculated as :

Type up derivation from David Tong notes?

$$\vec{B} = \frac{\mu_0}{4\pi}\frac{1}{r^3}\left[\frac{3(\vec{\mu}\cdot\vec{r})\cdot\vec{r}}{r^2} - \vec{\mu}\right]$$

The symmetry of the field enables us to, without any loss of generality, consider the direction of the dipole the z-axis. Then, defining x, y as usual by $r\cos\theta$ and $r\sin\theta$ respectively. We may then consider magnetic field in two separate components, parallel (B_z) and perpendicular (B_x, B_y):

$$B_{\parallel} = \frac{\mu_0}{r^3}(3\cos^2\theta - 1), \quad B_{\perp} = \frac{3\mu_0}{r^3}\cos\theta\sin\theta.$$

Then, we may use the Pythagorean principle to determine the overall magnitude B as

$$B = \sqrt{B_{\parallel}^2 + B_{\perp}^2}.$$

2.1.2 Gyromagnetic Ratio

Classical Derivation

The current in equation 2.1 is proportional to the angular momentum of the charge. That is, the dipole moment is always associated with an angular momentum $\vec{G} = \vec{r} \times \vec{p}$ with $\vec{r}$ the radius and $\vec{p}$ the momentum.

Dividing the magnetic dipole moment by the angular momentum we find the **gyromagnetic ratio**.

$$\gamma = \frac{\vec{\mu}}{\vec{G}}. \tag{2.2}$$

Without loss of generality we may consider the most simple case which is where the magnetic dipole moment is parallel (or anti-parallel) to the angular momentum. Then we may consider the absolute values for the dipole moment and the angular momentum:

$$\mu = IS, \quad I = \frac{qv}{2\pi R}, \quad S = \pi R^2 \tag{2.3}$$

We substitute I and S to find

$$\mu = \frac{qvR}{2} \tag{2.4}$$

and further, we equate the angular momentum vector, using the model of a planar loop to

$$G = m_q v R \tag{2.5}$$

leaving

$$\gamma = \frac{q}{2m_q}. \tag{2.6}$$

We finally consider that we may represent the, currently unknown, charge and mass as a sum of electron charges and masses.

$$\gamma = \frac{q}{2m_q} = \frac{\cancel{N}e}{2\cancel{N}m_e} \implies \gamma = \frac{e}{2m_e} \tag{2.7}$$

We therefore find that the gyromagnetic ratio of the electron depends only on fundamental constants [17].

Extending to Quantum Mechanics

Since the gyromagnetic ratio was calculated considering the motion of dipole in a loop, we may extend this to an electron in an orbit within the atom. The fundamental change required to extend the model to quantum mechanics is the treatment of angular momentum which should now be quantised. Thus, we replace our classical approximation of $\vec{G} = \vec{r} \times \vec{p}$ with the equation for the eigenvalues of the quantum mechanical representation of orbital angular momentum:

$$\hat{G} = \hbar \hat{J} \tag{2.8}$$

where $\hat{J}$ is the operator of the orbital angular momentum (quantum number of orbital momentum).

The angular momentum and total energy are conserved in general in a closed system. We consider the time independent Shrödinger equation

Write up / expand on Noether currents?

$$\hat{H}\Psi_n = E_n\Psi_n \tag{2.9}$$

We may choose Ψ_n such that it is an eigenfunction of the Hamiltonian, the total angular momentum squared ($J^2 = J_x^2 + J_y^2 + J_z^2$) and exactly one directional component of the angular momentum which is by convention chosen as J_z.

Need to develop why?

According to quantum mechanics the projection of J along the quantisation axis (M_J) may take integer values $-J, -J+1, \ldots, J-1, J$. Thus, we may describe a given quantum state by the spin J and the projection of the spin M_j. Thus, using Dirac notation we may write

Discuss why?

$$\hat{H}\left|J, M_J\right\rangle = E\left|J, M_J\right\rangle \tag{2.10}$$
$$\hat{J}^2\left|J, M_J\right\rangle = J(J+1)\left|J, M_J\right\rangle \tag{2.11}$$
$$\hat{J}_z\left|J, M_J\right\rangle = M_L\left|J, M_J\right\rangle. \tag{2.12}$$

Thus, the operator which describes the orbital magnetic moment may be written as (using equations 2.7, 2.8)

$$\hat{\vec{\mu}}_J = \gamma\hat{\vec{G}}_J = \gamma\hbar\hat{\vec{J}} = \frac{e\hbar}{2m_e c}\hat{\vec{J}}. \tag{2.13}$$

This leads to a quantity known as the **Bohr Magneton**, μ_B, given by

$$\mu_B = \frac{|e|\hbar}{2m_e c}. \tag{2.14}$$

Using this we may write equation 2.13 as

$$\hat{\vec{\mu}}_J = -\mu_B\hat{\vec{J}}. \tag{2.15}$$

Change all J's above this to L's

2.1.3 g-factor

The above expression is valid for the orbital electron but may be extended to a more general system by introducing a g-factor. The g-factor is equivalent to a dimensionless gyromagnetic ratio [18], so equation 2.15 may be written with $g = 1$ as

$$\hat{\vec{\mu}}_L = -g\mu_B\hat{\vec{L}}. \tag{2.16}$$

2.2 Landé g-factor

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2.3 Spin

As well as the orbital magnetic moment generated by the orbital angular momentum of the electron, the electron also possesses an intrinsic magnetic moment. Classically this implies an intrinsic angular momentum hence the magnetic moment of elementary particles is termed spin.

For a single electron spin may take the value $\pm 1/2$ since the system has only been observed in two possible states [19] and experiments confirm that the orbital angular momentum and spin angular momentum are of the same nature and thus may be summed. The magnetic moment of the spin may thus be expressed as 2.16 [20] where $g \approx 2.0023$ [21, 22].

In reality the electron is point-like and thus the current loop model is unsuitable. Spin is actually a purely quantum mechanical effect and a consequence of the algebra required to satisfy the Dirac equation of relativistic quantum mechanics. The manifestation of this degree of freedom however, is a system which has the physical characteristics as described in this section.

We thus consider the total angular momentum of a system J given by

$$J = L + S \tag{2.17}$$

which make take the values $L + S, L + S - 1, \ldots, |L - S|$.

2.4 Zeeman Effect

When no magnetic field is applied to a system, the magnetic dipoles of the orbital electron and spin have no preferred direction. The energy levels for all combinations of L and S (all J) are equivalent.

If a magnetic field is applied the magnetic moments interact with that field via the Zeeman interaction. The Zeeman effect consists of atomic energy level splitting when an external magnetic field is imposed on a sample [23].

The classical expression for the energy of a dipole in a magnetic field

$$E = -\vec{\mu} \cdot \vec{B} \tag{2.18}$$

may be replaced with the Hamiltonian for a quantum mechanical system

$$\hat{H}_{\text{Zeeman}} = -\hat{\vec{\mu}} \cdot \vec{B}. \tag{2.19}$$

The negative sign indicates that when the magnetic moment is parallel to the magnetic field the lowest energy is achieved.

Thus distinct quantum systems with different J and thus different projections of angular momentum (m_J) have different energies due to their interaction with a magnetic field.

Considering a simple two-level system ($S = 1/2$), the energy difference between the spin being aligned or anti-aligned with the field is called the Zeeman energy.

The Hamiltonian to describe the energy is, using the total angular momentum form of 2.16,

$$\hat{H}_{\text{Zeeman}} = g\mu_B \hat{\vec{S}} \cdot \vec{B}. \tag{2.20}$$

Without loss of generality we may direct the magnetic field along the z axis and reduce the scalar product to only the z component. Now, using $S = 1/2$ quantised along the $z - axis$, i.e. $m_S = \pm 1/2$ we find the Zeeman energy by solving the Shrödinger equation

$$\hat{H}_{\text{Zeeman}} \left|S, m_S\right\rangle = E_{\text{Zeeman}} \left|S, m_S\right\rangle \tag{2.21}$$

which, to a factor is equivalent to, by 2.12, to

$$\hat{S}_Z \left|S, m_S\right\rangle = M_S \left|S, m_S\right\rangle . \tag{2.22}$$

Thus we find the two eigenvalues to be

$$E_+ = \frac{1}{2} g\mu_B B, \qquad E_- = -\frac{1}{2} g\mu_B B$$

and thus the Zeeman energy is given by $g\mu_B B$.

The $S = 1/2$ system is thus doubly degenerate and the degeneracy is lifted by the application a magnetic field. The Zeeman energy is the difference between the two states and it grows linearly with B.

This may be generalised to a more complex system by considering the total angular momentum J where the energy difference between states is given by

$$\Delta E = g_J \mu_B B. \tag{2.23}$$

2.5 Spin-Orbit Interaction

The orbital magnetic dipole may interact with the intrinsic spin magnetic dipole via the Spin-Orbit interaction. This is represented by the spin-orbit Hamiltonian with λ representing the constant of the coupling:

$$H_{\mathrm{SO}} = \lambda \hat{\vec{L}} \cdot \hat{\vec{S}}. \tag{2.24}$$

This is caused by the interaction between the magnetic field generated by the relativistic motion of the electron around the nucleus and that of the spin magnetic moment. The coupling is proportional to the atomic mass.

2.6 Perturbation Theory

By considering a ground, non-degenerate state and a perturbation in the electron Zeeman interaction and the spin-orbit coupling we can develop insight into so called zero field splitting. The perturbation is given by

$$\hat{H}' = \hat{H}_{\mathrm{Zeeman}} + \hat{H}_{\mathrm{SO}} \tag{2.25}$$

for which we find

$$E_0 = E_0^{(0)} + \langle 0| \hat{H}' |0\rangle + \sum_n \frac{\langle 0| \hat{H}' |n\rangle \langle n| \hat{H}' |0\rangle}{E_0^{(0)} - E_n^{(0)}} \tag{2.26}$$

Now, if we consider arbitrary interactions of forms

$$\hat{H}_{\mathrm{Zeeman}} = g_L \mu_B \hat{\vec{L}} \cdot \vec{B} + g_S \mu_B \hat{\vec{S}} \cdot \vec{B} \tag{2.27}$$

$$\hat{H}_{\mathrm{SO}} = \lambda \hat{\vec{L}} \cdot \hat{\vec{S}} \tag{2.28}$$

we may compute the first and second order corrections.

First Order

Substituting 2.27 and 2.28 into 2.26 and integrating only over the orbital values to deduce the spin Hamiltonian (discussed more in section **??**) we find

Link section

$$\begin{aligned}
\langle 0| \hat{H}' |0\rangle &= \langle 0| g_L \mu_B \hat{\vec{L}} \cdot \vec{B} + g_S \mu_B \hat{\vec{S}} \cdot \vec{B} + \lambda \hat{\vec{L}} \cdot \hat{\vec{S}} |0\rangle \\
&= \langle 0| g_L \mu_B \hat{\vec{L}} \cdot \vec{B} |0\rangle + \langle 0| g_S \mu_B \hat{\vec{S}} \cdot \vec{B} |0\rangle + \langle 0| \lambda \hat{\vec{L}} \cdot \hat{\vec{S}} |0\rangle \\
&= g_L \mu_B \vec{B} \cdot \langle 0| \hat{\vec{L}} |0\rangle + g_S \mu_B \vec{B} \cdot \hat{\vec{S}} \langle 0|0\rangle + \lambda \hat{\vec{S}} \cdot \langle 0|\hat{\vec{L}}|0\rangle \\
&= g_L \mu_B \vec{B} \cdot \cancelto{0}{\langle 0| \hat{\vec{L}} |0\rangle} + g_S \mu_B \vec{B} \cdot \hat{\vec{S}} \cancelto{1}{\langle 0|0\rangle} + \lambda \hat{\vec{S}} \cdot \cancelto{0}{\langle 0|\hat{\vec{L}}|0\rangle} \\
&= g_s \mu_B \hat{S} \cdot \hat{B}.
\end{aligned} \tag{2.29}$$

Here we used the fact that $\langle 0|\hat{\vec{L}}|0\rangle = 0$ since, for example in the alegbraic basis $\hat{L}_z = -i\left(x\frac{\partial}{\partial y} - y\frac{\partial}{\partial x}\right)$ is a Hermitian operator is therefore has eigenvalues which are strictly real numbers, i.e.

$$\hat{L}_z \left|\psi\right\rangle = m_L \left|\psi\right\rangle . \tag{2.30}$$

By considering 2.30 we see that if we apply an imaginary operator to a real valued eigenfunction the corresponding eigenvalue must be imaginary or zero. We know the state is strictly real since it is non-degenerate[1]. In this case, the expectation value of $\hat{L}$ can only be 0.

Zeeman Splitting. The result of the first order perturbation is thus a more formal confirmation of the result of section 2.4, specifically 2.23.

Second Order

At second order, again substituting 2.27 and 2.28 into 2.26 and integrating only over the orbital values we find

$$\begin{aligned}
&\sum_n \frac{\langle 0|\,\hat{H}'\,|n\rangle\,\langle n|\,\hat{H}'\,|0\rangle}{E_0^{(0)} - E_n^{(0)}} \\
&= \frac{\langle 0|g_L\mu_B\hat{\vec{L}}\cdot\vec{B} + g_S\mu_B\hat{\vec{S}}\cdot\vec{B} + \lambda\hat{\vec{L}}\cdot\hat{\vec{S}}|n\rangle\,\langle n|g_L\mu_B\hat{\vec{L}}\cdot\vec{B} + g_S\mu_B\hat{\vec{S}}\cdot\vec{B} + \lambda\hat{\vec{L}}\cdot\hat{\vec{S}}|0\rangle}{E_0^{(0)} - E_n^{(0)}} \\
&= \frac{\langle 0|g_L\mu_B\hat{\vec{L}}\cdot\vec{B} + \lambda\hat{\vec{L}}\cdot\hat{\vec{S}}|n\rangle\,\langle n|g_L\mu_B\hat{\vec{L}}\cdot\vec{B} + \lambda\hat{\vec{L}}\cdot\hat{\vec{S}}|0\rangle}{E_0^{(0)} - E_n^{(0)}} \\
&= (g_L\mu_B\vec{B} + \lambda\hat{\vec{S}})\underbrace{\sum_n \frac{\langle 0|\hat{\vec{L}}|n\rangle\,\langle n|\hat{\vec{L}}|0\rangle}{E_0^{(0)} - E_n^{(0)}}}_{\Lambda}(g_L\mu_B\vec{B} + \lambda\hat{\vec{S}})
\end{aligned} \tag{2.31}$$

Here Λ is a matrix composed of the elements as shown. Expanding out, this allows us to write the second order perturbation as

$$\sum_n \frac{\langle 0|\,\hat{H}'\,|n\rangle\,\langle n|\,\hat{H}'\,|0\rangle}{E_0^{(0)} - E_n^{(0)}} = g_L^2\mu_B^2\vec{B}\cdot\Lambda\cdot\vec{B} + 2\lambda g_L\mu_B\hat{\vec{S}}\cdot\Lambda\cdot\vec{B} + \lambda^2\hat{\vec{S}}\cdot\Lambda\cdot\hat{\vec{S}}. \tag{2.32}$$

Since for EPR we are only interested in the spin-dependent terms, the first term may be neglected as it represents a global shift in the energy spectra.

[1] A complex wavefunction ψ is at least doubly degenerate; the complex conjugate ψ^* has the same energy.

Combined Perturbation

Combining 2.29 and 2.32 we find

$$\begin{aligned}\langle 0|\,\hat{H}'\,|0\rangle + \sum_n \frac{\langle 0|\,\hat{H}'\,|n\rangle\,\langle n|\,\hat{H}'\,|0\rangle}{E_0^{(0)} - E_n^{(0)}} &= g_S\mu_B\hat{\vec{S}}\cdot\vec{B} + 2\lambda g_L\mu_B\hat{\vec{S}}\cdot\Lambda\cdot\vec{B} + \lambda^2\hat{\vec{S}}\cdot\Lambda\cdot\hat{\vec{S}} \\ &= \mu_B\hat{\vec{S}}\cdot\underbrace{(g_S + 2g_L\lambda\Lambda)}_{g}\cdot\vec{B} + \hat{\vec{S}}\cdot\underbrace{\lambda^2\Lambda}_{D}\cdot\hat{\vec{S}}\end{aligned} \tag{2.33}$$

In this expression g and D are matrix quantities depending on Λ and represent the (possibly anisotropic) g factor and D the fine structure splitting.

For this work we will consider only systems in which the differences in angular momentum is due only to the spin and thus g is reduced to a scalar quantity in the spin Hamiltonian.

The term depending on D has no dependence on magnetic field and thus this fine-structure splitting is known as zero field splitting (ZFS) and is observed in systems with $S > 1/2$.

$$H_{\mathrm{FS}} = \hat{\vec{S}}\cdot D\cdot\hat{\vec{S}}. \tag{2.34}$$

2.7 Zero Field Splitting

ZFS is in fact due to the combined effects of fine structure and a dipole-dipole interaction. These effects manifest themselves identically which makes them difficult to separate experimentally. They each depend on a traceless matrix D, as will be shown, which can be totally described by two parameters, conventionally labelled D and E. For simplicity in this work we will consider the combined effect of both the fine-structure and the dipole-dipole interaction as the ZFS interaction. This means when D and E are measured for a specific system, they represent the compound effect of fine-structure splitting and the dipole interaction, but totally describe the zero-field splitting.

2.7.1 Fine Structure

The matrix D in 2.34 has form

$$D = \begin{pmatrix} D_{xx} & D_{xy} & D_{xz} \\ D_{yx} & D_{yy} & D_{yz} \\ D_{zx} & D_{zy} & D_{zz} \end{pmatrix} \tag{2.35}$$

which may be simplified by alignment to the wider system axis and diagonalising the matrix as

$$D = \begin{pmatrix} D_{xx} & 0 & 0 \\ 0 & D_{yy} & 0 \\ 0 & 0 & D_{zz} \end{pmatrix}. \tag{2.36}$$

The trace of the matrix $\mathrm{Tr}(D)$ is unchanged by the change of basis. Since for EPR we are only concerned with the changes in energy and not the absolute, we may choose

the value of the trace without any loss of generality, so we set it equal to zero.

$$\mathrm{Tr}(D) = 0. \tag{2.37}$$

This means that the diagonal form of D may be fully determined by just two parameters

$$D = D_{zz} - (D_{xx} + D_{yy})/2 \tag{2.38}$$
$$E = (D_{xx} - D_{yy})/2 \tag{2.39}$$

Here D represents the axially symmetric parameter and E represents any non-axial contribution of the fine-structure interaction.

Substituting 2.38 and 2.39 into 2.34 and expanding allows us to write our fine-structure Hamiltonian as

$$H_{\mathrm{FS}} = D\left(\hat{S}_z^2 - \frac{1}{3}S(S+1)\right) + E\left(\hat{S}_x^2 - \hat{S}_y^2\right). \tag{2.40}$$

2.7.2 Dipole-Dipole Interaction

We will now show that the interaction between the magnetic dipoles of two electrons has the same form as 2.34 by considering two electrons ($S = 1/2$).

We begin with the classical expression for the energy between two magnetic dipoles, μ_1, μ_2

$$E = \frac{1}{r^3}\left(\mu_1 \cdot \mu_2 - \frac{3(\mu_1 \cdot \vec{r})(\mu_2 \cdot \vec{r})}{r^2}\right). \tag{2.41}$$

Substituting the quantum mechanical operators for the two electron magnetic dipoles we find

$$H_{\mathrm{DD}} = g_S^2 \mu_B^2 \frac{1}{r^3}\left(\hat{\vec{S}}_1 \cdot \hat{\vec{S}}_2 - \frac{3(\hat{\vec{S}}_1 \cdot \vec{r})(\hat{\vec{S}}_2 \cdot \vec{r})}{r^2}\right). \tag{2.42}$$

Considering the total spin of the system we may expand this to obtain [13]

$$H_{\mathrm{DD}} = \frac{1}{2r^5} g_S^2 \mu_B^2 \hat{\vec{S}} \cdot \underbrace{\begin{pmatrix} r^2 - 3x^2 & -3xy & -3xz \\ -3xy & r^2 - 3y^2 & -3yz \\ -3xz & -3yz & r^2 - 3z^2 \end{pmatrix}}_{D} \cdot \hat{\vec{S}}. \tag{2.43}$$

As with 2.36 the matrix D in 2.43 has a constant trace (which we may select to be 0) leaving the form of the dipole-dipole interaction identical to that of the fine structure interaction

$$H_{\mathrm{DD}} = \hat{\vec{S}} \cdot D \cdot \hat{\vec{S}}. \tag{2.44}$$

We therefore decompose the traceless matrix D into the axial and non-axial parameters D and E as above.

2.7.3 Zero Field Splitting Hamiltonian

When measuring the values of D and E experimentally, the combined effect will be contained within those measurements so we may therefore describe the zero field splitting interaction as a whole using

$$H_{\mathrm{ZFS}} = D\left(\hat{S}_z^2 - \frac{1}{3}S(S+1)\right) + E\left(\hat{S}_x^2 - \hat{S}_y^2\right). \tag{2.45}$$

2.8 Nuclear Hamiltonians

2.8.1 Hyperfine Interaction

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2.8.2 Nuclear Quadrupole

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2.9 Stark Effect

Deduce Stark Effect Hamiltonian and write up

The classical energy of an electric dipole $\vec{d} = e\vec{r}$ in an electric field $\vec{E}$ is given by [24]

$$E = \vec{d} \cdot \vec{E}. \tag{2.46}$$

ref perturbation theory section

As we did in section **??** we will perturb the potential using $H' = \vec{d} \cdot \vec{E}$. To simplify the calculation we will assume the $\vec{E}$ field is applied along the z axis i.e.

$$H' = -\mu_z \mathcal{E} \tag{2.47}$$

Update notation here

Thus for we find for the ground state

$$E_0 = E_0^{(0)} - \langle 0|H'|0\rangle + \sum_n \frac{\langle 0|H'|n\rangle \langle n|H'|0\rangle}{E_0^{(0)} - E_n^{(0)}} \tag{2.48}$$

and we may compute the first and second order effect.

First Order

Substituting 2.47 into 2.48 in the first order terms we find

$$\langle 0|H'|0\rangle = -\mathcal{E} \langle 0|\mu_Z|0\rangle . \tag{2.49}$$

Second Order

Similarly, substituting 2.47 into 2.48 in the second order terms we find

$$\sum_n \frac{\langle 0|H'|n\rangle \langle n|H'|0\rangle}{E_0^{(0)} - E_n^{(0)}} = \mathcal{E}^2 \left(\sum_n \frac{\langle 0|\mu_z|n\rangle \langle n|\mu_z|0\rangle}{E_0^{(0)} - E_n^{(0)}} \right) \tag{2.50}$$

2.10 Stark Effect (Group Theory)

For our Spin Hamiltonian given by

$$\begin{aligned} H &= H_{\text{ZFS}} + H_{\text{Zeeman}} + H_{\text{Hyperfine}} + H_{\text{Zeeman (n)}} + H_{\text{Quadrupole}} \\ H &= \hat{\vec{S}} \cdot \vec{D} \cdot \vec{S} + g\mu_b \hat{\vec{S}} \cdot \vec{B} + \hat{\vec{S}} \cdot A \cdot \hat{\vec{I}} - \mu_n g_n \hat{\vec{I}} \cdot \vec{B} + \hat{\vec{I}} \cdot Q \cdot \hat{\vec{I}}. \end{aligned} \tag{2.51}$$

In the most general sense, an applied electrical field change any of the parameters. We will not consider the effect of an applied electrical field on the nuclear Zeeman term as the nuclear is paramagnetically shielded. Thus, in a general sense we may add the contributions of an applied electrical field $\vec{E}$ as $H + H_{\text{Stark}}$ where

$$H_{\text{Stark}} = \vec{E} \cdot \left(\hat{\vec{S}} \cdot R \cdot \hat{\vec{S}} + T\mu_B \hat{\vec{S}} \cdot \vec{B} + \hat{\vec{S}} \cdot F \cdot \hat{\vec{I}} + \hat{\vec{I}} \cdot q \cdot \hat{\vec{I}} \right). \tag{2.52}$$

Here R, T, Fq are matrices for each component of the electric field given by

$$R_{ijk} = \frac{\partial D_{jk}}{\partial E_i}, \quad T_{ijk} = \frac{\partial g_{jk}}{\partial E_i}, \quad F_{ijk} = \frac{\partial A_{jk}}{\partial E_i}, \quad q_{ijk} = \frac{\partial Q_{jk}}{\partial E_i}. \tag{2.53}$$

We immediately simplify at least the T term as for this work we assume isotropic and constant g thus in all cases T is zero.

Also, as discussed in section **??** for this work any change in the nuclear Hamiltonian will represent a global shift of the energy spectra in EPR and is therefore not relevant.

Ref nuclear section

This allows us to then consider, in the Spin Hamiltonian, only the energy change due to the shift in the D parameter, that is the values of R.

We can consider R as a square matrix for each component of the applied $\vec{E}$. Exactly as we did for ZFS in section **??**, we may reduce each of these matrices to a symmetric and traceless form. Here we define D and E in exactly the same way as **??**.

link section

link D E def

Additionally, each matrix R must be symmetric, that is $R^T = R$,

Explain or reference section 3.5 Mims

Now consider the expansion of $\hat{\vec{S}} \cdot R \cdot \hat{\vec{S}}$ which is given explicitly for $S = 1$ as

$$\begin{aligned} \hat{\vec{S}} \cdot R \cdot \hat{\vec{S}} &= \begin{pmatrix} \hat{S}_x & \hat{S}_y & \hat{S}_z \end{pmatrix} \cdot \begin{pmatrix} R_{xx} & R_{xy} & R_{xz} \\ R_{xy} & R_{yy} & R_{yz} \\ R_{xz} & R_{yz} & R_{zz} \end{pmatrix} \cdot \begin{pmatrix} \hat{S}_x \\ \hat{S}_y \\ \hat{S}_z \end{pmatrix} \\ &= R_{xx}\hat{S}_x^2 + R_{yy}\hat{S}_y^2 + R_{zz}\hat{S}_z^2 \\ &+ R_{xy}(\hat{S}_x\hat{S}_y + \hat{S}_y\hat{S}_x) + R_{xz}(\hat{S}_x\hat{S}_z + \hat{S}_z\hat{S}_x) + R_{yz}(\hat{S}_y\hat{S}_z + \hat{S}_z\hat{S}_y) \end{aligned} \tag{2.54}$$

As described above, we set the constant trace equal to zero as for ZFS and rewrite the $R_{xx}\hat{S}_x^2 + R_{yy}\hat{S}_y^2 + R_{zz}\hat{S}_z^2$ as $R_D\left(\hat{S}_z^2 - \frac{1}{3}S(S+1)\right) + R_E\left(\hat{S}_x^2 - \hat{S}_y^2\right)$. Where R_D and R_E are defined in terms of R the same way D and E are in terms of D.

Then, we may write H_{Stark} in this basis as

$$\begin{aligned} H_{\text{Stark}} = \vec{E} \cdot \Bigg(& R_D \left(\hat{S}_z^2 - \frac{1}{3} S(S+1) \right) + R_E \left(\hat{S}_x^2 - \hat{S}_y^2 \right) \\ & + R_{xy}(\hat{S}_x \hat{S}_y + \hat{S}_y \hat{S}_x) + R_{xz}(\hat{S}_x \hat{S}_z + \hat{S}_z \hat{S}_x) + R_{yz}(\hat{S}_y \hat{S}_z + \hat{S}_z \hat{S}_y) \Bigg). \end{aligned} \tag{2.55}$$

The final step is to apply symmetry arguments, which in the case of C_{3v} as required for this work reduces the Hamiltonian again to

$$\begin{aligned} H_{\text{Stark}} = & R_{113} \left(E_x(\hat{S}_x \hat{S}_y + \hat{S}_z \hat{S}_x) + E_y(\hat{S}_y \hat{S}_z + \hat{S}_z \hat{S}_y) \right) \\ & - R_{2E} \left(E_x(\hat{S}_x \hat{S}_y + \hat{S}_y \hat{S}_x) + E_y(\hat{S}_x^2 - \hat{S}_y^2) \right) \\ & + R_{3D} \left(\hat{S}_z^2 - \frac{1}{3} S(S+1) \right) \end{aligned} \tag{2.56}$$

Finally, the coefficient R_{113} represents a mixing of the $m_S = 0$ and $m_S = \pm 1$ states which have an energy splitting of $\mathcal{O}(10^9)$ Hz. The Stark energies are $\sim \mathcal{O}(10^3)$Hz and of at least second order, thus may be ignored [25].

This allows us to write our Hamiltonian in the final form as

$$H_{\text{Stark}} = d_{\parallel} \left(\hat{S}_z^2 - \frac{1}{3} S(S+1) \right) - d_{\perp} E_y(\hat{S}_x^2 - \hat{S}_y^2) + d_{\perp} E_x(\hat{S}_x \hat{S}_y + \hat{S}_y \hat{S}_x) \tag{2.57}$$

where we have the axial contribution as $d_{\parallel}$ and the off-axis contribution as $d_{\perp}$ to match the convention of existing literature.

By direct comparison to 2.45 it is easy to see the first two terms of 2.57 will contribute to the effective ZFS.

In general longitudinal fields along the defect's symmetry axis result in equal shifts of all levels, whereas transverse fields split the orbitals into two branches whose energy difference grows with increasing field.

Track refs in git history

2.11 Total Hamiltonian

We may thus, by disregarding the nuclear interactions consider our total Hamiltonian for $s = 1, 3/2$ systems for this work to be

$$H = H_{\text{SO}} + H_{\text{ZFS}} + H_{\text{Zeeman}} + H_{\text{Stark}} \tag{2.58}$$

using

$$H_{\text{ZFS}} = D \left(\hat{S}_z^2 - \frac{1}{3} S(S+1) \right) + E(\hat{S}_x^2 - \hat{S}_y^2) \tag{2.59}$$

$$H_{\text{Zeeman}} = g \mu_B \hat{\vec{S}} \cdot \vec{B} \tag{2.60}$$

$$H_{\text{Stark}} = d_{\parallel} E_z \hat{S}_z^2 - d_{\perp} E_x(\hat{S}_x^2 - \hat{S}_y^2) + d_{\perp} E_y(\hat{S}_x \hat{S}_y + \hat{S}_y \hat{S}_x) \tag{2.61}$$

2.12 Spin Hamiltonian

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2.13 Strain and Pressure

The effect of a strain is treated as an effective electric field

2.14 Quantum Sensing

Quantum sensing involves using a qubit system acting as a quantum sensor that interacts with an external variable of interest, such as a magnetic field, electric field, strain or acoustic wave, or temperature [26].

Quantum sensors have a higher sensitivity within a nanoscale or microscale sampling volume compared to a fully classical counterpart which would require higher field densities or higher volume interrogation to be effective.

[27]

[28]

[29]

[30]

2.14.1 DiVincenzo Criteria

To construct a working quantum sensor with any candidate system, DiVincenzo and Degen outlined a set of three necessary conditions that must be followed [31, 32, 33]

1. The quantum system must have discrete resolvable energy levels (or an ensemble of two-level systems with a lower energy state $|0\rangle$ and an upper energy state $|1\rangle$) that are separated by a finite transition energy.

2. It must be possible to initialise the quantum sensor into a well-known state and to read out its state.

3. The quantum sensor can be coherently manipulated, typically by time-dependent fields.

Spin defects are mostly paramagnetic and radiative point defects (or colour centres). Colour centres possessing a non-zero electron spin are excellent candidates for optical spin quantum bits (qubits) [26].

Colour centres can produce detectable luminescence even at room temperature. Optical radiation is generally used as a readout but the excitation can also be used for spin manipulation and control.

[32] [31]

2.14.2 Crystal Defects

[34]

[35]

Quantisation

Polarisation

[36]

Coherent Manipulation

[12]

[37]

[38]

[10]

[39]

[40]

Efficient Readout

[41]

[42]

[43]

[44]
[45]

2.14.3 Coherence

[11],[46], [47] [48], [49]
[50]

Spin Relaxation

Dephasing

Hahn Echo

[51]

Example: NV Diamond

2.14.4 Sensitivity

[52]
[53]
[54]
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Most of the SiC colour centres have a residual spin and therefore all could be in principle used in quantum sens- ing. However, they can be distinguished and grouped by their ground state spin value and the zero field (magnetic) split- ting (ZFS), which defines their properties and the different methods for their initialisation, control, and read-out. Colour centres with the high-spin ground state (S = 1, 3/2) can be used as two or three levels quantum systems (figure 1(c)). They can be controlled optically and using a microwave (MW) or radio frequency (RF) excitation due to the higher sensitivity to the presence of the magnetic field.

The spin Hamiltonian of an S = 3/2 elec- tron spin defect within a nuclear spin bath can be written as:

$$\hat{H} = \underbrace{g\mu_B \hat{\mathbf{S}} \cdot \mathbf{B}_0}_{H_{\text{Zeeman}}} + D\left(\hat{\mathbf{S}}_z^2 - \frac{S(S+1)}{3}\right) + E(\hat{\mathbf{S}}_x^2 - \hat{\mathbf{S}}_y^2) + \sum_j \hat{\mathbf{S}}_i \cdot \mathbf{A}_{ij} \cdot \hat{\mathbf{I}}_j \qquad (2.62)$$

where g is the isotropic centre specific Lande factor (g = 2.0028), µB is the Bohr magneton, B0 is the external magnetic field, D and E account for the zero magnetic fields splitting for the axial (along the spin polarisation axis) or the off-axis component of the spin defect operator $\hat{S}$ = (Sˆx , Sˆy , $\hat{S}$z) respect- ively. Aij is the hyperfine

tensor that describes the central spin coupling to many nuclear spins indexed by j with spin operat- ors Iˆj .

2.15 Spin Population

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2.16 Optical Polarisation

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2.17 ODMR

[58]

[59]

2.18 Silicon Carbide

SiC is considered an excellent semiconductor material for high-power, high-temperature, and high-frequency electronics [60, 61]. Studies have demonstrated SiC's potential as a host material for qubits, which enables the development of quantum sensors.

A qubit system is, in the simplest terms, as a two-level system. The power of a qubit lies in quantum coherence and/or temporal superposition of quantum states which allow for computation or manipulation with no classical analogue before being collapsed back to a measurement basis.

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2.18.1 Production of SiC

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2.18.2 Colour Defects in SiC

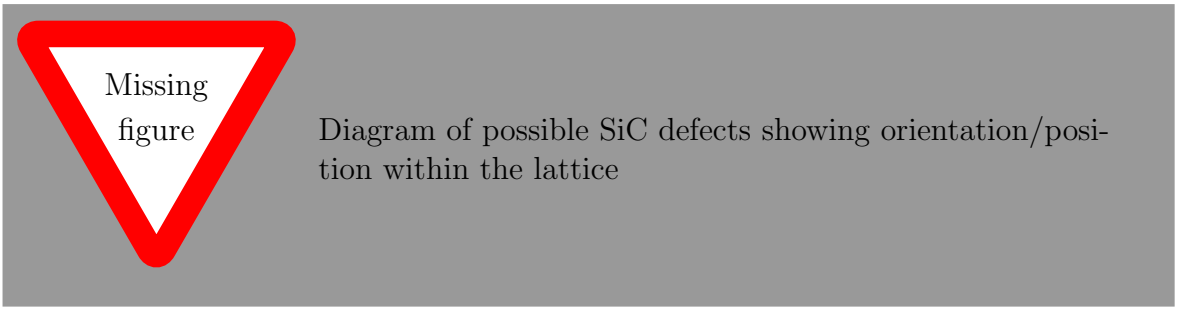

Diagram of possible SiC defects showing orientation/position within the lattice

Figure 2.1:

Electronic Structure

Charge State

Spin System

2.19 Multimodal Sensors

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Chapter 3

Design

In this chapter we will provide an overview of how the defects in SiC can be used for magnetometry, thermometry and electrometry in isolation. We will then develop a framework where by combining specific defects we may simultaneously measure multiple parameters.

3.1 $S = 1$ Magnetometry

We begin by considering a triplet state, that is a $S = 1$ system.

Under the influence of a magnetic field, the Hamiltonian can be expressed as:

$$H = H_{\mathrm{D}} + H_{\mathrm{Z}} \tag{3.1}$$

Here the labels D and Z describe the electron spin-spin interactions and the Zeeman interaction with an external magnetic field.

They have the following forms:

$$H_{\mathrm{D}} = DS_z^2 + E(S_x^2 + S_y^2) \tag{3.2}$$

$$H_{\mathrm{Z}} = g\mu_B \sum_j^{x,y,z} B_j \cdot S_j \tag{3.3}$$

$$\tag{3.4}$$

Spin-Spin Interaction

The E and D in equation 3.2 represent the fine structure constants of the spin system, describing the spin-spin interaction and S_j the corresponding spin operators in x,y and z-direction.

D is non-zero in system with axis of threefold (or other manifold) symmetry. The definiteness, orientation and magnitude of D is dependent on the specific spin system being studied.

E occurs when there is a distortion of the point group symmetry, for example strain or an $\vec{E}$ field. Similarly, the value of E is a characteristic of the nature of the distortion and the specifics of the spin system being studied.

Zeeman Interaction

B_j in equation 3.3 is the magnetic field along the x, y and z direction, g is the g-factor of the vacancy and μ_B the Bohr-Magneton.

By combining H_D and H_Z we find

$$H = DS_z^2 + E(S_x^2 + S_y^2) + g\mu_B \sum_j^{x,y,z} B_j \cdot S_j \tag{3.5}$$

The $S = 1$ spin operators S_j in matrix representation are

$$S_x = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, S_y = \frac{i}{\sqrt{2}} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & -1 \\ 0 & 1 & 0 \end{pmatrix}, S_z = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}. \tag{3.6}$$

Then, aligning the magnetic field (with strength B_0) along the z-axis (the quantisation axis), the reduced Hamiltonian will have the form

$$H = \begin{pmatrix} D + B_0 & 0 & E \\ 0 & 0 & 0 \\ E & 0 & D - B_0 \end{pmatrix}, \tag{3.7}$$

with Eigenvalues

$$E_x = E_y = D \pm \sqrt{B_0^2 + E^2},\ E_z = 0. \tag{3.8}$$

The corresponding non-normalised Eigenvectors are then

$$|X\rangle = \frac{1}{E}\left(B_0 + \sqrt{B_0^2 + E^2}\right)|{+1}\rangle + |{-1}\rangle \tag{3.9}$$

$$|Y\rangle = \frac{1}{E}\left(B_0 - \sqrt{B_0^2 + E^2}\right)|{+1}\rangle + |{-1}\rangle \tag{3.10}$$

$$|Z\rangle = |0\rangle, \tag{3.11}$$

with

$$|1\rangle = \begin{pmatrix} 1 & 0 & 0 \end{pmatrix}, |0\rangle = \begin{pmatrix} 0 & 1 & 0 \end{pmatrix}, |{-1}\rangle = \begin{pmatrix} 0 & 0 & 1 \end{pmatrix}, \tag{3.12}$$

the Eigenvectors for H with $E = 0$.

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In the case where $E \ll B_0$ the Eigenvectors are well described by the bases $|0\rangle$ and $|{\pm}1\rangle$.

For an arbitrary external magnetic field, H can be expressed using spherical coordinates:

$$H = \begin{pmatrix} D + B_0 \cdot \cos\theta & \frac{B_0}{\sqrt{2}} \cdot e^{-i\cdot\varphi} \cdot \sin\theta & E \\ \frac{B_0}{\sqrt{2}} \cdot e^{i\cdot\varphi} \cdot \sin\theta & 0 & \frac{B_0}{\sqrt{2}} e^{-i\cdot\varphi} \cdot \sin\theta \\ E & \frac{B_0}{\sqrt{2}} \cdot e^{i\cdot\varphi} \cdot \sin\theta & D - B_0 \cdot \cos\theta \end{pmatrix} \tag{3.13}$$

Here, we transformed the magnitude of the arbitrary magnetic field into spherical coordinates as

$$B_x = B_0 \cos\varphi \sin\theta \tag{3.14}$$
$$B_y = B_0 \sin\varphi \sin\theta \tag{3.15}$$
$$B_z = B_0 \cos\theta \tag{3.16}$$

with θ the azimuthal and φ the polar angle. Then using equations 3.5 and 3.6 we compute 3.13.

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It immediately follows from the characteristic equation that Eigenvalues λ satisfy

$$0 = \lambda^3 - 2\cdot\lambda^2\cdot D + \frac{D\cdot B_0^2}{2} + \lambda(D^2 - E^2 - B_0^2) - \frac{1}{2}B_0^2 \underbrace{\left(D\cdot\cos(2\theta) - 2\cdot E\cos(2\varphi)\cdot\sin(\theta)^2\right)}_{\Delta_{\varphi\theta}} \tag{3.17}$$

3.1.1 $\vec{B}$ Parallel to Defect Axis

The simplest implementation of the magnetometer is when the applied magnetic field, B_0 is parallel to the defect axis.

In this case, the entire magnitude of the field contributes to the Zeeman splitting of the energy level. This means in the CW-ODMR spectra the difference between the two frequencies $f_1 > f_2$ is directly proportional to B_0 and related as detailed in Section 2.4.

$$f_1 = D + \gamma B_0, \quad f_2 = D - \gamma B_0$$

It is then straightforward to calculate B_0 using

$$B_0 = \frac{f_1 - f_2}{2\gamma}$$

which is visualised for the DNV system in figure 3.1.

$\vec{B}$ at Angle θ to Defect Axis

The Zeeman effect is proportional to $\cos\theta$, thus, when $\vec{B}$ is perpendicular to the defect axis the Zeeman effect reduces to zero, varying the azimuthal angle θ is effectively the same as scaling B_0 by $\cos\theta$.

3.1.2 $S = 1$ Vector Magnetometry

Vector magnetometry with a $S = 1$ system can be achieved by comparing the relative intensities from defects known to be at specific angles.

For example, in diamond the nitrogen vacancy is aligned with the tetragonal crystal structure and thus may take one of four orientations as illustrated in Figure 3.3.

The 4 possible DNV orientations in the lattice are 111, $1\overline{1}\overline{1}$, $\overline{1}1\overline{1}$ and $\overline{1}\overline{1}1$. Once the projections of the magnetic field along these axes have been measures, we reconstruct the magnetic field in the laboratory frame.

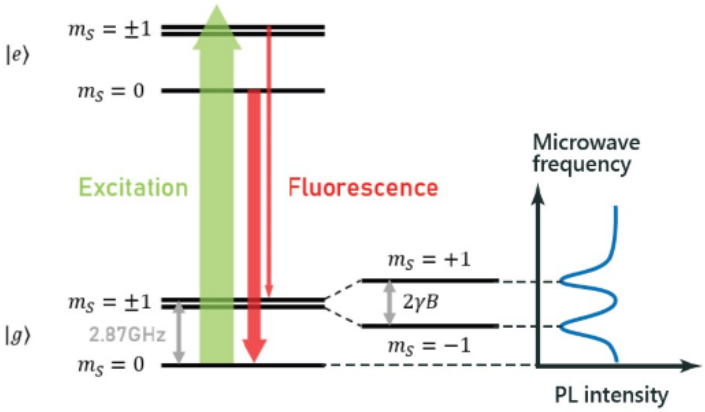

Figure 3.1: Magnetometry with $\theta = 0$. Left shows the lifting of degeneracy of the spin system energy levels with the applied $\vec{B}$ field. Right shows the corresponding ODMR spectra and two EPR frequencies [1].

The ODMR sprectrum for a sample of diamond with approximately equal distribution of the four defect orientations.

The measured field components m_i do not directly give the magnetic field B_i, but are affected by some noise in- herent to the measurement which is accounted for using a maximum-likelihood method.

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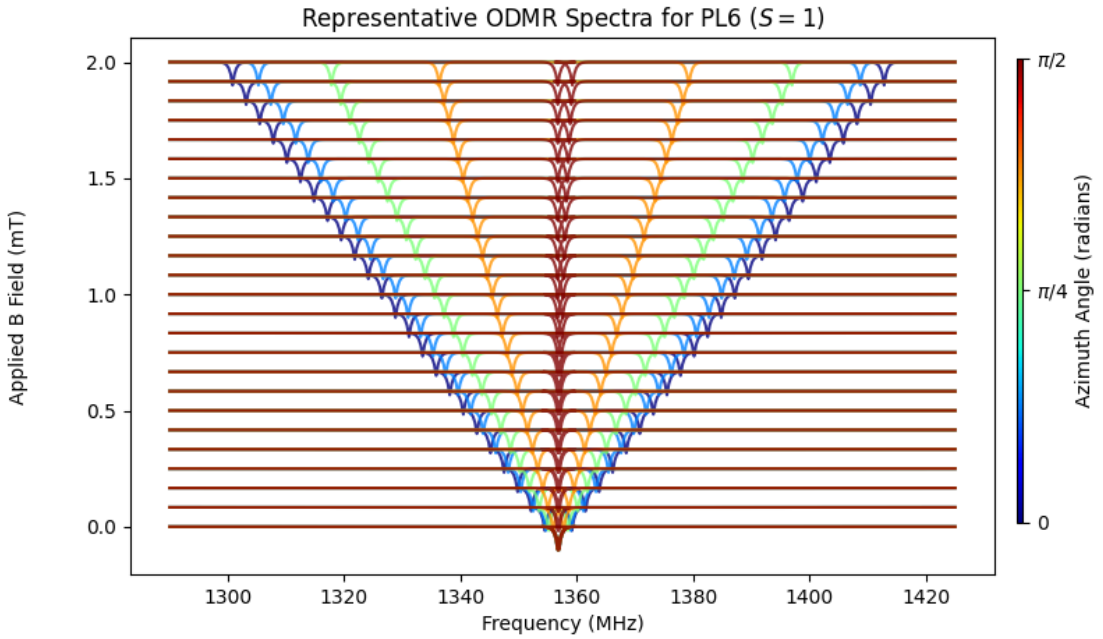

Figure 3.2: ODMR/Energy level plot showing the reduction of the effective parallel $\vec{B}$ field with increasing θ.

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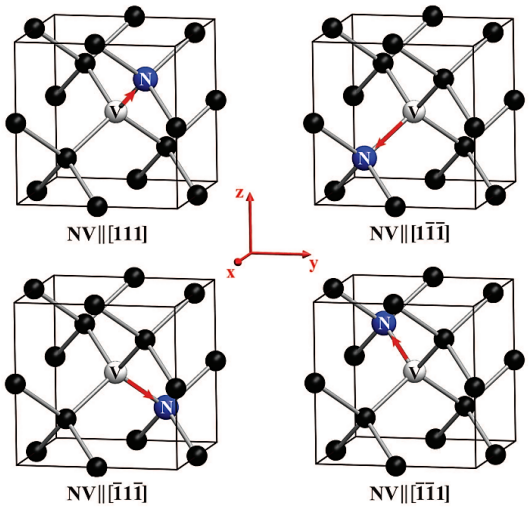

Figure 3.3: Diagram showing the four possible orientations of NV centers in diamond [2].

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3.2 $S = 3/2$ Magnetometry

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If the ZFS interaction of the $S = 3/2$ defect is sufficiently strong, the eigenvalues of the spin Hamiltonian show a strong dependence on the orientation of the applied magnetic field.

This induces a non-linear shift of resonance transitions in EPR frequencies, which is seen in the ODMR spectra. This allows information about the applied external magnetic field to be extracted from ESR spectra provided the ZFS is known.

In zero magnetic field the V_{Si} V2 vacancy has an ODMR line maximum around 70 MHz with very weak dependence on temperature. That is, the ZFS parameter is known and resistant to the environmental influence of temperature.

In a $S = 3/2$ spin system the orientation related terms are, like for $S = 1$ systems, in the eigenvalue equation. This results in the orientation dependent shift of EPR frequencies which are not explained by $g\mu_B B_0$ as they are for $S = 1$.

Therefore in order to reconstruct the energy eigenstates we must use the observed resonant energies. There are $2S + 1$ states for a system with spin S from which $2S$ transition frequencies may be found.

For the V2 V_{Si}, $E \ll D$ and a uniaxial symmetry exists therefore the Hamiltonian for the system is given as in equation **??**. Here we use the 4-dimensional $S = 3/2$ matrix representation

ref correct hamiltonian

(3.18)

Add spin 3/2 matrices

find ref

Add hamiltonian matrix

For this defect, using the same polar co-ordinate conversion as in section ?? we may write the Hamiltonian in matrix form and find the eigenvalue equation

$$\lambda^4 - \left(2D^2 + 6E^2 + \frac{5}{2}(g\mu_B B_0)^2\right)\lambda^2 - 2(g\mu_B B_0)^2\left(D(3\cos^2\theta - 1) + 3E\sin^2\theta\cos 2\varphi\right)\lambda$$
$$+ \frac{9}{16}(g\mu_B B_0)^4 + D^4 - \frac{1}{2}D^2(g\mu_B B_0)^2 - D^2(g\mu_B B_0)^2(3\cos^2\theta - 1) + 3E^2(3E^2 + 2D^2)$$
$$+ E(g\mu_B B_0)^2(6D\sin^2\theta\cos 2\varphi + \frac{9}{2}E\cos 2\theta) = 0 \tag{3.19}$$

Considering B_0 componentwise we may find [87] for B_0 parallel to the defect axis

$$\lambda = \frac{1}{2}g\mu_B B_0 \pm \sqrt{(D + g\mu_B B_0)^2 + 3E^2} \text{ or, } \lambda = -\frac{1}{2}g\mu_B B_0 \pm \sqrt{(D - g\mu_B B_0)^2 + 3E^2}. \tag{3.20}$$

For B_0 perpendicular to the defect axis we find:

$$\begin{aligned}\lambda &= \frac{1}{2}g\mu_B B_0 \pm \sqrt{(g\mu_B B_0)^2 + D^2 + 3E^2 - (D - 3E)g\mu_B B_0} \text{ or,}\\ \lambda &= -\frac{1}{2}g\mu_B B_0 \pm \sqrt{(g\mu_B B_0)^2 + D^2 + 3E^2 + (D - 3E)g\mu_B B_0}.\end{aligned} \tag{3.21}$$

We may write the general equation for the eigenvalues as

$$\sum_{n=0}^{2S+1} C_n \lambda^n = 0 \tag{3.22}$$

we then substitute each eigenvalue λ_i into this general expression to obtain $2S + 1$ equations.

The goal is now to remove all λ_i terms by considering instead the transition frequencies between eigenstates, which are observed in the ODMR spectra. The energy states are not in general sorted with respect to the energy values, so we use the convention that $\lambda_i > \lambda_{i-1}$.

To reduce our number of equations to $2S - 1$ we make the substitutions

$$\lambda_i + \underbrace{\lambda_{i+1} - \lambda_i}_{f_{i+1,i}} = \lambda_{i+1}, \qquad \lambda_i - \underbrace{(\lambda_i - \lambda_{i-1})}_{f_{i,i-1}} = \lambda_{i-1}$$

for each $i = 2, \ldots, 2S$ and calculate both

$$\sum_{n=0}^{2S+1} \frac{C_n\left((\lambda_i + f_{i+1,i})^n - \lambda_i^n\right)}{C_{2S+1}} = 0 \text{ and } \sum_{n=0}^{2S+1} \frac{C_n\left((\lambda_i - f_{i,i-1})^n - \lambda_i^n\right)}{C_{2S+1}} = 0$$

to find two new simultaneous equations

$$\sum_{n=0}^{2S} C'_{i,n}\lambda_i^n = 0 \text{ and } \sum_{n=0}^{2S} C''_{i,n}\lambda_i^n = 0.$$

We may combine these as

$$\sum_{n=0}^{2S} \frac{C'_{i,n}\lambda_i^n}{C'_{i,2S}} - \frac{C''_{i,n}\lambda_i^n}{C''_{i,2S}} = 0$$

to obtain an equation for the eigenvalue of the energy eigenstate $|i\rangle$ where $i = 2, \ldots, 2S$:

$$\sum_{n=0}^{2S-1} C_{i,n}^{(2S-1)}\lambda_i^n = 0. \tag{3.23}$$

This process is repeated until only one linear equation exists for each eigenvalue, which may be expressed in terms of resonant energies. $f_{i,i-1}$ can then be substituted to find expressions for all other eigenvalues.

For the V2 V_{Si}, we obtain equations for λ_2 expressed in terms of $f_{2,1}, f_{3,2}$ and λ_3 expressed in terms of $f_{3,2}, f_{4,3}$. Finally, using $f_{3,2} = \lambda_3 - \lambda_2$ we find formulas for each eigenvalues in terms of the resonant frequencies:

$$\lambda_1 = -\frac{3}{4}f_{2,1} - \frac{1}{2}f_{3,2} - \frac{1}{4}f_{4,3} \tag{3.24}$$

$$\lambda_2 = \frac{1}{4}f_{2,1} - \frac{1}{2}f_{3,2} - \frac{1}{4}f_{4,3} \tag{3.25}$$

$$\lambda_3 = \frac{1}{4}f_{2,1} + \frac{1}{2}f_{3,2} - \frac{1}{4}f_{4,3} \tag{3.26}$$

$$\lambda_4 = \frac{1}{4}f_{2,1} + \frac{1}{2}f_{3,2} + \frac{1}{4}f_{4,3}. \tag{3.27}$$

We substitute one of these expressions into one of the equations of the form of equation 3.23 and we obtain

$$\begin{aligned} 5(g\mu_B B_0)^2 = &\left(\frac{\sqrt{3}}{2}f_{4,3} + f_{3,2} + \frac{\sqrt{3}}{2}f_{2,1}\right)^2 \\ &+ (1-\sqrt{3})(f_{4,3}+f_{2,1})f_{3,2} - f_{4,3}f_{2,1} - 4(D^2+3E^2). \end{aligned} \tag{3.28}$$

A second quantity η, useful for angle resolution (next section) related to the polar and azimuthal angle is also defined

$$\eta \equiv E(2\cos^2\varphi\sin^2\theta + \cos^2\theta) + D\cos^2\theta \tag{3.29}$$

which in terms of the resonant frequencies is given by

$$\eta = \frac{4\left(8(D+3E) + 5(f_{4,3}-f_{2,1})\right)(g\mu_B B_0)^2 + (f_{4,3}-f_{2,1})\left(16(D^2+3E^2) - (f_{4,3}-f_{2,1})^2 - 4f_{3,2}^2\right)}{96(g\mu_B B_0)^2} \tag{3.30}$$

Overall, this shows that if the ZFS is known and three EPR frequencies are observed, the applied magnetic field strength can be found using equation 3.28.

3.2.1 $S = 3/2$ Angle Resolved Magnetometry

We may approximate η defined in equation 3.29 for the V2 V_{Si}, which exhibits uniaxial symmetry, i,e, $E \ll D$, to

$$\eta \sim D \cos^2 \theta. \tag{3.31}$$

By exploiting this approximation, we may also determine the polar angle that the magnetic field vector makes with the defect axis, however at this stage we may not determine anything about the x, y components of the vector.

To do so we explicitly compute η using equation 3.2 then we find the polar angle as

$$\theta = \cos^{-1} \sqrt{\frac{\eta}{D}} \tag{3.32}$$

3.2.2 $S = 3/2$ Vector Magnetometry

link reference

Vector magnetometry is achieved in the case of the DNV as described in section **??** and theoretically a similar approach is possible in SiC. There exists two distinct and differently oriented Silicon vacancies in 4H-SiC and three in 6H-SiC [88]. In practice however, in practice at least one of the defects in each polytope is difficult to observe at room temperature making this approach unsuitable for vector magnetometry.

In a general $S = 3/2$ system, ambiguity is found when computing θ using equation 3.32 as the EPR frequencies can not be mapped to specific transitions.

The following approach exploits the fact that a crossing of resonant frequencies occurs at a given angle (see figure 3.4). The method should be considered for $g\mu_B B_0 \gg 2\sqrt{D^2 + 3E^2}$ explicitly as interactions such as level anti-crossing produce a complex spectra [89] when $g\mu_B B_0 \approx 2\sqrt{D^2 + 3E^2}$ and the invariance of a particular EPR frequency when $g\mu_B B_0 \ll 2\sqrt{D^2 + 3E^2}$ makes determination of the polar angle θ impossible.

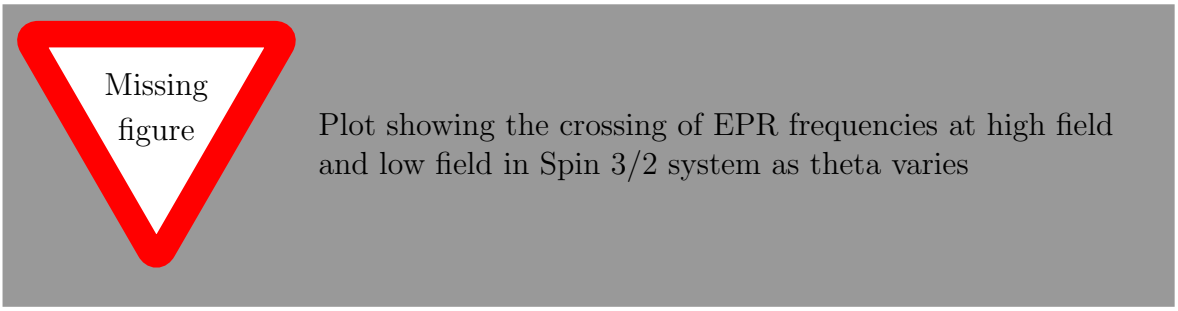

Figure 3.4:

At a high B0 field (gB B0 ZFS), B0 can be obtained from the observed ESR spectra but the polar angle cannot be determined due to the ambigu- ity of differentiating two outer transitions. In contrast, at low gB B0 (ZFS), as long as one can explicitly identify at least three transitions including the allowed lowest energy transition, the external magnetic field vec- tor can be reconstructed. In the field strength comparable to the ZFS, it is hard to find a useful scheme because very complex patterns appear

due to mixing of some of the eigenstates. In the case of the NV centers in dia- mond (ZFS/h=2.87 GHz), this missing range is around 100 mT . The VSi in SiC can fill out this gap since its ZFS is quite small (ZFS/h 100 MHz) thus this mag- netic field range can be considered as a high field range in which the three necessary transitions are well observ- able25,29, and at least the field strength can be experi- mentally determined. When the VSi in SiC is used to realize such schemes at sub-mT, if the lowest transition energy is observable by ELDOR, one can determine both B0 and without ambiguity.

[90]

3.3 Sensing Pressure*

[91] [36] [92]

3.4 $S = 1$ Thermometry

[93] [94] [76] [81] [80] [95] [82]

[83]

Distribute references properly

We can use spin defects in SiC for temperature sensing. There are two main approaches to thermometry:

ZFS Temperature Dependence. The ZFS parameters D and E may, depending on the specific spin system being studies, be sensitive to changes in temperature.

Photoluminescence. The photoluminescence of the spin system may have a dependence on temperature.

This work will focus on the first method of thermometry. Unlike $\vec{B}$ and $\vec{E}$ field sensing, there is no direction associated with temperature so the sensing regime may be simpler.

For SiC divacancies, which are triplet states, the ZFS parameter E shows no dependence on temperature. However, the ZFS parameter D varies with temperature.

The value of D for both the PL5 and PL6 defects in SiC has been measured from close to 0K to around 550K and the dependence of D has been fitted to the change in temperature. Both defects show an approximately linear relationship near room temperature which is shown in Figure 3.5

Font size

Update the T dependence of PL5 and PL6 and regen the figure. Also update temp linear range in figure caption.

In the simplest case thermometry is then achieved in the presence of a well known applied magnetic field.

The measurement stems from the change of the value of D mapped into the change of the oscillation frequency of the relative variation of the photoluminescence intensity induced by the microwave pulse sequence.

Since the degeneracy is raised symmetrically, the value of D is the average of the two resonant frequencies. The value of D can then be mapped to a temperature.

This is visualised in figure 3.6.

In practice this . Lorem ipsum dolor sit amet, consectetuer adipiscing elit. Ut pu-

Write up the Ramsey Interference methods for c-axis and basal from Castello

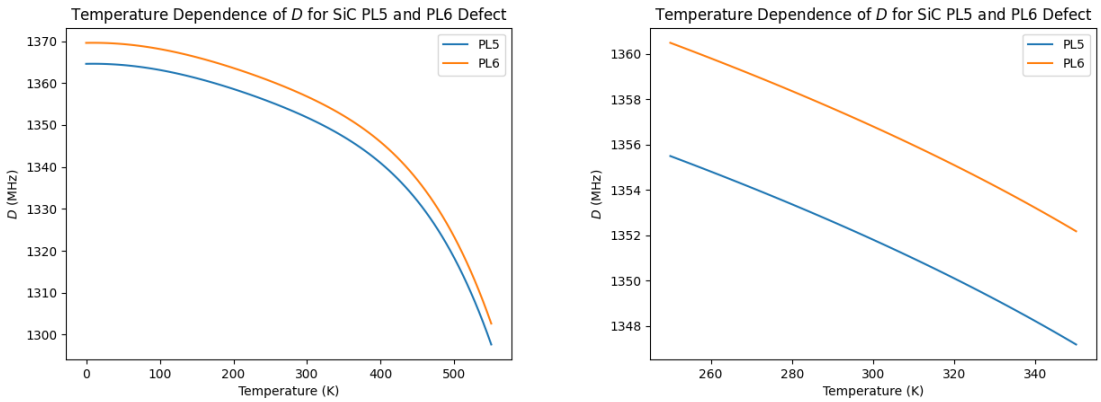

Figure 3.5: ZFS parameter D temperature dependence for the PL5 and PL6 $S = 1$ defect in SiC from 0-550 K (left) and 250-350 K (right).

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3.5 $S = 3/2$ Thermometry

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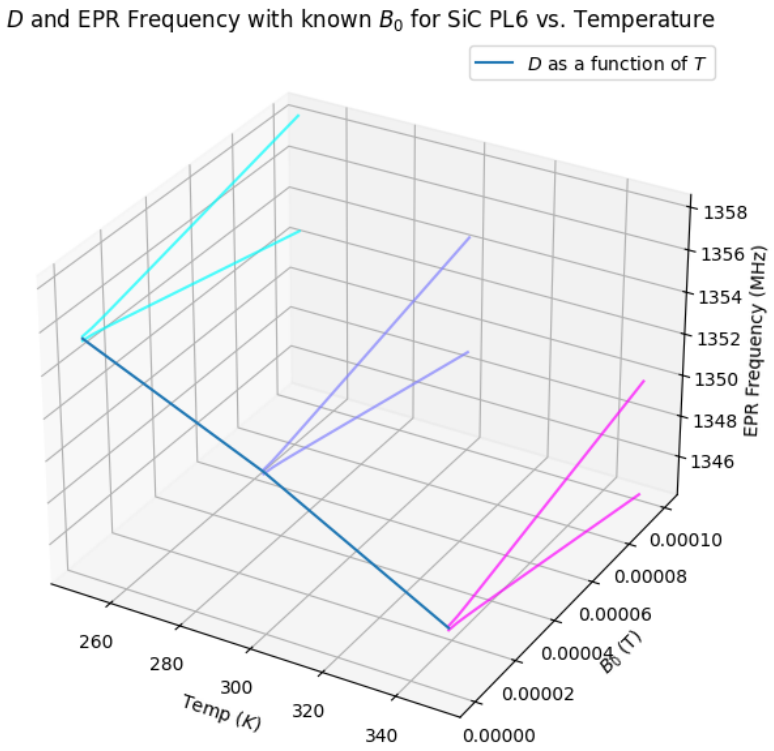

Figure 3.6:

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3.6 $S = 1$ Electometry

Add matrix Hamiltonian as well as eigenval solutions. Include the formula for $\Delta\omega$ and dicuss the diminishing returns when $B \neq 0$ or if $B \not\perp z$

3.7 $S = 3/2$ Electrometry

3.8 Sensing Strain*

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Strain and applied $\vec{E}$ field are indistinguishable so can use E techniques in shielded environment to determine strain.

3.9 Multimodality

To develop our multimodal system we will start with a very simple model with the assumption that the applied $\vec{B}$ field is parallel to the defect axis. From there we will iterate our ensemble and work to reduce the number of assumptions.

3.9.1 $|\vec{B}|$ and T

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3.9.2 Angle Resolved $|\vec{B}|$ and T

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3.9.3 $\vec{B}$ and T

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3.9.4 $|\vec{B}|$, $|\vec{E}|$ and T

The influence of an $\vec{E}$ field parallel to the defect axis is indistinguishable from the influence of a change of temperature. Similarly, the influence of an $\vec{E}$ field perpendicular to the defect axis is indistinguishable from the influence of a $\vec{B}$ field parallel to the defect axis. The exception is when ...

When B_0 is smaller than ZFS E when the effects can be distinguished

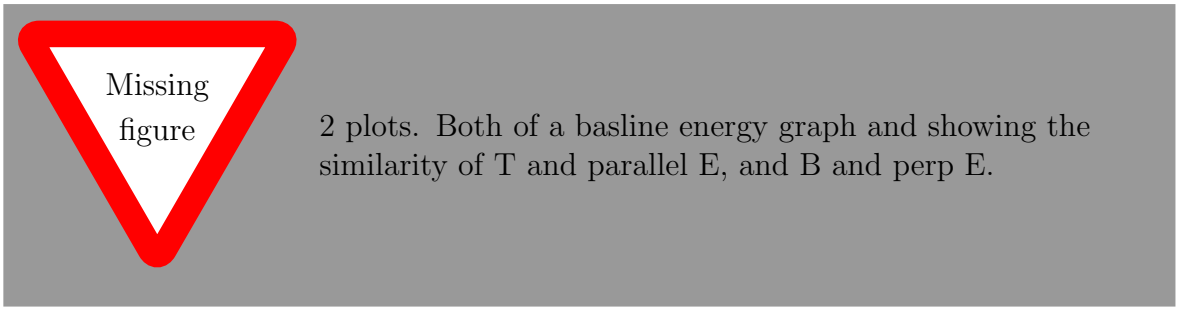

Figure 3.7:

Thus, to extend the multi-modality to include the $\vec{E}$ field we must isolate the influence of the $\vec{E}$ field from the other environmental factors.

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3.9.5 $\vec{B}$, $\vec{E}$ and T

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Chapter 4

Results and Analysis

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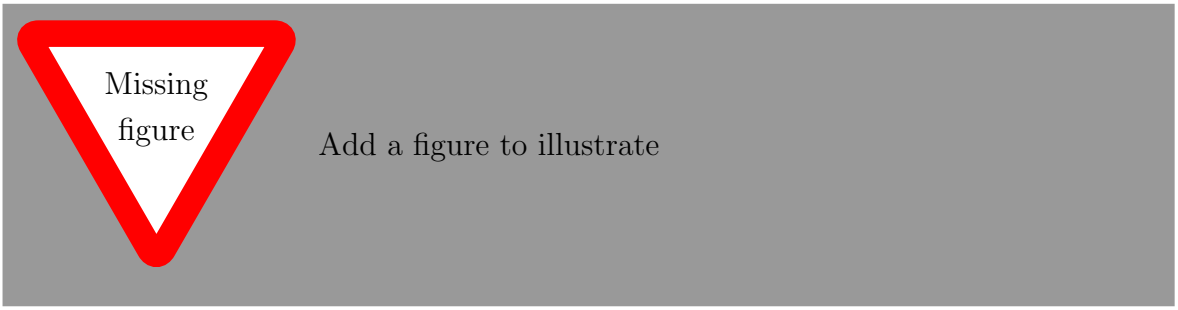

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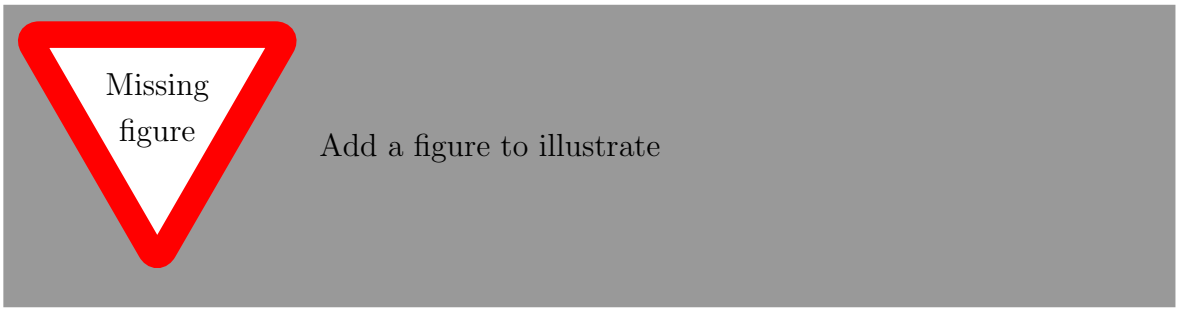

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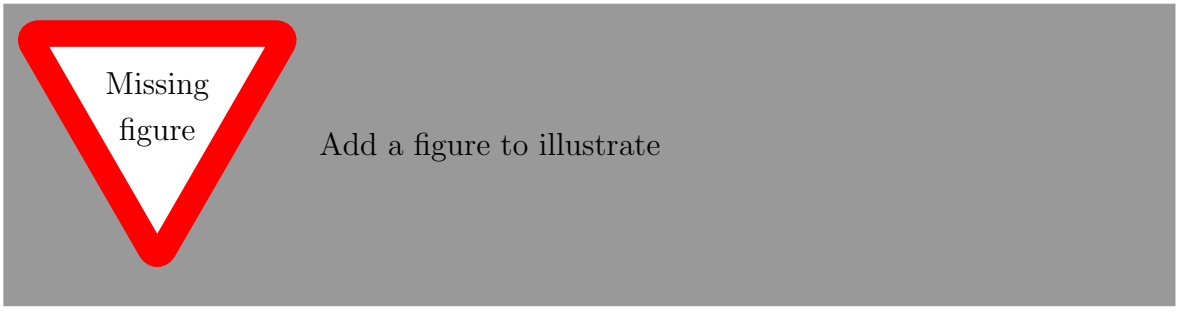

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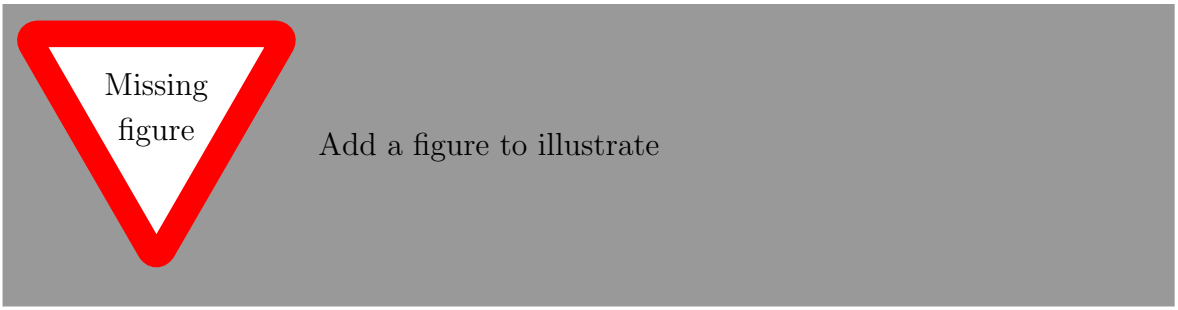

Chapter 5

Conclusions

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5.1 Multimodal Spin Based Sensors

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5.2 Wider Scientific Context

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5.3 Future Work

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