Title of my Dissertation

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MSc in Theoretical Physics

The University of Edinburgh

2023

Abstract

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

Declaration

I declare that this dissertation was composed entirely by myself.

Personal Statement

You **must** *include a Personal Statement in your dissertation. This should describe what you did during the project, and when you did it. Give an account of problems you faced and how you attempted to overcome them. The examples below are based on personal statements from MSc and MPhys projects in previous years, with (mostly-obvious) changes to make them anonymous.*

Acknowledgements

Contents

List of Tables

List of Figures

Chapter 1

Introduction

1.1 Defect Orientation

Colour centres or defects in general are part of the crystal lattice and thus have an associated orientation and direction within the lattice. This allows the definition of a **defect axis**. For example, in diamond the NV axis is defined as the vector from the vacancy towards the Nitrogen atom when the vacancy is taken as the origin of your co-ordinate system.

In a tetragonal crystal, due to symmetry there are four possible orientations of a defect within the lattice: 111, $1\overline{1}\overline{1}$, $\overline{1}1\overline{1}$ and $\overline{1}\overline{1}1$ directions.

1.2 Miller Indices

The notation for defect orientation above is known as a Miller Index, and we consider the 111 direction to be aligned with the defect axis.

This means that if we know the orientation of our crystal then we can establish the orientations of the defect axis inside. For example, using a crystal for which all surfaces belong to the $\{001\}$ lattice planes, each surface normal is aligned with a Cartesian axis. Thus, by fixing the crystal in place, there remain just **four** possible angles which a defect axis can have with respect to the crystal surface.

Calculating the scalar product of any of the surface planar directions in the family of $\{001\}$ and the four possible orientations of the defect within the lattice we find $\cos\theta = \pm 0.6$. Then, considering the physical solutions (from $0, 2\pi$) gives four possible angles that the (directed) defect axis may make with the surface of the crystal: 53.13°, 306.87°, 126.9° and 233.13° (0.927, 5.355, 2.214 and 4.069 radians respectively).

1.3 Spintronic Magnetometry

The Hamiltonian, and thus the energy, of a spin is sensitive to the magnetic field because of the Zeeman interaction.

How the electron Zeeman energy varies with magnetic field is known to a very large

precision. Therefore, by measuring the energy difference we may determine the magnetic field. This is the mechanism which enables us to use the spin of an electron as a magnetic-field sensor.

In practice, for example with diamond a fluorescence microscope to measure the electron spin resonances of an ensemble of NV centres. This allows the determination of both magnitude and direction of an external magnetic field.

1.3.1 Applied Magnetic Field

To use defects as magnetometers, we must understand the nature of their spin states when an external magnetic field is applied. From this we may determine both the amplitude and direction of the external magnetic field from the electron spin resonance frequencies of the defect.

1.3.2 Spin-1 Defect

A spin-1 defect has $S = 1$ electron spin. Therefore, it has 3 possible spin states, $m_S = 0, 1, +1$.

With no external applied magnetic field, in general, the $m_S = \pm 1$ states are degenerate, that is they have the same energy. Applying a magnetic field lifts the degeneracy and the $m_S = \pm 1$ states will have different energies, E_u and E_l (subscripts refer to "upper" and "lower"). These are equivalent to transition frequencies by $E = hf$, which we denote f_u and f_l.

The exact values of these frequencies are functions of both amplitude and direction of the magnetic field, specifically the cosine of the angle between the applied magnetic field and the defect axis θ.

Thus, by experimentally determining the transition frequencies, the magnitude and relative angle of the applied magnetic field may be determined. Details of the derivation are included in 4.2.2 and we find we may determine

$$\gamma B = \frac{1}{3}\sqrt{f_u^2 + f_l^2 - f_u f_l - D^2} \tag{1.1}$$

$$\cos^2\theta = \frac{-(f_u + f_l)^3 + 3f_u^3 + 3f_l^3}{27D(\gamma B)^2} + \frac{2D^2}{27(\gamma B)^2} + \frac{1}{3} \tag{1.2}$$

Here $\gamma = 28\text{GHz/T}$ is the gyromagnetic ratio of the electron. $D = 2.87\text{GHz}$ is the zero-field splitting of the defect ground state, that is the energy difference between $m_S = 0$ and $m_S = \pm 1$ with no external field applied.

The simplest possible case is when the defect axis aligns with the applied field for which we get a linear relationship

$$f_u = D + \gamma B \qquad f_l = D - \gamma B. \tag{1.3}$$

Include plot of the electron spin resonances vs applied magnetic field.

Chapter 2

Background Theory

2.1 Spintronics

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. Careful construction of an appropriate system allows for this magnetic moment to be initialised, manipulated and measured to infer the physical properties of the environment surrounding the system.

2.2 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 [1].

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.

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2.2.1 DiVincenzo Criteria

[6] [7]

2.2.2 Crystal Defects

[8]

[9]

Quantisation

Polarisation

[10]

Coherent Manipulation

[11]
[12]
[13]
[14]
[15]
[16]

Efficient Readout

[17]
[18]
[19]
[20]
[21]

2.2.3 Coherence

[22],[23], [24] [25], [26]
[27]

Spin Relaxation

Dephasing

Hahn Echo

[28]

Example: NV Diamond

2.2.4 Sensitivity

[29]

[30]
[31]
[32]
[33]
[34]

2.2.5 ODMR

[35]
[36]

2.2.6 Multimodal Sensors

[37]
[38]
[39]
[40]
[41]
[42]
[43]
[44]
[45]
[46]
[47]
[10]

2.3 Silicon Carbide

[48]
[49]
[50]
[51]
[52]
[53]
[53]
[54]
[55]

2.3.1 Production of SiC

[56]
[57]
[15]
[58]
[59]

2.3.2 Colour Defects in SiC

Electronic Structure

Charge State

Spin System

2.3.3 Wider Scientific Context

[60]

Chapter 3

To Sort

3.1 Spin

The magnetic moment of elementary particles is called spin.

3.2 Defect Orientation

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3.4 Lattice Symmetry

Tetragonal lattice has the

Chapter 4

Task

4.1 Brief

I think, as a start can go through section 3.2.4 in the attached PhD thesis? In particular check in details how to diagonalise the NV centre spin S=1 Hamiltonian to get Eq. 3.31? You could also do some python simulations to plot how the spin levels (i.e. the eigenvalues of the spin Hamiltonian) change with applied magnetic field.

Once we've learned this, we can apply it to other spin defects in SiC.

4.2 Work

4.2.1 Concepts and Nomenclature

Spin-Spin Interactions

Zeeman Splitting

Hyperfine Interaction

4.2.2 System Hamiltonian

The ground state of the NV^- spin system in diamond is a triplet state, thus a $S = 1$ system.

The corresponding Hamiltonian, which it seems can be generalised to an electron spin system of a defect, can be expressed as:

$$H_{\mathrm{NV}} = H_{\mathrm{D}} + H_{\mathrm{Zeeman}} + H_{\mathrm{HF}} \tag{4.1}$$

Here the labels D, Z and HF describe the electron spin-spin interactions, the Zeeman interaction with an external magnetic field and the hyperfine interaction between the nuclparallel spin I and the electron spin S of the NV.

They have the following forms:

$$H_{\mathrm{D}} = DS_z^2 + E(S_x^2 + S_y^2) \tag{4.2}$$

$$H_{\mathrm{Z}} = g\mu_B \sum_j^{x,y,z} B_j \cdot S_j \tag{4.3}$$

$$H_{\mathrm{HF}} = \vec{S} \cdot A \cdot \vec{I}. \tag{4.4}$$

Spin-Spin Interaction

The E and D in equation 4.2 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 symmetry or spin quantization axis points along the connection of the nitrogen atom and vacancy forming the defect. In bulk diamond D is around 2.87 GHz at room temperature.

The definiteness, orientation and magnitude of D is thus 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 electrical field. In bulk diamond E is typically negligibly small but especially in NDs, E can be of the order of several MHz.

Thus, 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 4.3 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, a constant.

It seems often the scaled parameter $g\mu_B$ is considered, for the NV^- system this is around 28 GHz T^{-1}, but again, will be a characteristic of the system being studied.

Hyperfine Interaction

Equation 4.4 related the nuclear spin to the electron spin via the hyperfine tensor A which has the form

$$A = \begin{pmatrix} A_\perp & 0 & 0 \\ 0 & A_\perp & 0 \\ 0 & 0 & A_\parallel \end{pmatrix}. \tag{4.5}$$

$A_\parallel$ and $A_\perp$ are the axial and non-axial hyperfine parameters which encode two different interactions.

Fermi Contact Interaction. This interaction is calculated by

$$f_A = \frac{A_\parallel + 2A_\perp}{3}. \tag{4.6}$$

Anisotropic Interaction. This interaction is found by considering both spins as magnetic dipoles is calculated by

$$d_A = \frac{A_\parallel - A_\perp}{3}. \tag{4.7}$$

For the NV^- system in diamond specifically, using the values for $A_\parallel$ and $A_\perp$ we calculate that f_A is an order of magnitude stronger than d_A for both N^{14} and N^{15}.

Reduced Hamiltonian

By combining H_D and H_Z and neglecting H_{HF} we find

why (specifically) do we get to neglect this? Can we generalise?

$$H_{NV} = DS_z^2 + E(S_x^2 + S_y^2) + g\mu_B \sum_j^{x,y,z} B_j \cdot S_j \tag{4.8}$$

The spin operators S_j in matrix representation are

$$S_x = \frac{1}{\sqrt{2}} \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, S_y = \frac{i}{\sqrt{2}} \begin{pmatrix} 0 & -1 & 0 \\ 1 & 0 & -1 \\ 0 & 1 & 0 \end{pmatrix}, S_z = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix}. \tag{4.9}$$

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

$$H_{NV} = \begin{pmatrix} D + B_0 & 0 & E \\ 0 & 0 & 0 \\ E & 0 & D - B_0 \end{pmatrix}, \tag{4.10}$$

with Eigenvalues

$$E_x = E_y = D \pm \sqrt{B_0^2 + E^2},\ E_z = 0. \tag{4.11}$$

The corresponding non-normalised Eigenvectors are then

$$|X\rangle = \frac{1}{E}\left(B_0 + \sqrt{B_0^2 + E^2}\right)|+1\rangle + |-1\rangle \tag{4.12}$$

$$|Y\rangle = \frac{1}{E}\left(B_0 - \sqrt{B_0^2 + E^2}\right)|+1\rangle + |-1\rangle \tag{4.13}$$

$$|Z\rangle = |0\rangle, \tag{4.14}$$

with

$$|1\rangle = \begin{pmatrix} 1 & 0 & 0 \end{pmatrix}, |0\rangle = \begin{pmatrix} 0 & 1 & 0 \end{pmatrix}, |-1\rangle = \begin{pmatrix} 0 & 0 & 1 \end{pmatrix}, \tag{4.15}$$

the Eigenvectors for H_{NV} with $E = 0\ldots$

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

For $E \gg B_0$, when transforming the spin operators S_j into the diagonalised system with Hamiltonian H_{NV} they read

$$\hat{S}_x^{\parallel} \propto \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \hat{S}_y^{\parallel} \propto \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & -i \\ 0 & i & 0 \end{pmatrix}, \hat{S}_z \propto \begin{pmatrix} 0 & 0 & -1 \\ 0 & 0 & 0 \\ -1 & 0 & 0 \end{pmatrix}, \tag{4.16}$$

and

$$\hat{H}_{\mathrm{NV}} = \begin{pmatrix} D + \sqrt{B_0^2 + E^2} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & D - \sqrt{B_0^2 - E^2}. \end{pmatrix} \tag{4.17}$$

Another solution for a $\pi/2$ shifted, modulating magnetic field leads to

$$\hat{S}_x^{\perp} \propto \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}, \hat{S}_y^{\perp} \propto \begin{pmatrix} 0 & -i & 0 \\ i & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \hat{S}_z \propto \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}, \tag{4.18}$$

a physical interpretation of which is that a linear modulating B-field aligned along the x-axis where strain is applied only allows transitions between the state $|X\rangle$ and $|0\rangle$, whereas fields perpendicular to the strain and the NV quantization axis only allow coupling between $|Y\rangle$ and $|0\rangle$.

For an arbitrary external magnetic field, H_{NV} can be expressed using spherical coordinates:

$$H_{\mathrm{NV}} = \begin{pmatrix} D + B_0 \cdot \cos\theta & \frac{B_0}{\sqrt{2}} \cdot e^{-i\cdot\varphi} \cdot \sin\theta & E \\ \frac{B_0}{\sqrt{2}} \cdot e^{i\cdot\varphi} \cdot \sin\theta & 0 & \frac{B_0}{\sqrt{2}} e^{-i\cdot\varphi} \cdot \sin\theta \\ E & \frac{B_0}{\sqrt{2}} \cdot e^{i\cdot\varphi} \cdot \sin\theta & D - B_0 \cdot \cos\theta \end{pmatrix} \tag{4.19}$$

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

$$B_x = B_0 \cos\varphi \sin\theta \tag{4.20}$$
$$B_y = B_0 \sin\varphi \sin\theta \tag{4.21}$$
$$B_z = B_0 \cos\theta \tag{4.22}$$

with θ the azimuthal and φ the polar angle. Then using equations 4.8 and 4.9 we compute 4.19.

It immediately follows from the characteristic equation that Eigenvalues λ satisfy

$$0 = \lambda^3 - 2\cdot\lambda^2\cdot D + \frac{D\cdot B_0^2}{2} + \lambda(D^2 - E^2 - B_0^2) - \frac{1}{2}B_0^2 \underbrace{\left(D\cdot\cos(2\theta) - 2\cdot E\cos(2\varphi)\cdot\sin(\theta)^2\right)}_{\Delta_{\varphi\theta}} \tag{4.23}$$

Chapter 5

Design

Chapter 6

Results and Analysis

Chapter 7

Conclusions

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