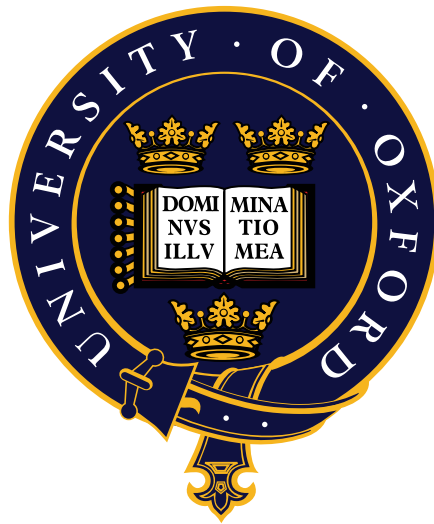


Applications of quantum coherence in condensed matter nanostructures



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Thesis submitted for the degree of
Doctor of Philosophy
at the
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This thesis is concerned with studying the fascinating quantum properties of real-world nanostructures embedded in a noisy condensed matter environment. The interaction with light is used for controlling and manipulating the quantum state of the systems considered here. In some instances, laser pulses also provide a way of actively probing and controlling environmental interactions.

The first two research chapters assess two different ways of performing all-optical spin qubit gates in self-assembled quantum dots. The principal conclusion is that an ‘adiabatic’ control technique holds the promise of achieving a high fidelity when all primary sources of decoherence are taken into account.

In the next chapter, it is shown that an optically driven quantum dot exciton interacting with the phonons of the surrounding lattice acts as a heat pump. Further, a model is developed which predicts the temperature-dependent damping of Rabi oscillations caused by bulk phonons, finding an excellent agreement with experimental data.

A different system is studied in the following chapter: two electron spin qubits with no direct interaction, yet both exchange-coupled to an optically active mediator spin. The results of this study show that these general assumptions are sufficient for generating controlled electron spin entanglement over a wide range of parameters, even in the presence of noise.

Finally, the Radical Pair model of the avian compass is investigated in the light of recent experimental results, leading to the surprising prediction that the electron spin coherence time in this molecular system seems to approach the millisecond timescale.

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List of publications

This thesis is based on the following publications:

1. *Excitonic qubits as a heat pump*. E. M. Gauger, and J. Wabnig. Manuscript in preparation.
2. *Quantum coherence and entanglement in the avian compass*. E. M. Gauger, E. Rieper, J. J. L. Morton, S. C. Benjamin, and V. Vedral. arXiv:0906.3725.
3. *Damping of exciton Rabi rotations by acoustic phonons in optically excited InGaAs/GaAs quantum dots*. A. J. Ramsay, A. V. Gopal, E. M. Gauger, A. Nazir, B. W. Lovett, A. M. Fox, and M. S. Skolnick. Physical Review Letters **104** 017402 (2010).
4. *Robust adiabatic approach to optical spin entangling in coupled quantum dots*. E. M. Gauger, A. Nazir, S. C. Benjamin, T. M. Stace & B. W. Lovett. New Journal of Physics **10** 073016 (2008).
5. *Strategies for entangling remote spins with unequal coupling to an optically active mediator*. E. M. Gauger, P. P. Rohde, A. M. Stoneham & B. W. Lovett. New Journal of Physics **10** 073027 (2008).
6. *High fidelity all-optical control of quantum dot spins: Detailed study of the adiabatic approach*. E. M. Gauger, A. Nazir, S. C. Benjamin & B. W. Lovett. Physical Review B **77** 115322 (2008).

Chapter 1

Motivation and outline

1.1 Motivation

Quantum mechanics provides the most accurate known description of physical systems at the fundamental level. Nevertheless, the profound and fascinating quantum phenomena of entanglement, quantum superposition and coherence are not usually directly observable in the macroscopic world, yet they are likely to play an important role in the implementation of powerful emerging technologies.

It is fair to say that modern technology has relied on the quantum nature of matter for several decades. For instance, the semiconductor building blocks of digital electronics and photovoltaics cannot be understood within a purely classical description. As another example, technologies such as scanning tunnelling microscopy or the electron microscope have long exploited quantum tunnelling and matter waves. The recent advances in the practical implementation (and even commercial availability) of quantum cryptography devices have taken the use of quantum systems to a new level, in the sense that the device's entire operation can be formulated in the abstract language of quantum information processing, rather than by looking at the specific description of the physical system used for its implementation. Taking this

idea further, the realisation of a full-scale quantum computer with unprecedented computational power remains the ultimate dream and goal of the field.

This new class of applications requires different quantum properties from the ones utilised in the more conventional twentieth century technologies. The desired properties usually only exist in atomic, molecular or other nanoscale systems, and they have proven easily corruptible and fragile, tending to persist only in perfectly isolated systems. On the other hand, environmental interactions, the culprit of the the aforementioned corruption, can also lead to new fascinating phenomena and challenge our understanding of the physics governing this regime.

Meanwhile, it is an intriguing thought that nature, with her head start of millions of years of evolutionary progress, might well be harnessing some of the wealth of quantum effects for biological information processing tasks, which is presently the subject of intense scientific speculation. In particular, the avian compass could be a striking example of a system with remarkably long-lived quantum coherence and entanglement. By contrast, it has been suggested that the process of photosynthesis would actually benefit from a surprisingly rapid loss of quantum correlations.

The central theme of this thesis is the study of the fascinating properties of real-world structures at the nanoscale, which are typically embedded in a noisy condensed matter environment. Many of these systems can be activated or even controlled with light, allowing us to manipulate them and, in some instances, dynamically probe and control their interactions with their respective environments. Especially, we aim to advance our understanding of how environmental interactions influence and affect the evolution of those systems, and which routes this offers towards the optimisation of particular quantum control and processing tasks.

1.2 Outline

We start this thesis with a general introduction of the concepts of quantum information processing in Chapter 2. This will provide the motivation and background for some of the research chapters of this thesis, but will be less relevant for others that focus more on particular physical properties of a specific nanosystem rather than their direct applicability to quantum information processing.

Chapter 3 contains a description of the techniques we employ to analyse and model the interaction of a small physical system with a larger environment. This forms the theoretical basis of our work and is directly relevant to all of the research chapters. Further, we here also establish how matter systems interact with an electromagnetic field and derive a generic interaction Hamiltonian for electron-phonon coupling in a crystal.

In Chapter 4, we introduce self-assembled quantum dots and their optical and electronic properties as a particularly important model system for this thesis. This last background chapter covers selected aspects of self-assembled quantum dot physics, enabling us to progress directly into the following research chapters on quantum dot systems. On the other hand, the research chapters on systems other than quantum dots will each have a few introductory pages containing a brief review of the pertinent literature and an explanation of the system.

We proceed by analysing a ‘hybrid scheme’ for optical spin manipulation in Chapter 5. In particular, we investigate and compare two different ways of performing an all-optical single qubit phase gate using a Markovian master equation technique, which comprises all principal decoherence mechanisms that typically afflict optical control operations in quantum dots.

In Chapter 6, we provide a natural extension of the single qubit phase gate study by considering an entangling two-qubit gate. Deriving a suitable Markovian master

equation, we compare two different control methods with the result that only the slower ‘adiabatic’ approach of performing the CPHASE operation has the potential of achieving a high fidelity.

Chapter 7 is concerned with the interaction between driven excitonic qubits in quantum dots and lattice phonons. In the first part, we study the processes of phonon absorption and emission by solving the system dynamics while at the same time resolving the number of phonon excitations in the environment. As a second theme of this chapter, we investigate the temperature dependence of phonon-induced damping. Comparing the theoretical predictions to experimental data provides strong evidence for acoustic phonons as the dominant excitonic dephasing mechanism at low temperature.

Chapter 8 presents a scheme for the controlled generation of entanglement between two molecular or solid state spin qubits with no direct interaction between them. Instead, the entanglement is mediated by a third party which can be activated optically. Again, we compare two different optical control techniques assessing their intrinsic robustness as well as their resilience to optical decay.

Finally, in Chapter 9 we investigate a model of the avian compass in the light of recent experimental results. We build our study on the well-established radical pair model of the bird’s ability to sense the inclination of the Earth’s magnetic field. Reconciling several crucial parameters of such a model with experimental observations then allows us to deduce an extraordinary lifetime – and coherence time – of electron spins in this molecular system.

As the research chapters address a rather broad range of different questions in different physical systems, they will each have their own conclusion section in which we summarise key results of the study and discuss interesting remaining questions and avenues for further work.

Chapter 2

Introduction to quantum information processing

We begin this thesis with an introduction to the most prominent and challenging application of a quantum technology. The quest to build a scalable quantum computer has motivated a great number of studies and produced many seminal pieces of work. Despite all these efforts, the ultimate goal still seems elusive, and we will review the difficulties encountered below. Nonetheless, many great achievements, both experimental and theoretical, have been realised, vindicating the stance that the scientific knowledge that has been gained by the endeavour to build a quantum computer constitutes important and exciting research in its own right.

2.1 Quantum Computing

Quantum Computing (QC) is a novel approach to information processing based on exploiting quantum phenomena to drive computations more efficiently. Richard Feynman may have been the first to fully realise that it takes a ‘quantum mechanical computer’ to efficiently simulate physics on the level of quantum mechanics in 1981

[1]. However, it was not until a seminal paper by David Deutsch in 1985 [2], where he introduced the notion of universal QC as a more powerful extension of the universal Turing machine, that a substantial amount of interest in the field of QC developed.

Subsequently, it has been shown that for some classes of problems, quantum information processing has the potential to outperform any known classical algorithm with respect to the required processing time [3]. The most famous example for an exponential speed-up is Shor's algorithm for the prime factorisation of large numbers [4]. A still significant quadratic speed up is provided by Grover's search algorithm for an unsorted database [5]. Moreover, QC makes the efficient simulation of quantum mechanical systems feasible [1, 6, 7]. A QC may thus provide unprecedented insight into all systems that are larger than single atoms or molecules but still small enough to behave in a genuinely 'quantum' manner. The scientific and technological implications of this application could be immense.

Regarding the question of which physical systems might be suitable for scalable quantum information processing, there are a multitude of implementation proposals, including photons [8], neutral [9] and charged atom traps [10], nuclear magnetic resonance techniques [11] and quantum dots [12] among many others. In order to achieve universal QC, coherent quantum superposition and quantum entanglement are crucial resources. These delicate quantum properties are present in small isolated systems but tend to decay rapidly when such systems interact with a macroscopic environment, a process that is known as decoherence. While perfectly flawless coherent control is not a requirement for achieving universal QC, a stringent threshold has to be met to be able to offset further errors with the help of error correcting codes, which can deal with up to one error for every $10^4 - 10^5$ calculation steps [3]. The relatively new paradigm of measurement based QC might allow conditions to be relaxed further once a suitable graph state (i.e., an entangled state of a large number of qubits) has been constructed [13]. Nonetheless, measurements and single

qubit operations still need to be performed with a high level of accuracy.

In the remainder of this chapter, we first introduce the basics of quantum information processing by contrasting it to classical information processing. We shall then give a concise overview of a selection of the more prominent implementation proposals and discuss their respective weaknesses and merits. Finally, we shall focus on the additional difficulties one faces when imposing the restraint that the architectures of the QC must be scalable. By scalable we mean that it must be possible within a given architecture to move beyond a small proof-of-principle scenario to a device that supports arbitrarily large calculations.

2.2 Carriers of information

‘Information is physical’ - this famous quote attributed to Rolf Landauer [3] fittingly epitomises the paradigm of quantum information theory. Classically, a bit of information is regarded as an abstract quantity which can assume one of two mutually exclusive states that are conventionally labelled as 0 and 1. As such, the information is completely independent of its carrier and the physics of the underlying implementation. Like its classical counterpart, quantum information processing (QIP) uses two level systems (2LS) called ‘qubits’ (for quantum bits) as a basic unit of information. The basis states of a qubit are usually referred to as $|0\rangle$ and $|1\rangle$. The quantum mechanical nature of a qubit manifests itself in the fact that a qubit is not restricted to being in one of its basis states but can also be in superposition of the basis states. Hence, a general qubit state is given by $|\phi\rangle = \alpha|0\rangle + \beta|1\rangle$ where α and β can be arbitrary complex numbers satisfying the normalisation condition $|\alpha|^2 + |\beta|^2 = 1$ [3].

As shown in Figure 2.1, the state of a qubit can be geometrically visualised as a point on the surface a unit sphere, the Bloch sphere. Up to an irrelevant global

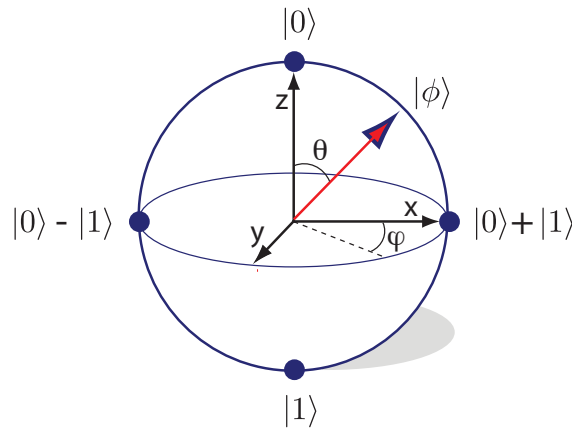


Figure 2.1: A qubit state represented as a vector on the Bloch sphere.

phase (which cannot be observed experimentally), the state of the qubit is fully characterised by two angles in this picture: θ describes the distribution of amplitude between the basis states while φ gives the relative phase between them, so that

$$|\phi\rangle = \cos \frac{\theta}{2} |0\rangle + e^{i\varphi} \sin \frac{\theta}{2} |1\rangle. \quad (2.1)$$

Here, it has been assumed that the qubit is in a pure state. When including the effects of decoherence, it can also be helpful to think of the qubit state as a vector whose direction is determined by the two angles θ and φ . The length of the vector equals one for a pure state and becomes shorter as the pure state degrades into a mixed state. A totally mixed state then corresponds to the null vector [3].

2.3 Quantum logic

Classical information processing proceeds by a concatenation of unary and binary Boolean logic operations, such as the NOT, AND and XOR operations [3]. Similarly, a quantum computation requires a sequence of single and multi-qubit gates to be applied to the qubit registers. The demand to preserve proper normalisation of the

quantum states entails that all logical quantum gates must be unitary transformations. They can be conveniently written in a matrix form with respect to the qubit basis $\{|0\rangle, |1\rangle\}$.

The only non-trivial unary gate for a classical bit flips the bit from 0 to 1 or vice versa and is called the NOT gate. Interchanging α and β is a straightforward extension of this action to a qubit with the following matrix form:

$$U_{NOT} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}. \quad (2.2)$$

However, since a qubit is determined by the two continuous variables θ and φ , it contains a greater number of degrees of freedom than a classical bit. Hence, it is only natural that there should be many more ways of manipulating the information stored in it. The phase gate,

$$U_{PHASE} = \begin{pmatrix} 1 & 0 \\ 0 & e^{i\phi} \end{pmatrix}, \quad (2.3)$$

and the Hadamard gate,

$$H = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 1 \\ 1 & -1 \end{pmatrix}, \quad (2.4)$$

are examples of important single qubit gates without classical analogues.

Let $\{|00\rangle, |01\rangle, |10\rangle, |11\rangle\}$ be the basis of a two-qubit system. The two-qubit gates of great practical relevance are the controlled NOT gate (CNOT), which flips

the target bit depending on the state of the control bit,

$$U_{CNOT} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{pmatrix}, \quad (2.5)$$

and the controlled phase gate (CPHASE),

$$U_{CPHASE} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & -1 \end{pmatrix}. \quad (2.6)$$

Contrary to the CNOT gate, the CPHASE gate is symmetric in the input states and induces a phase flip only when both input qubits are in the $|1\rangle$ state. The CNOT and the CPHASE gate are equivalent in the sense that one can be constructed from the other by additional application of Hadamard gates on the individual qubits [3]. Many implementation proposals rely on the CPHASE rather than the CNOT gate, as the symmetry of the former is more likely to reflect physical realities [3].

While one might worry that a general quantum circuit would require many more complex multi-qubit gates, this is not the case as an arbitrary unitary operation on n qubits can be efficiently decomposed into single and two qubit operations [3]. In fact, it has been shown that any entangling two qubit gate, such as the CPHASE, along with arbitrary single qubit operations constitutes a universal set of gates and is sufficient for universal QC [11]. Furthermore, it is a well-known and important result that rotations around two different axes on the Bloch sphere are sufficient for construction of an arbitrary unitary transformation of the single qubit

[3]. From a geometrical picture this is not surprising since any unitary transformation is simply a rotation on the Bloch sphere around some axis through the origin. Therefore, a concatenation of at most three rotations around two different axes, e.g. $R_Z(\theta)R_Y(\phi)R_Z(\psi)$, describes a universal unitary evolution of the Bloch vector [3].

2.4 Quantum algorithms

A computer algorithm is a list of instructions describing how to produce an output state, the result of the calculation, from a given input state. For practical execution, the algorithm needs to be re-expressed as a sequence of elementary operations on the input (qu)bits. In the case of a classical computer, the program consists of Boolean operations, whereas a QC uses a unitary evolution of the qubits with controlled interactions.

An application of quantum parallelism was first demonstrated by the Deutsch-Jozsa algorithm [14]. Consider a black box function $f : \mathcal{S} \rightarrow \{0, 1\}$, which is known to be either balanced or constant on a set of inputs $\mathcal{S}$. A constant function $f(x)$ either evaluates to 0 for all values of x or to 1 for all values of x , whereas a balanced f would give both 1 and 0 for exactly half the number of possible input values. Let N be the number of distinct values of x in the domain $\mathcal{S}$, i.e. $|\mathcal{S}| = N$. Classically, it could take up to $N/2 + 1$ separate evaluations of f until one can determine with certainty whether f is constant or balanced. On a quantum computer, however, the Deutsch-Jozsa algorithm accomplishes the same task with a single evaluation call. The crucial step is to initialise each of the n input qubits in a superposition state of $|0\rangle$ and $|1\rangle$. Hence, the evaluation is performed on a superposition of all possible input states at the same time. Notwithstanding the fact that this problem is not of great practical relevance, it nicely elucidates the power of parallel computing, which is an intrinsic feature of QC, but is hard to realise with a classical approach.

The most famous application for QC is Shor's algorithm [4]. It deals with finding the prime factors of large numbers, a task that is virtually impossible with classical computing resources due to computational complexity. The number of calculation steps needed for factoring an n digit number scales exponentially with n and to this day, no classical algorithm performing the task with polynomial scaling has been found. This fact is exploited by current internet encryption protocols for the generation of public keys, such as the RSA key exchange scheme [15]. However, the quantum factoring algorithm proposed by Shor in 1994 scales polynomially as $\mathcal{O}(n^3)$ and thus outperforms all known classical algorithms exponentially [4]. The problem of factorisation is known to be equivalent to identifying the periodicity of a mathematical function. Shor showed how to make the periodicity of this function appear in the quantum registers and then perform a quantum Fourier transform, which turns the period into an observable. The factorisation is but one example of a larger problem class called the hidden subgroup problem.

Grover's algorithmic search [5] is applicable to problems that can be phrased as looking for a specific n out of a large number of N elements, for which a black box function g (traditionally referred to as the oracle) gives $g(n) = 0$. Classically, this would on average require $N/2$ queries to the oracle. Owing to the fact that Grover's algorithm works on quantum superpositions, a quadratic speed-up is provided and the answer can be found in only $\sqrt{N}$ steps [5]. An important example of this kind of problem is a search in an unsorted database. While this is 'only' a quadratic improvement over classical search algorithms, it could still be valuable once N becomes sufficiently large.

Efficient simulation of medium and large-scale quantum systems could arguably be the QC application with the highest practical relevance, since a better understanding of quantum mechanics on the micro- and mesoscopic level might lead to exciting new technological advances through the design of novel purpose-built mate-

rials as well as tremendously facilitating nanoscale device fabrication. As Feynman pointed out [1], simulating an n -particle quantum system with k internal quantum states per particle, requires an amount of $\mathcal{O}(k^n)$ classical computational resources and is hence a formidable task. On the other hand, quantum resources proportional to only $\mathcal{O}(kn)$ can hold the same amount of information. Therefore, the resource overhead of the quantum simulator is polynomial compared to the exponential overhead of the classical approach. Feynman’s idea has since been elaborated and formalised into algorithms by Abrams and Lloyd [6, 7].

All of these algorithms have been experimentally demonstrated in small-scale ‘proof-of-concept’ scenarios. The first experimental realisations of quantum algorithms were achieved with nuclear magnetic resonance (NMR) techniques, such as the Deutsch-Jozsa Algorithm in 1998 [16] as well as the factorisation of $N = 15$ with the Shor algorithm [17] and a continuous, analogue version of the Grover algorithm [18]. The Deutsch-Jozsa algorithm has also been realised in an ion trap setup [19] and a single trapped ion simulator for a Hamiltonian of spin half particles has been demonstrated [20]. Most recently, Lu *et al.* have demonstrated Shor’s algorithm with four photonic qubits [21], while at the same time unequivocally proving entanglement of the qubits. While these formidable experimental results are certainly encouraging, there has not yet been any success in scaling up to the level of practical usefulness.

2.5 Decoherence

Quantum computing derives its advantage over classical computation from working on coherent superpositions of the input basis states. Unfortunately, quantum coherence is delicate and easily lost by the inevitable interactions of any real physical system with its environment. This process is known as decoherence and leads to a

corruption of the information in the quantum registers. Therefore, decoherence is possibly the main obstacle to full scalable QC and thus needs to be carefully considered when aspiring to devise an architecture that can sustain a large number of qubits.

Generally, there are two different processes contributing to decoherence [3]. First, a dissipative relaxation of a system from an excited state into the ground state or another state with lower energy causes a change in the populations of the computational basis. This process is characterised by the decay time, usually referred to as the T_1 time. Second, there are dephasing processes which are elastic or energy conserving but lead to a loss of phase coherence between the computational states. Dephasing happens on the so-called T_2 timescale, which is typically considerably shorter than the T_1 time and is limited by $T_2 \leq 2T_1$ [22, 23].

While it is a first priority in the design of QIP architectures to identify systems with low intrinsic decoherence rates, even a minute amount of unavoidable decoherence will eventually corrupt a lengthy computation. Fortunately, this corruption of quantum information can be reversed by using quantum error correction codes [24, 25, 26, 27]. Similar to classical error correction, these work by introducing redundancy into the encoding of the logical information. Neglecting qubit loss, there are two fundamental kinds of errors a qubit is exposed to: bit flip and phase flip errors. Analogously to the classical case, bit flip errors can be rectified by simply encoding each logical bit in three identical physical bits $|0\rangle_L = |000\rangle$ and $|1\rangle_L = |111\rangle$. Obviously, a bit flip in one of the physical qubits can now be recovered [24, 26]. Phase flip errors do not exist in classical information theory. They are most easily corrected by changing the basis used to represent the logical qubits in such a way that phase flips will then appear simply as bit flip errors [27]. This is achieved by a Hadamard rotation of the basis states $|0\rangle \rightarrow |+\rangle = (|0\rangle + |1\rangle)/\sqrt{2}$ and $|1\rangle \rightarrow |-\rangle = (|0\rangle - |1\rangle)/\sqrt{2}$. The logical qubits are then encoded as follows

$|0\rangle_L = |+++ \rangle$ and $|1\rangle_L = |-- - \rangle$. To protect the qubit against phase and bit flips simultaneously, the encodings presented above can be concatenated. This requires nine physical qubits per logical qubit and is known as the Shor code [26]. Needless to say, these error correction schemes require additional operations on the physical qubits and hence entail a significant computational overhead. Nevertheless, such correction codes show that once the error threshold of less than one error in 10^4 to 10^5 operations on the physical qubit can be realised, an arbitrarily long calculation can be performed and this is often referred to as fault-tolerant QC [3]. The threshold marks the point at which the error correction process is more likely to fix errors than to introduce them; the encoding can then be concatenated or nested to the required level to keep errors negligible for algorithms of arbitrary length. Recent theoretical developments suggest that new protocols could lower the threshold for fault-tolerance by a few orders of magnitude to make up to one error in a hundred operations tolerable [28]. An interesting example of a similarly reduced error threshold is given by the framework of topological codes that has been developed by Raussendorf and co-workers [29, 30].

Another way of dealing with decoherence is that of protecting the information in so-called Decoherence Free Subspaces (DFS). This is always practical if a subspace of the full Hilbert space can be found that is not affected by the prevalent decoherence processes [31]. Naturally for this approach, the identification of DFS heavily depends on the specifics of the physical system under consideration. Nonetheless, Zanardi and Rasetti have been able to describe the technique using an abstract mathematical formulation [32]. More recently, the concept of identifying a DFS has been successfully applied to one-way quantum computing in a theoretical work [33] and also in an experimental realisation of the Deutsch algorithm [34, 35].

2.6 Architecture and physical implementation

Having become familiar with a few of the abstract concepts of QC, we shall now turn to the more physical questions with regard to constructing a viable QC device. To address this difficult question, we shall first list a selection of prominent candidates for single qubits. Then, we shall consider the problems one faces when trying to scale up from a handful of qubits to a more useful number.

2.6.1 Physical candidates for quantum information

When looking for a suitable physical representation of a qubit, one is not necessarily restricted to a genuine 2LS. Rather, it suffices to define a suitable subsystem of two states in a larger quantum system as the computational basis. These states must of course support an application of the required gates. Another important requirement is that no population leakage occurs to states outside the computational basis.

Photonic, atomic and molecular qubits

Photons - being a natural 2LS, the polarisation of a photon is an ideal quantum information carrier. Alternatively, in the so-called dual-rail representation, a single photon in one of two possible optical modes provides another qubit realisation. Due to the weakness of their interactions with the environment (when travelling through free space or in optical fibres) photons have excellent decoherence properties. In both representations, single qubit operations are implemented by well-understood linear optical devices such as phase shifters, polarisation rotators and beam splitters [36, 37]. The conventionally suggested way of performing a CPHASE gate is based on exploiting optical non-linearities such as the Kerr effect [3]. However, in practice there is a serious flaw in this approach as photon absorption usually dominates over the much weaker Kerr effect, making the implementation of an entangling CPHASE

gate with decent fidelity impossible [38, 39].

NMR - the nuclear magnetic resonance approach uses different nuclear spins on large molecules as qubit registers. For signal amplification, an ensemble of molecules is used, meaning the calculation is always performed on a large number of molecules simultaneously, each of which is a small QC on its own. The read-out signal is then an ensemble average over up to 10^{15} molecules [3]. Gates are performed by applying magnetic field pulses. The first proposal of a CNOT gate based on NMR was suggested by DiVincenzo [11]. NMR QC experienced its heyday in the late 90s with a series of impressive experimental successes: in 1998, Chuang and co-workers were able to demonstrate a simple variant of the Deutsch-Josza algorithm [16]. A few years later, Vandersypen *et al.* successfully factorised the number 15 using Shor's algorithm [17]. In spite of these early experimental successes, there is not much scope for going to larger scales, since the small number of distinct and accessible nuclear spins on any given molecule obviously has its consequence in a very limited scalability. Although recent work has shown ways around this at the expense of some additional overhead [40, 41], there is also a deeper problem of initialising the qubits since the pseudo-pure states that are used for the computation are created from what is physically a highly mixed state of the NMR ensemble [42, 43].

Ion traps - individual ions confined in an ion trap constitute the qubits, which can be defined in two different ways: firstly, as a stable electronic ground state in the hyperfine structure of the ion. Secondly, as a ground state versus an excited optical level. Ion trap QC saw its advent when a CNOT gate was proposed by Cirac and Zoller in 1995 [10] and in the same year demonstrated by Wineland and co-workers [44]. Besides demonstration of the Deutsch-Josza algorithm [19], a simple case of a quantum simulation has also been achieved experimentally [20]. As with NMR, there are potential issues with scaling up the essentially one-dimensional geometry of the ion trap. To solve this problem, some ideas of shuttling large numbers of ions

around between active processing and storage areas of an elaborate ion trap maze have been conceived [45].

Fullerenes - embedding a single atom in a carbon cage (e.g. a nitrogen atom in a bucky ball, N@C_{60}) preserves the favourable properties of electronic and nuclear spins, whilst trapping the ion in a structure that is far easier to handle mechanically. The homogeneous carbon cage provides an exquisite shield to protect the spin from its environment [46]. A full set of quantum gates is yet to be demonstrated; however, some first attempts at robust coherent manipulation of qubits defined in this way have been successful [47].

Qubits in the solid state

NV⁻ centres in diamond and other defects in solids - the electronic ground-state spin properties of nitrogen-vacancy defects (NV⁻ centres) are well-suited for QIP [48]. A phosphorus atom embedded in a silicon matrix is another good example of a defect in a solid with properties conducive to QIP [49, 50]. Promisingly, the coherence time of these systems is typically long even at room temperature, which overcomes some of the practical problems inherent in other solid state implementations. In recent years NV⁻ centres, which are relative newcomers to the large number of QIP proposals, have been found to possess various auspicious properties, making them promising candidate systems for future research [48, 51, 52].

Quantum dots - for a long time, semiconductor structures like quantum dots have been considered promising candidates for QIP [12, 53, 54], partially owing to a large amount of manufacturing expertise and scientific knowledge already existing in the field. Quantum confinement of charge carriers in the dots leads to an atom-like discrete energy structure, opening several possibilities to define a qubit, either through the presence or absence of an electron-hole pair (exciton) in the dot¹ or as

¹As a variation of such a charge qubit, one can also define the qubit as the position of a charged

a spin qubit of a single confined electron in the conductance band.

Superconducting qubits are generally based on a Cooper pair and one or more Josephson tunnel junctions. The qubit can essentially be defined as either a charge qubit [55] or a flux qubit [56]. Recently, there has been significant progress in coupling superconducting qubits to microwave resonators, as demonstrated by several impressive QED experiments [57, 58]. While superconducting qubits are arguably the most successful and advanced solid state implementation of a qubit to date [59], they suffer from the same scalability issues that have hampered the advance of other solid state architectures, and scalable QIP is still a long way from appearing on the horizon [60].

It is worth mentioning that the list of potential qubits presented above is by no means exhaustive and a plethora of more exotic proposals exists (such as e.g. cavity QED proposals [61] etc.). However, despite the large number of systems that have been put forward as candidates for QIP, the majority of promising qubit representations fall into just three broad categories [3]: photons, electron spins and nuclear spins.

2.6.2 Architecture

Finding a good physical representation of the qubit and demonstrating high fidelity single and two-qubit gates is not sufficient for building a universal QC. The famous ‘DiVincenzo checklist’ [62] stipulates a list of requirements to be fulfilled by any proposal aspiring to enable universal QC. “So, without further ado, here are the [...] requirements for the implementation of quantum computation” [62]

1. A scalable physical system with well characterised qubits.
 2. The ability to initialise the state of the qubits to a simple fiducial state, such
-
- particle on one of two dots.

as $|000\dots\rangle$.

3. Long relevant decoherence times, much longer than the gate operation time.
4. A ‘universal’ set of quantum gates.
5. A qubit-specific measurement capability.

While some of the aforementioned systems, notably NMR and ion traps, meet most of DiVincenzo’s criteria at first glance, they do not scale very well and in these systems it has been difficult to advance beyond relatively small numbers of qubits. However, there may be a way out of some practical problems by looking at QC from an entirely different angle, which we sketch in the following.

Conventionally, the paradigm underlying QC has been the *circuit-based model* [3], which most closely resembles the standard model of classical information processing. As long as the tough threshold for fault-tolerant QC cannot be met, a gating failure that happens halfway through a calculation cannot be recovered and the calculation needs to be redone from scratch. This poses a huge problem for the circuit-based model and has proven particularly problematic in optical QC schemes, where single qubit gates can be performed with high fidelity but the entangling two-qubit gate relying on optical non-linearities is associated with significant photon loss. In 2001, Knill, Laflamme and Milburn published an important paper showing that optical QC can be driven solely by single qubit operations, beam splitters and photon detection [8], a result often referred to as the KLM scheme which is a well-known representative of the wider class of Linear Optical Quantum Computing (LOQC) schemes. The essential trick in LOQC is to use projective measurements instead of non-linear optical materials to induce an entanglement between two photons. However, this scheme relies on post-selection and is hence inherently probabilistic, which means that a substantial number of failed attempts for each two qubit gate needs to be

discarded. Therefore, it entails a huge overhead in terms of computational and physical resources. Interestingly, more recent theoretical results suggest the overhead required for entangling operations in LOQC could be reduced by exploiting weak cross-Kerr nonlinearities to achieve near deterministic CNOT gates [63, 64, 65].

Can something analogous to LOQC also be achieved with fermions in the solid state? While single electron Hamiltonians and single spin measurements are insufficient for universal QC [66], Beenakker *et al.* have shown this can in principle be overcome by additionally using the charge degree of freedom (which is outside the computational Hilbert space) [67]. A scheme for an all-measurement based entangling operation between two interacting quantum dot spin qubits has subsequently been proposed by Kolli *et al.* [68].

In addition to using measurements in lieu of an entangling gate, one can go even further and drive the entire calculation with one-qubit measurements, given a particular entangled input state (a so-called graph or cluster state), as has been shown by Raussendorf and Briegel [13]. This approach is fully equivalent to the circuit model and is known as *Measurement-Based QC* (MBQC) or as a one-way quantum computing. The latter name refers to the fact that the entanglement of the input state is used up while the quantum algorithm is performed through measurements [13]. Of course this approach cannot solve the problem of creating the required entanglement of the input state. However, by considering entanglement as a computational resource that can be generated before the calculation starts, error prone entangling gates can be dealt with far more efficiently (e.g. by distillation protocols [69, 70]) and no longer cause havoc by destroying ongoing calculations. Once the entangled cluster state exists, the calculation is performed by one-qubit measurement in different measurement bases. Since many physical systems have a preferred measurement basis, this may still require high fidelity single qubit rotations around the Bloch sphere.

How does one construct such a desirable cluster state? Numerous proposals and strategies for building and purifying cluster states exist. For instance, optical lattices are a potential candidate for creating cluster states with the required properties [71]. An influential paper by Barrett and Kok has opened another route to growing cluster states by showing how to remotely entangle two suitable matter qubits in different cavities by photon path erasure [72]. While this scheme is probabilistic with a maximal success probability of $p = 1/2$, successful entanglement is heralded by the photon measurement outcome. This procedure has the advantage of elegantly precluding scalability issues: whenever it works for two qubits, there is no fundamental physical reason why it would not work for any number, technical issues aside. Not long ago, such an entanglement between single atoms has been experimentally observed for the first time [73], albeit with extremely low success rate of 10^9 attempts per entangled pair. Meanwhile, a more elaborate protocol making use of an additional ‘broker’ qubit on each site [69] has also been developed. Additional to the Barrett and Kok ‘double-heralding’ scheme there also exists a ‘repeat until success’ scheme by Lim *et al.* [74] that allows probabilistic remote entanglement of two photon sources in different cavities.

At this point it is far from clear which architecture and computational model will eventually prove successful. Nonetheless, the concept of remotely creating entanglement between stationary matter qubits certainly offers an elegant route to avoid scalability issues if large scale cluster states suitable for one-way QC can indeed be constructed.

2.7 Summary

We have given a concise explanation of the ideas behind QC and introduced a few applications for which QC could prove beneficial. Our emphasis has been on the

endeavours to identify viable physical implementations of QIP. We have given an overview of the various physical systems which have been suggested for QIP and briefly touched upon the state-of-the-art of experimental achievements.

This concludes our review of quantum information processing and we shall, in the following chapter, focus on the techniques required for studying solid state nanostructures, with the aim of developing models which allow us to better understand and analyse the performance of quantum operations in these systems.

Chapter 3

Open quantum systems

In the previous chapter, we have seen that designing viable building blocks for quantum technologies can be an immensely difficult task since the ubiquitous corruption of information by decoherence needs to be considered on all levels. It is therefore of crucial importance to analyse and accurately predict the way the particular quantum systems are affected by their interaction with the environment. We shall in the following present the techniques that can be employed to tackle this difficult problem. Furthermore, it will become apparent that interactions with the environment are not necessarily always bad, and we shall see that coupling of electronic states to electromagnetic radiation provides an easy and fast route to manipulating quantum systems in a controlled way.

3.1 Density Operator

An isolated physical system in a pure state is described by its state vector $|\varphi\rangle$, whose time evolution is governed by the well-known Schrödinger equation [75],

$$i\hbar\frac{d}{dt}|\varphi\rangle = H|\varphi\rangle, \quad (3.1)$$

where H is the system's Hamilton operator. However, this description is insufficient when there is uncertainty about the preparation of the physical system, so that we only know it to be in one of several possible pure states $|\varphi_i\rangle$ with classical probability p_i . Therefore, we need to generalise the state vector and introduce the density operator ρ (often also called the density matrix) in its stead:

$$\rho = \sum_i p_i |\varphi_i\rangle \langle \varphi_i|. \quad (3.2)$$

In this case, one refers to the system as being in a mixed state; a particular instance of a mixed state is the state of thermal equilibrium. The choice of the $|\varphi_i\rangle$ is not unique and the density operator can indeed be written in any complete basis without changing the physics of the system. The diagonal elements of the density matrix describe the population of the basis states while the off-diagonal terms are the coherences, which indicate a coherent superposition of basis states if non-zero. Density operators have the following important properties [75]:

- Positivity: $\langle v | \rho | v \rangle \geq 0 \quad \forall |v\rangle \in \mathcal{H}$, where $\mathcal{H}$ is the system's Hilbert space.
- Hermiticity: $\rho = \rho^\dagger$.
- $\text{tr}(\rho) = 1$, this is equivalent to the demand that the classical probabilities p_i add up to one: $\sum_i p_i = 1$.

Of course, the density matrix description encompasses the case of pure systems, for which $\text{tr}(\rho^2) = 1$, in contrast to $\text{tr}(\rho^2) < 1$ for genuinely mixed states: for a system in a pure state $|\mu\rangle$, one can simply choose $|\varphi_j\rangle = |\mu\rangle$ as one of the basis vectors such that $p_j = 1$ and all other $p_{i \neq j} = 0$. Hence, it follows that $\sum_i p_i^2 = 1$, whereas in general $\sum_i p_i^2 < 1$.

The Schrödinger equation is readily extended to the von Neumann equation, which governs the time evolution of ρ ,

$$i\hbar \frac{d}{dt} \rho = [H, \rho]. \quad (3.3)$$

We will also need to calculate the expectation value of an operator A , which is given by $\langle A \rangle = \langle \psi | A | \psi \rangle$ for a pure state $|\psi\rangle$. This definition can be extended to the density operator in a straightforward manner:

$$\begin{aligned} \langle A \rangle &= \sum_i p_i \langle \varphi_i | A | \varphi_i \rangle \\ &= \text{tr}(A\rho). \end{aligned} \quad (3.4)$$

Quite often, we encounter the situation of dealing with a physical subsystem S which interacts with a larger environment E . Let the state of the combined system be described by a density matrix ρ_{SE} . We can then define the reduced density operator for the system S as

$$\rho_S = \text{tr}_E(\rho_{SE}), \quad (3.5)$$

where tr_E denotes the partial trace over the environment E , which is defined as [3]

$$\text{tr}_E(|s_i\rangle\langle s_j| \otimes |e_l\rangle\langle e_m|) = |s_i\rangle\langle s_j| \text{tr}(|e_l\rangle\langle e_m|). \quad (3.6)$$

The reduced density operator ρ_S provides a complete physical description of the subsystem S in the sense that it predicts the correct statistics for measurements made on S . Nevertheless, we still need to solve the unitary evolution of the combined system before being able to access the reduced density operator. The theory of open quantum systems [76, 77] provides a solution to this difficult problem by describing the effects of the environment on the system, without keeping track of the specific

dynamics taking place within the environment. Following this route, we shall derive an equation of motion for the system density operator ρ_S by formally integrating the von Neumann equation of the joint system and then tracing out the environmental part in the following sections.

3.2 Markovian Master Equations

In order to track the full dynamical evolution of a system experiencing continuous environmental interactions, we need to employ density matrix *master equations* (ME). In many situations the problem only becomes tractable when a couple of approximations are performed: the Born approximation assumes that the state of the system and the environment can always be written as a product, while the Markovian approximation demands that the evolution at time t must only depend on the state at this particular time and is not influenced by the previous history of the system. Fortunately, the Born-Markov approximation provides a good description for many physically relevant processes, although it is not always well-justified. The following Lindblad master equation is commonly used and represents the most general generator of a quantum dynamical semigroup [76, 78],

$$\dot{\rho}_S = -\frac{i}{\hbar}[H_S, \rho_S] + \sum_i \gamma_i \left(L_i \rho_S L_i^\dagger - \frac{1}{2}(L_i^\dagger L_i \rho_S + \rho_S L_i^\dagger L_i) \right), \quad (3.7)$$

where the L_i are the Lindblad operators describing the action of the environment on the system and the γ_i are the associated decoherence rates [78].

3.2.1 Derivation of the Markovian Master Equation

In this section, we derive a Markovian master equation similar to Eq. (3.7) based on a generic interaction Hamiltonian, which describes the coupling between the system

of interest and the environment. The derivation can be found in several textbooks and the version presented here is loosely based on Refs [76, 77]. Let the joint system be described by the density matrix ρ_{SE} whose time evolution is governed by the total Hamiltonian,

$$H = H_S + H_E + H_{SE}, \quad (3.8)$$

where H_S and H_E are the free system and environment Hamiltonians respectively, while H_{SE} describes the system-environment interaction. For simplicity, we assume for now that H has no explicit time-dependence, allowing us to move into the interaction picture with respect to both the system and bath Hamiltonian with the transformation $U(t) = \exp(-i(H_S + H_E)t/\hbar)$. In the interaction picture, the density matrix is given by $\tilde{\rho}_{SE}(t) = U(t)\rho_{SE}(t)U^\dagger(t)$ and its dynamics is governed by

$$\frac{d}{dt}\tilde{\rho}_{SE} = -\frac{i}{\hbar}[\tilde{H}_{SE}(t), \tilde{\rho}_{SE}(t)], \quad (3.9)$$

where $\tilde{H}_{SE}(t) = U^\dagger(t)H_{SE}(t)U(t)$. As we will remain in the interaction picture throughout the remainder of this section unless explicitly stated otherwise, we can safely drop the ‘ $\sim$ ’ symbol without causing confusion. We proceed by formally solving Eq. (3.9),

$$\rho_{SE}(t) = \rho_{SE}(0) - \frac{i}{\hbar} \int_0^t ds [H_{SE}(s), \rho_{SE}(s)], \quad (3.10)$$

and reinserting this solution into Eq. (3.9) while at the same time performing the partial trace over the environment, yielding

$$\frac{d}{dt}\rho_S(t) = -\frac{1}{\hbar^2} \int_0^t ds \operatorname{tr}_E [H_{SE}(t), [H_{SE}(s), \rho_{SE}(s)]]. \quad (3.11)$$

Here, we have also made the unproblematic assumption that $[H_{SE}(t), \rho_{SE}(0)] = 0$, which can be achieved by a suitable definition of the original Hamiltonian H [76]. So far Eq. (3.11) is still exact, yet a general closed form solution to this equation does not exist, and we must therefore make several approximations in the following.

The first of these is the *Born approximation* which relies on the assumption that there is no significant back-action from the small system on the much larger environmental bath,¹ so that the joint density matrix can be written as a tensor product at all times,

$$\rho_{ES}(t) = \rho_S(t) \otimes \rho_E. \quad (3.12)$$

Substituting this into Eq (3.11) results in an equation that is still non-local in time as the time evolution of $\rho_S(t)$ does not only depend on the current time t but also on the previous history. Therefore, we proceed by making the *Markov approximation* which consists of two parts. Firstly, we assume the bath's memory kernel time is short, so that we can safely replace $\rho_S(s)$ by $\rho_S(t)$ in the integral, leading to a time-local expression known as the Redfield equation

$$\frac{d}{dt}\rho_S(t) = -\frac{1}{\hbar^2} \int_0^t ds \operatorname{tr}_E [H_{SE}(t), [H_{SE}(s), \rho_S(t) \otimes \rho_E]]. \quad (3.13)$$

Secondly, we make the substitution s to $t - s$ to remove the explicit reference to the initial preparation time at $t = 0$. Provided the integrand vanishes sufficiently quickly, as is the case for rapid bath relaxation [76, 77], we can then also take its upper limit to infinity to obtain the Markovian quantum master equation:

$$\frac{d}{dt}\rho_S(t) = -\frac{1}{\hbar^2} \int_0^\infty ds \operatorname{tr}_E [H_{SE}(t), [H_{SE}(t-s), \rho_S(t) \otimes \rho_E]]. \quad (3.14)$$

¹The system may still create excitations in the bath as long as these decay sufficiently quickly compared to the system dynamics; effectively, we consider a coarse-grained time axis [76].

With the goal of finally arriving at a master equation in Lindblad form, we rewrite the Schrödinger picture interaction Hamiltonian H_{SE} in the form

$$H_{SE} = \sum_{\alpha} A_{\alpha} \otimes B_{\alpha}, \quad (3.15)$$

where the operators A_{α} act on the system and the B_{α} act on the environment. We proceed by decomposing H_{SE} into eigenoperators of the system Hamiltonian. Let the system have eigenvalues ϵ with corresponding projectors onto the eigenspace $\Pi(\epsilon)$. We can then define the operators

$$A_{\alpha}(\omega) = \sum_{\epsilon' - \epsilon = \hbar\omega} \Pi(\epsilon) A_{\alpha} \Pi(\epsilon'), \quad (3.16)$$

where the sum in the expression is extended over all energy eigenvalues ϵ' and ϵ with a fixed energy difference of $\hbar\omega$. It follows from this definition that

$$[H_S, A_{\alpha}(\omega)] = -\hbar\omega A_{\alpha}(\omega), \quad (3.17)$$

$$[H_S, A_{\alpha}^{\dagger}(\omega)] = +\hbar\omega A_{\alpha}^{\dagger}(\omega), \quad (3.18)$$

and the operators are therefore said to be eigenoperators of the system Hamiltonian. Further, they possess the following interaction picture representation:

$$e^{\frac{i}{\hbar}H_S t} A_{\alpha}(\omega) e^{-\frac{i}{\hbar}H_S t} = e^{-i\omega t} A_{\alpha}(\omega), \quad (3.19)$$

$$e^{\frac{i}{\hbar}H_S t} A_{\alpha}^{\dagger}(\omega) e^{-\frac{i}{\hbar}H_S t} = e^{+i\omega t} A_{\alpha}^{\dagger}(\omega). \quad (3.20)$$

Summing over all energy differences recovers $A_{\alpha} = \sum_{\omega} A_{\alpha}(\omega) = \sum_{\omega} A_{\alpha}^{\dagger}(\omega)$, and allows us to cast the Schrödinger picture system-bath coupling Hamiltonian into a

form,

$$H_{SE} = \sum_{\alpha, \omega} A_{\alpha}(\omega) \otimes B_{\alpha} = \sum_{\alpha, \omega} A_{\alpha}^{\dagger}(\omega) \otimes B_{\alpha}^{\dagger}, \quad (3.21)$$

which gives rise to a particularly simple interaction picture expression for H_{SE} ,

$$H_{SE} = \sum_{\alpha, \omega} e^{-i\omega t} A_{\alpha}(\omega) \otimes B_{\alpha}(t) = \sum_{\alpha, \omega} e^{i\omega t} A_{\alpha}^{\dagger}(\omega) \otimes B_{\alpha}^{\dagger}(t), \quad (3.22)$$

where $B_{\alpha}(t) = e^{iH_B t/\hbar} B_{\alpha} e^{-iH_B t/\hbar}$. Inserting this expression into Eq. (3.14) yields after some lengthy but straightforward algebra

$$\begin{aligned} \frac{d}{dt} \rho_S(t) &= \frac{1}{\hbar^2} \int_0^{\infty} ds \operatorname{tr}_E (H_{SE}(t-s) \rho_S(t) \rho_E H_{SE}(t) - H_{SE}(t) H_{SE}(t-s) \rho_S(t) \rho_E) + \text{H.c.} \\ &= \sum_{\omega, \omega'} \sum_{\alpha, \beta} e^{i(\omega' - \omega)t} \Gamma_{\alpha\beta}(\omega) \left(A_{\alpha}(\omega) \rho_S(t) A_{\beta}^{\dagger}(\omega') - A_{\beta}^{\dagger}(\omega') A_{\alpha}(\omega) \rho_S(t) \right) + \text{H.c.}, \end{aligned} \quad (3.23)$$

where H.c. denotes the Hermitian conjugate and we have introduced the bath correlation functions $\Gamma_{\alpha\beta}(\omega)$, which are defined as

$$\Gamma_{\alpha\beta}(\omega) = \frac{1}{\hbar^2} \int_0^{\infty} ds e^{i\omega s} \langle B_{\beta}^{\dagger}(t) B_{\alpha}(t-s) \rangle. \quad (3.24)$$

When the environment is in a stationary state, the reservoir correlation functions are homogeneous in time with the consequence that the $\Gamma_{\alpha\beta}(\omega)$ are time-independent and we thus have

$$\langle B_{\beta}^{\dagger}(t) B_{\alpha}(t-s) \rangle = \langle B_{\beta}^{\dagger}(s) B_{\alpha}(0) \rangle. \quad (3.25)$$

Looking at the bath correlators Eq. (3.24) allows us to consider the validity of the Markov approximation: the Markovian approximation can generally be expected to be well-justified whenever the memory kernel time of the bath is short compared to

the timescale of the system dynamics, where a short memory time manifests itself in the fast decay of the bath correlation functions.

Frequently, we can make a final approximation and neglect the non-secular terms, i.e. those terms with $\omega' \neq \omega$, in Eq. (3.23). This is an instance of a rotating wave approximation (RWA), where fast oscillating terms are neglected because they are expected to average out and consequently do not influence the systems dynamics. Physically, this is the case when the intrinsic system dynamics is much faster than the timescale τ_R of environment-induced relaxation processes; where the relevant system dynamics timescale τ_S is characterised by the inverse frequency differences $\tau_S = |\omega' - \omega|^{-1}$. Under the condition $\tau_S \ll \tau_R$, it is then sufficient to consider the following form of the Markovian master equation:

$$\frac{d}{dt}\rho_S(t) = \sum_{\omega, \alpha, \beta} \Gamma_{\alpha\beta}(\omega) \left(A_{\alpha}(\omega)\rho_S(t)A_{\beta}^{\dagger}(\omega) - A_{\beta}^{\dagger}(\omega)A_{\alpha}(\omega)\rho_S(t) \right) + \text{H.c.} \quad (3.26)$$

Finally, we transform the interaction picture master equation back to the Schrödinger picture by adding the unitary von Neumann part of the system evolution to the right-hand side of Eqs. (3.23, 3.26) [76].²

3.2.2 Interpretation of the Master Equation

To analyse how Eq. (3.26) affects the system's density matrix, it is convenient to decompose the Fourier transforms of the bath correlation functions for fixed ω as

²Note that only the off-diagonal Lindblad terms of Eq. (3.23) need to be transformed to the Schrödinger picture explicitly.

follows

$$\Gamma_{\alpha\beta}(\omega) = \frac{1}{2}\gamma_{\alpha\beta} + iS_{\alpha\beta}(\omega), \quad (3.27)$$

$$\gamma_{\alpha\beta}(\omega) = \Gamma_{\alpha\beta}(\omega) + \Gamma_{\beta\alpha}^*(\omega) = \int_{-\infty}^{\infty} ds e^{i\omega(s)} \langle B_{\beta}^{\dagger}(s) B_{\alpha}(0) \rangle, \quad (3.28)$$

$$S_{\alpha\beta}(\omega) = \frac{1}{2i} (\Gamma_{\alpha\beta}(\omega) - \Gamma_{\beta\alpha}^*(\omega)). \quad (3.29)$$

Hence, the matrix $\gamma_{\alpha\beta}(\omega)$ is positive while $S_{\alpha\beta}(\omega)$ forms a Hermitian matrix that contributes to the unitary evolution of the system. Defining the Lamb shift Hamiltonian (which commutes with the unperturbed system Hamiltonian H_S by virtue of the definition of the $A_{\alpha}(\omega)$),

$$H_{LS} = \hbar \sum_{\omega, \alpha, \beta} S_{\alpha\beta}(\omega) A_{\beta}^{\dagger}(\omega) A_{\alpha}(\omega), \quad (3.30)$$

and moving back into the Schrödinger picture, we can rewrite Eq. (3.26):

$$\frac{d}{dt} \rho_S(t) = -\frac{i}{\hbar} [H_S + H_{LS}, \rho_S(t)] + \mathcal{D}[\rho_S(t)]. \quad (3.31)$$

Typically, H_{LS} renormalises the energy levels and is often neglected because it is either only a small correction or, in other instances, it diverges [77]. The more important non-unitary contribution stems from the dissipator $\mathcal{D}[\rho_S(t)]$ of the master equation,

$$\begin{aligned} \mathcal{D}[\rho_S(t)] \equiv \sum_{\omega, \alpha, \beta} \gamma_{\alpha\beta}(\omega) & \left(A_{\alpha}(\omega) \rho_S(t) A_{\beta}^{\dagger}(\omega) - \right. \\ & \left. \frac{1}{2} \left(A_{\beta}^{\dagger}(\omega) A_{\alpha}(\omega) \rho_S(t) + \rho_S(t) A_{\alpha}^{\dagger}(\omega) A_{\beta}(\omega) \right) \right). \end{aligned} \quad (3.32)$$

To conclude this section, we note that the dissipator can in general be written in Lindblad form as in Eq (3.7) by diagonalising the matrix $\gamma_{\alpha\beta}(\omega)$.

3.3 Physical interaction Hamiltonians

3.3.1 Interaction of matter with radiation

In this section, we establish how matter interacts with a radiation field, of either a classical or a quantum nature. We begin with the description of the electromagnetic field. A non-relativistic electromagnetic field in free space can be conveniently expressed by a single vector potential $\mathbf{A}(\mathbf{r}, t)$ in the Coulomb gauge, which must satisfy [79]

$$\nabla^2 \mathbf{A} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \mathbf{A} = 0, \quad (3.33)$$

and which bears the following relation to the electric and the magnetic field:

$$\mathbf{E}(\mathbf{r}, t) = -\frac{\partial}{\partial t} \mathbf{A}(\mathbf{r}, t), \quad (3.34)$$

$$\mathbf{B}(\mathbf{r}, t) = \nabla \times \mathbf{A}(\mathbf{r}, t). \quad (3.35)$$

In the presence of such an electromagnetic field, we need to use a modified version of the Schrödinger equation to describe the evolution of a charged particle. Specifically, the momentum operator gains a term proportional to the vector potential operator ($-i\hbar\nabla \rightarrow -i\hbar\nabla + e\mathbf{A}(\mathbf{r}, t)$), so that the Hamiltonian now reads [36, 37]

$$H = \frac{1}{2m} (-i\hbar\nabla + e\mathbf{A})^2 + V(\mathbf{r}), \quad (3.36)$$

where m is the mass of the particle and $V(\mathbf{r})$ is the external potential. Substituting the time-dependent form of the momentum operator, $-i\hbar\nabla = m d\mathbf{r}/dt$, in the mixed term $-i\hbar e/m \nabla \cdot \mathbf{A}$ and dropping the quadratic term in $\mathbf{A}$, as is allowed in the regime

where the field is not too strong, yields,

$$H = -\frac{\hbar^2 \nabla^2}{2m} + e\mathbf{A} \cdot \frac{d}{dt}\mathbf{r} + V(\mathbf{r}). \quad (3.37)$$

Within the *dipole approximation*, based on the assumption that the electromagnetic field has no spatial variation across the extent of the matter system or atom, we may rewrite the second term of above Hamiltonian according to

$$\mathbf{A} \cdot \frac{d}{dt}\mathbf{r} = \frac{d}{dt}(\mathbf{A} \cdot \mathbf{r}) - \frac{d}{dt}\mathbf{A} \cdot \mathbf{r} = -\frac{d}{dt}\mathbf{A} \cdot \mathbf{r}, \quad (3.38)$$

since the first term on the right-hand side of the equation disappears in the Coulomb gauge, where

$$\mathbf{A}(\mathbf{r}, t) = \frac{1}{2}\mathbf{B}(t) \times \mathbf{r}. \quad (3.39)$$

Hence, using Eq. (3.34), our Hamiltonian takes the following form

$$H = -\frac{\hbar^2 \nabla^2}{2m} + e\mathbf{E}(t) \cdot \mathbf{r} + V(\mathbf{r}). \quad (3.40)$$

By comparison with the free Hamiltonian of the particle in the absence of an external field, we are now in a position to identify the matter radiation interaction term,

$$H_{dipole} = -\mathbf{E}(t) \cdot \mathbf{d}, \quad (3.41)$$

where we have introduced the electron dipole operator defined as $\mathbf{d} = -e\mathbf{r}$. Suppose the matter system is a 2LS with a ground state $|g\rangle$ and an excited state $|e\rangle$. The dipole operator may then be written in terms of its matrix elements $\mathbf{d}_{ij} = \langle i|\mathbf{d}|j\rangle$. Since $\mathbf{r}$ is an operator with odd parity, it follows that diagonal terms vanish,³ $\mathbf{d}_{gg} = \mathbf{d}_{ee} = 0$. Moreover, we have $\mathbf{d}_{eg} = \mathbf{d}_{ge}^*$ because the operator is Hermitian [80].

³Provided the system possesses inversion symmetry, as is normally the case for atomic systems.

3.3.2 Optical manipulation of an atomic two level system

We proceed by considering a single quantised field mode, e.g. a cavity mode, with frequency ω given by (also in the dipole approximation) [36, 80]

$$\mathbf{E}(t) = \sqrt{\frac{\hbar\omega}{2\epsilon_0 V}} \boldsymbol{\epsilon} \left(\hat{b} e^{-i\omega t} + \hat{b}^\dagger e^{i\omega t} \right), \quad (3.42)$$

where $\hat{b}, \hat{b}^\dagger$ represent the annihilation and creation operators of the field mode, and where $\boldsymbol{\epsilon}$ is the mode's polarisation. The above field operator is in the Heisenberg picture and already includes the time-dependence of annihilation and creation operators, which simply lose the $\exp(\pm i\omega t)$ terms in the Schrödinger picture. Therefore, the dipole interaction can be written as (in the Schrödinger picture)

$$H_{dipole} = -\sqrt{\frac{\hbar\omega}{2\epsilon_0 V}} \boldsymbol{\epsilon} \left(\hat{b} + \hat{b}^\dagger \right) \cdot \left(\mathbf{d}_{ge} |g\rangle\langle e| + \mathbf{d}_{ge}^* |e\rangle\langle g| \right) \quad (3.43)$$

$$= g \left(\hat{b} + \hat{b}^\dagger \right) (\sigma_+ + \sigma_-), \quad (3.44)$$

where, for simplicity, we have assumed that $\mathbf{d}_{ge}$ is real in the second line of the equation. Further, we have introduced the atomic raising operator $\sigma_+ = |e\rangle\langle g|$ and lowering operator $\sigma_- = |g\rangle\langle e|$ and defined g for notational convenience,

$$g = -\sqrt{\frac{\hbar\omega}{2\epsilon_0 V}} \boldsymbol{\epsilon} \cdot \mathbf{d}_{ge}. \quad (3.45)$$

Adding the free Hamiltonian of the field mode and the 2LS, we obtain the full Hamiltonian of the atom-photon system

$$H = -\frac{1}{2}\hbar\omega_0\sigma_z + \hbar\omega\hat{b}^\dagger\hat{b} + g \left(\hat{b} + \hat{b}^\dagger \right) (\sigma_+ + \sigma_-). \quad (3.46)$$

Here, the energy splitting between ground state $|g\rangle$ and excited state $|e\rangle$ is expressed by $\hbar\omega_0$ and written in pseudo-spin notation, where $\sigma_z = |g\rangle\langle g| - |e\rangle\langle e|$ is the standard

Pauli matrix [75, 3]. Suppose the frequency of the field mode is close to resonance with the atom, i.e. $\omega \approx \omega_0$. In this case, the terms proportional to $\hat{b}^\dagger \sigma_+$ and $\hat{b} \sigma_-$ couple states which are far off-resonance and it is therefore a good approximation to drop them, provided the coupling g is not too strong [3]. This approximation, which is equivalent to the rotating wave approximation for classical fields, leads to the famous *Jaynes-Cummings Hamiltonian*:

$$H = -\frac{1}{2}\hbar\omega_0\sigma_z + \hbar\omega\hat{b}^\dagger\hat{b} + g\hat{b}\sigma_+ + g\hat{b}^\dagger\sigma_-. \quad (3.47)$$

Hamiltonian (3.47) has the important property that it preserves the excitation number, since the coupling terms only allow the state of the system to be raised when a photon is absorbed and vice versa. For a fixed number of n excitations in the joint system, we may therefore write the Hamiltonian as a 2×2 matrix in the basis $\{|g, n\rangle, |e, n-1\rangle\}$

$$H_n = \begin{pmatrix} \hbar(\omega n - \frac{1}{2}\omega_0) & \sqrt{n}g \\ \sqrt{n}g & \hbar(\omega(n-1) + \frac{1}{2}\omega_0) \end{pmatrix}. \quad (3.48)$$

On resonance, the terms on the diagonal are equal and the eigenstates of the Hamiltonian are the entangled atom-photon states $\frac{1}{\sqrt{2}}(|g, n\rangle \pm |e, n-1\rangle)$. Initialising the system in a Fock state, i.e. in a simple number state with a given number of photons all in the same mode, will then lead to a periodic exchange of a quantum of energy between the 2LS and the field mode, the so-called *Rabi* oscillation. The Rabi frequency is given by the off-diagonal terms and depends on the number of photons in the mode [80].

Instead of the quantised field, we may also consider the interaction of an atomic 2LS with a classical field. It is an unsurprising result that both approaches coincide in the limit of a large photon occupation in the mode [80]. In the classical description,

the electric field is simply given by

$$\mathbf{E}(t) = E_0 (\boldsymbol{\epsilon} e^{-i\omega t} + \boldsymbol{\epsilon}^* e^{i\omega t}), \quad (3.49)$$

leading to the following matrix form of the combined system and dipole interaction Hamiltonian in the basis $\{|g\rangle, |e\rangle\}$

$$H(t) = \begin{pmatrix} -\hbar\omega_0/2 & E_0 \mathbf{d}_{eg}^* \cdot (\boldsymbol{\epsilon} e^{-i\omega t} + \boldsymbol{\epsilon}^* e^{i\omega t}) \\ E_0 \mathbf{d}_{eg} \cdot (\boldsymbol{\epsilon} e^{-i\omega t} + \boldsymbol{\epsilon}^* e^{i\omega t}) & \hbar\omega_0/2 \end{pmatrix}. \quad (3.50)$$

The time-dependence of the Hamiltonian makes it difficult to solve the Schrödinger equation exactly. However, the problem becomes tractable by following a commonly used procedure. First, we perform a unitary transformation $U(t)$, which takes us into a frame rotating at the same frequency as the optical field,

$$U(t) = \begin{pmatrix} e^{i\omega t/2} & 0 \\ 0 & e^{-i\omega t/2} \end{pmatrix}. \quad (3.51)$$

Since the transformation is time-dependent, we must use the following prescription for transforming the Hamiltonian:

$$\tilde{H}(t) = U^\dagger(t) H(t) U(t) + i\hbar \left(\frac{d}{dt} U^\dagger(t) \right) U(t), \quad (3.52)$$

as can easily be checked by deriving the appropriate Schrödinger equation for the transformed states $|\tilde{\psi}\rangle = U(t)|\psi\rangle$. This yields

$$\tilde{H}(t) = \begin{pmatrix} -\hbar(\omega_0 - \omega)/2 & E_0 \mathbf{d}_{eg}^* \cdot (\boldsymbol{\epsilon} + \boldsymbol{\epsilon}^* e^{2i\omega t}) \\ E_0 \mathbf{d}_{eg} \cdot (\boldsymbol{\epsilon}^* + \boldsymbol{\epsilon} e^{-2i\omega t}) & \hbar(\omega_0 - \omega)/2 \end{pmatrix}. \quad (3.53)$$

The off-diagonal elements of the Hamiltonian now each have a time-independent and an oscillatory term. When the external field is close to resonance, $|\omega_0 - \omega| \ll \omega$, these rapidly oscillating terms have little effect on the dynamics and can be neglected. This approximation is known as the *rotating wave approximation* (RWA), and leads to the time-independent *Rabi Hamiltonian*

$$\tilde{H} = \frac{\hbar}{2} \begin{pmatrix} -\delta & \Omega^* \\ \Omega & \delta \end{pmatrix}, \quad (3.54)$$

where we have introduced the *detuning* $\delta = \omega_0 - \omega$ and the *Rabi frequency* $\Omega = \frac{2}{\hbar} E_0 \mathbf{d}_{eg} \cdot \boldsymbol{\epsilon}^*$. Being time-independent, the Rabi Hamiltonian is amenable to analytical treatment and will be encountered in a similar form many times throughout this thesis.

3.3.3 Interaction with lattice phonons

Phonons are quantised excitations of vibrational modes of the crystal lattice. Electronic excitations modify the overall electrostatic potential of the crystal produced by all charge carriers, which in turn determines the equilibrium positions of lattice ions. As a consequence, we may expect a coupling between the state of an electronic excitation and lattice phonons. In the following, we will derive a generic interaction term to describe electron-phonon coupling following Ref. [81].

We begin by assuming that the interaction between an electron at position $\mathbf{r}$ and an ion at $\mathbf{R}$ only depends on the distance between them. Summing over all electrons and lattice ions yields

$$H_{ep} = \sum_{ij} V(\mathbf{r}_i - \mathbf{R}_j). \quad (3.55)$$

Let us decompose the vector $\mathbf{R}_j$ into the ionic equilibrium position, $\mathbf{R}_j^{(0)}$, and a small displacement $\mathbf{Q}_j$, so that $\mathbf{R}_j = \mathbf{R}_j^{(0)} + \mathbf{Q}_j$. A Taylor expansion of the above summation terms then yields:

$$V(\mathbf{r}_i - \mathbf{R}_j^{(0)} - \mathbf{Q}_j) = V(\mathbf{r}_i - \mathbf{R}_j^{(0)}) - \mathbf{Q}_j \cdot \nabla V(\mathbf{r}_i - \mathbf{R}_j^{(0)}) + \mathcal{O}(\mathbf{Q}^2). \quad (3.56)$$

We do not need to consider the first term $V(\mathbf{r}_i - \mathbf{R}_j^{(0)})$ of this expansion further; it is the periodic potential experienced by the electrons in the unperturbed lattice, and the Bloch function solutions to this equation are usually known. We shall neglect the higher order terms $\mathcal{O}(\mathbf{Q}^2)$ and proceed with the linear electron-phonon interaction term, which we label $V_{ep}(\mathbf{r})$.⁴

The periodicity of the lattice permits writing the potential gradient as a sum over its Fourier components $\nabla V(\mathbf{r}) = \frac{i}{N} \sum_{\mathbf{q}} \mathbf{q} V(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{r}}$. Inserting this into the linear coupling expression gives

$$V_{ep}(\mathbf{r}) = \frac{i}{N} \sum_{\mathbf{q}} V(\mathbf{q}) e^{i\mathbf{q} \cdot \mathbf{r}} \mathbf{q} \cdot \sum_j \mathbf{Q}_j e^{-i\mathbf{q} \cdot \mathbf{R}_j^{(0)}}. \quad (3.57)$$

It is now convenient to rewrite the displacement operator $\mathbf{Q}_j$ with a form given by the phonon mode creation and annihilation operators [81],

$$\frac{i}{N} \sum_j \mathbf{Q}_j e^{-i\mathbf{q} \cdot \mathbf{R}_j^{(0)}} = - \sum_{\mathbf{G}} \sqrt{\frac{\hbar}{2\mu V \omega_{\mathbf{q}}}} \boldsymbol{\xi}_{\mathbf{q}+\mathbf{G}} (\hat{a}_{\mathbf{q}} + \hat{a}_{\mathbf{q}}^\dagger), \quad (3.58)$$

where the summation $\mathbf{G}$ is over the reciprocal lattice vectors, μ is the mass density of the solid, V the lattice volume, and $\boldsymbol{\xi}_{\mathbf{q}}$ is the phonon polarisation vector of mode

⁴In the following, we make a sequence of approximations, which are collectively called the *Born-Oppenheimer approximation* (see Ref. [81] for details).

$\mathbf{q}$. The coupling term now reads

$$V_{ep}(\mathbf{r}) = - \sum_{\mathbf{q}, \mathbf{G}} e^{i\mathbf{r} \cdot (\mathbf{q} + \mathbf{G})} V(\mathbf{q} + \mathbf{G}) (\mathbf{q} + \mathbf{G}) \cdot \boldsymbol{\xi}_{\mathbf{q}} \sqrt{\frac{\hbar}{2\mu V \omega_{\mathbf{q}}}} (\hat{a}_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^{\dagger}). \quad (3.59)$$

Inserting this back into Eq. (3.55), we can simplify the resulting expression further by integrating over the charge density of the solid $\varrho(\mathbf{r})$ to arrive at

$$\begin{aligned} H_{ep} &= \int d\mathbf{r} \varrho(\mathbf{r}) V_{ep}(\mathbf{r}) \\ &= \sum_{\mathbf{q}, \mathbf{G}} M_{\mathbf{q}+\mathbf{G}} \hat{\varrho}(\mathbf{q} + \mathbf{G}) (\hat{a}_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^{\dagger}), \end{aligned} \quad (3.60)$$

where $\hat{\varrho}(\mathbf{q})$ is the charge density operator, i.e. the Fourier transform of the electron density, and $M_{\mathbf{q}}$ is a coupling matrix element defined as follows:

$$M_{\mathbf{q}} = - \sqrt{\frac{\hbar}{2\mu V \omega_{\mathbf{q}}}} V(\mathbf{q}) \mathbf{q} \boldsymbol{\xi}_{\mathbf{q}-\mathbf{G}}. \quad (3.61)$$

Bound excitonic states in semiconductor structures tend to possess fairly low energy and only couple to long wavelength phonons, which means we are justified in neglecting all contributions outside the first Brillouin zone and set $\mathbf{G} = 0$. There are two relevant coupling mechanisms for those systems, both of which can be conveniently expressed with measured phenomenological parameters. Firstly, the *deformational potential* matrix element is obtained by the writing $V(\mathbf{q})\mathbf{q} = D|\mathbf{q}|$, where D is the deformation potential coupling strength. Secondly, *piezoelectric coupling* has an imaginary matrix element $V(\mathbf{q})\mathbf{q} = iP$ with P being the piezoelectric coupling strength, so that the overall electron-phonon interaction Hamiltonian is

$$H_{ep} = \sum_{\mathbf{q}} \sqrt{\frac{\hbar}{2\mu V \omega_{\mathbf{q}}}} (D|\mathbf{q}| + iP) \hat{\varrho}(\mathbf{q}) (\hat{a}_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^{\dagger}). \quad (3.62)$$

Because these two interactions are out of phase, they do not interfere up to second order and the two mechanisms can be treated independently [81].

3.4 The quantum optical master equation

We will now apply our general master equation framework to the specific case of a simple atomic 2LS that interacts with a reservoir of optical modes. Let the system be described by the Hamiltonian $H_S = -\hbar\omega_0/2\sigma_z = \epsilon\sigma_z$ and the coupling to each mode of the reservoir be given by the Jaynes-Cummings Hamiltonian as in Eq. (3.47). Note that the environment must have many modes in order for the correlation functions to decay rapidly, so that the following technique is not appropriate for a 2LS coupled to a single cavity mode.

From Eq. (3.47), we can immediately identify the eigenstate-projected system and bath operators as required for Eq. (3.22)

$$A_+(-\omega_0) = \sigma_+ \quad \text{and} \quad B_+(t) = \sum_i g_i \hat{b}_i e^{-i\omega_i t}, \quad (3.63)$$

$$A_-(\omega_0) = \sigma_- \quad \text{and} \quad B_-(t) = \sum_i g_i \hat{b}_i^\dagger e^{i\omega_i t}. \quad (3.64)$$

Explicit calculation of the bath correlation functions is straightforward when the bath is assumed to be in a thermal state at temperature T ,

$$\rho_E = \frac{1}{Z_B} e^{-\beta H_B}, \quad (3.65)$$

where Z_B is the bath's partition sum and $\beta = 1/(k_B T)$. For this particular environ-

mental state, one can derive the relations [76]

$$\langle \hat{b}_i \hat{b}_j \rangle = \langle \hat{b}_i^\dagger \hat{b}_j^\dagger \rangle = 0, \quad (3.66)$$

$$\langle \hat{b}_i \hat{b}_j^\dagger \rangle = \delta_{ij} (N(\omega_i) + 1), \quad (3.67)$$

$$\langle \hat{b}_i^\dagger \hat{b}_j \rangle = \delta_{ij} N(\omega_i). \quad (3.68)$$

Here, $N(\omega)$ denotes the Bose-Einstein occupation number, which describes the average number of photons in a mode with frequency ω ,

$$N(\omega) = \frac{1}{e^{\beta \hbar \omega} - 1}. \quad (3.69)$$

Putting all this into Eq. (3.26), we obtain for the master equation and correlation functions

$$\dot{\rho}_S(t) = -\Gamma_+ (\sigma_+ \rho_S(t) \sigma_- - \sigma_- \sigma_+ \rho_S(t)) - \Gamma_- (\sigma_- \rho_S(t) \sigma_+ - \sigma_+ \sigma_- \rho_S(t)) + \text{H.c.}, \quad (3.70)$$

with

$$\Gamma_+ = \sum_i \int_0^\infty ds e^{i(\omega_i - \omega_0)s} \frac{|g_i|^2}{\hbar^2} N(\omega_i), \quad (3.71)$$

$$\Gamma_- = \sum_i \int_0^\infty ds e^{-i(\omega_i - \omega_0)s} \frac{|g_i|^2}{\hbar^2} (N(\omega_i) + 1). \quad (3.72)$$

We can further simplify the above expressions by using the relation

$$\int_0^\infty ds e^{\pm i \varepsilon s} = \pi \delta(\varepsilon) \pm i \frac{\mathcal{P}}{\varepsilon}, \quad (3.73)$$

where $\mathcal{P}$ denotes Cauchy's principal value, and converting the sum to an integral over the density of states of photon modes $J(\omega)$. With the definition of the following

parameters,

$$\Delta = \mathcal{P} \int d\omega \frac{|g(\omega)|^2}{\hbar^2} J(\omega) \frac{N(\omega) + 1/2}{\omega - \omega_0}, \quad (3.74)$$

$$\Gamma = 2\pi \frac{|g(\omega_0)|^2}{\hbar^2} J(\omega_0), \quad (3.75)$$

and after some algebra, we can write the final ME in the form of Eq. (3.31):

$$\begin{aligned} \dot{\rho}_S = & -\frac{i}{\hbar} [(\epsilon + \hbar\Delta)\sigma_z, \rho_S] + \Gamma N(\omega_0)(2\sigma_+\rho_S\sigma_- - \sigma_-\sigma_+\rho_S - \rho_S\sigma_-\sigma_+) \\ & + \Gamma(N(\omega_0) + 1)(2\sigma_-\rho_S\sigma_+ - \sigma_+\sigma_-\rho_S - \rho_S\sigma_+\sigma_-) \quad . \quad (3.76) \end{aligned}$$

This important final result is called the the *quantum optical master equation*. The Stark and Lamb shift term Δ in Eq. (3.76) is often either neglected or directly absorbed into ϵ , a renormalisation which has no influence on the form of the dissipator of the master equation [77].

Let us briefly discuss the terms of the dissipator of Eq. (3.76). The first group of terms proportional to $\Gamma N(\omega)$ raises the energy of the system by inducing the transition from $|g\rangle$ to $|e\rangle$. The required quantum of energy is taken from the environment through photon absorption, which explains why this process only occurs at finite temperature when the photon modes are thermally occupied. The second group of terms consists of two processes which lower the system's energy by means of photon emission. Of these the part proportional to $\Gamma N(\omega)$ corresponds to stimulated emission and the part proportional to Γ describes spontaneous emission. As expected, the latter is independent of the occupancy of photon modes, taking place independent of temperature.

3.5 Non-Markovian Dynamics

Markovian dissipation processes described by a ME in Lindblad form cause an exponential decay of the coherences in the system's density matrix while the system evolves into a mixed state. Conversely, a non-exponential decay of the coherences is a signature for non-Markovian processes. Indeed, for phonon-induced decoherence in self-assembled quantum dots (a system we shall study later on), non-Markovian features have been observed in measured data of experiments with fast optical excitation on sub-picosecond timescales [82, 83].

A simple 2LS coupled to a bath of bosons (e.g. phonons) is often referred to as the spin-boson model. For this model, exact analytical (non-Markovian) solutions exist [84, 85]. However, quite often we require a more general Hamiltonian with an additional term proportional to σ_x to adequately describe the physics of a 2LS, which leads to a generalised spin-boson Hamiltonian of the form

$$H = \epsilon\sigma_z + \Delta\sigma_x + \sum_{\mathbf{q}} \omega_{\mathbf{q}} \hat{a}_{\mathbf{q}}^\dagger \hat{a}_{\mathbf{q}} + \sigma_z \sum_{\mathbf{q}} g_{\mathbf{q}} (\hat{a}_{\mathbf{q}}^\dagger + \hat{a}_{\mathbf{q}}), \quad (3.77)$$

where ϵ is the energy difference and Δ the coupling between the qubit basis states, $\omega_{\mathbf{q}}$ is the frequency of the oscillation mode $\mathbf{q}$ and $g_{\mathbf{q}}$ describes the corresponding coupling strength of this mode to the qubit. $\hat{a}_{\mathbf{q}}^\dagger$ and $\hat{a}_{\mathbf{q}}$ are the creation and annihilation operators of the oscillation modes and σ_z and σ_x are the Pauli matrices in the basis of the qubit. Although it has proven difficult to find a general solution for this Hamiltonian, there are approaches for dealing with certain parameter regimes. Since we are looking at non-Markovian solutions, dynamics of the system will inevitably be determined by memory kernels, typically a time-dependent bosonic correlation function $C(t - t')$.

In the limit where the coupling to the bosons is expected to be much stronger

than the tunnelling, $g_{\mathbf{q}} \gg \Delta$, the polaron transform is a useful technique [86, 87]. The polaron transform is a unitary transformation into a basis of system states which are ‘dressed’ by the combined effect of all phonon modes. This fully decouples the spin half system from the bath, thus allowing us to treat the exciton-phonon interaction exactly. However, now the tunnelling term of the system Hamiltonian has to be calculated using perturbation theory. For example, this can be achieved by deriving a standard weak coupling Born-Markov ME in the polaron frame, which treats the tunnelling term as a perturbation to second order [88]. Another approach, also based on the polaron transform, is offered by a set of integro-differential equations for the qubit’s density matrix in Ref. [86]. Depending on the structure of $C(t - t')$, a Laplace transform can in some instances help to untangle the equations and lead to analytical solutions [86].

In the opposite limit of $\Delta \gg g_{\mathbf{q}}$, DiVincenzo and Loss have derived equations of motion for the density matrix elements by performing a ‘rigorous Born approximation’ [89]. Again, the equations need to be uncoupled in Laplace space, and the feasibility of this once more depends on $C(t - t')$, though the definition of this bosonic correlation function is now different from the previous one.

Yet another technique based on performing the Born but not the Markov approximation is presented in Refs. [90, 91] for a quantum dot charge state coupled to a bath of phonons. The quantities of interest are expressed as the spectral overlap integral of the bath’s density of states with a spectral representation of the system control parameters. This approach is particularly intuitive and enables an illustrative comparison of the effects of non-Markovian and Markovian theory.

This finishes our short and by no means comprehensive review of non-Markovian dynamics of the generalised spin-boson Hamiltonian, an important area of ongoing research.

Chapter 4

Self-assembled quantum dots

The term quantum dot (QD) is frequently applied to a variety of different physical systems. However, they all share a strong quantum confinement in all three spatial dimensions, and are therefore effectively ‘zero-dimensional’ structures, accounting for the common name. We shall be primarily interested in self-assembled semiconductor heterostructures of a lower band-gap material enclosed by higher band-gap surroundings. As a result, charge carriers in the dot are fully trapped in the confinement potential, leading to a discrete density of states. In typical semiconductors this kind of energy quantisation occurs when the spatial dimensions of the confining potential do not exceed a few tens of nanometres.

Out of the different classes of QDs, lithographically produced dots, either vertically defined [92] or lateral [93], display properties particularly conducive to QIP applications, since the manufacturing techniques enable a great deal of control over the shape, arrangement and size of the confinement potentials. However, these dots are not optically active and require electrical gating. For this reason, we will not consider them further and instead concentrate on the properties of the equally promising self-assembled quantum dots.

The discrete energy-level structure of those QDs is reminiscent of atoms and has

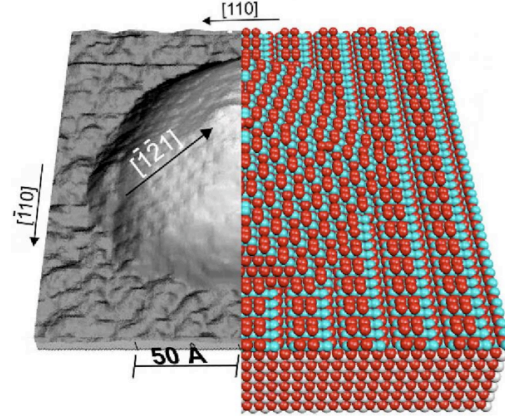


Figure 4.1: A self-assembled InAs quantum dot grown on GaAs substrate: the left half shows a high resolution STM image and the right the prediction of a numerical model based on Density Functional Theory [98]. Picture courtesy of Dr Thomas Hammerschmidt.

earned them the name ‘artificial atoms’. Correspondingly, many of the quantum optics techniques conventionally used with atoms can be applied to QDs. To list but a few experimental milestones, Rabi oscillations [94, 95], photon antibunching [96], and the optical Stark effect [97] have all been observed experimentally. The interest in self-assembled QDs is thus mainly founded on these atom-like properties combined with the advantages of solid state handling and stability, along with the potential for integration into conventional digital circuits.

4.1 Fabrication

Self-assembled QDs are typically grown with the Stranski-Krastanov (SK) technique [99, 100, 101]. A thin layer of semiconductor material is deposited on a planar substrate surface with a smaller lattice constant, typically using Molecular Beam Epitaxy (MBE). The strain due to the lattice mismatch makes it energetically favourable for the dot material to spontaneously nucleate and form little islands on top of a monolayer on the surface (the so-called wetting layer, see Fig. 4.1 for

an illustration). These islands are then overgrown by the substrate and thus form regions that are entirely embedded in the substrate material. The SK approach works particularly well for materials from the III-V groups of the periodic table of elements, for example $\text{In}_x\text{Ga}_{1-x}\text{As}$ on a GaAs substrate.

Since the positions of the nucleation centres are determined by random processes (on a perfectly smooth and regular surface), it can be difficult to grow the dots in ordered arrays. However, if several layers are stacked on top of each other, it has been observed that the dots preferentially line up with one another, as this gives the lattice a way of minimising overall strain [102, 103]. Self-assembled SK dots appear in fairly homogeneous ensembles but are not strictly of identical size. Yet, size fluctuations have a direct effect on the energy levels, so that the dots are non-resonant, allowing individual optical addressability. However, if required it is also possible to tune two dots into resonance with the AC Stark effect [104].

4.2 Electronic properties: single particle states

Obtaining an exact solution for a single particle state of a charge carrier in a QD involves solving the SE for a many-body problem of several thousands of ions and electrons and is therefore an extremely difficult undertaking. Fortunately, there exist some well-established techniques which tremendously simplify the problem, even bringing it into the realm of analytical solvability. The important approximations to make are the effective mass approximation [105, 106] and the envelope function approximation [107]. With the effective mass approximation, one makes the assumption that the crystal structure affects a single electron in such a way that it can be described as a free particle with an altered ‘effective mass’. Sweeping as this assumption may sound, it has proven to be a good approximation widely used across solid state and semiconductor physics [106, 108]. For the envelope wavefunc-

tion approximation, one factors out the fast oscillating part of the wavefunction, i.e. the Bloch function, which arises from the periodicity of the atomic lattice. The underlying physical assumption is that most properties of interest can be derived from the slowly varying wavefunction envelope over the region of the dot [109].

Forgoing the amount of mathematical detail delivered by dedicated textbooks on this subject [108, 109], let us nevertheless formalise these assumptions by starting with the time-independent Schrödinger equation for a Hamiltonian that includes the periodic crystal lattice potential and an additional confinement potential $V(\mathbf{r})$ describing the spatial variation of the band gap,

$$[H_{bulk} + V(\mathbf{r})] \psi(\mathbf{r}) = E\psi(\mathbf{r}). \quad (4.1)$$

The known bulk solutions satisfy $H_{bulk}U_{n\mathbf{k}}(\mathbf{r}) = \varepsilon(\mathbf{k})U_{n\mathbf{k}}(\mathbf{r})$, where the $U_{n\mathbf{k}}(\mathbf{r})$ are expressed in terms of Bloch functions sharing the periodicity of the lattice [109]

$$U_{n\mathbf{k}}(\mathbf{r}) = u_{n\mathbf{k}}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}. \quad (4.2)$$

Here, $\mathbf{k}$ is a wavevector within the first Brillouin zone and n denotes the band index. As the Bloch functions form a complete set, we can expand the general solution of the full Schrödinger equation as an integral over the first Brillouin zone

$$\psi(\mathbf{r}) = \sum_n \int \frac{d\mathbf{k}}{2\pi^3} \tilde{\phi}(\mathbf{k}) U_{n\mathbf{k}}(\mathbf{r}). \quad (4.3)$$

Our interest resides primarily with electron and hole states that are close to the band extrema at $\mathbf{k} = 0$, i.e. the important states for optical transitions in direct semiconductors such as GaAs. Hence, we make two approximations: first, we assume that only a single band contributes to the solution and drop the summation over n .¹

¹This is unproblematic for electron states but may be questionable for holes, which occupy two

Second, we assume that most of the $\mathbf{k}$ -dependence in $U_{n\mathbf{k}}(\mathbf{r})$ stems from the plane wave factor $e^{i\mathbf{k}\cdot\mathbf{r}}$ and we may therefore neglect the $\mathbf{k}$ -dependence of $u_{n\mathbf{k}}$. This allows us to write our general solution as a simple product of the periodic Bloch function u_{n0} and the slowly varying envelope function $\phi(\mathbf{r})$:

$$\psi(\mathbf{r}) = u_{n0}(\mathbf{r})\phi(\mathbf{r}), \quad (4.4)$$

where the envelope function $\phi(\mathbf{r})$ is the Fourier transform of the expansion coefficients $\tilde{\phi}(\mathbf{k})$. We proceed by making the assumption of a parabolic band shape near $\mathbf{k} = 0$,

$$\varepsilon_n(\mathbf{k}) = \varepsilon_n(0) + \frac{\hbar^2 \mathbf{k}^2}{2m^*}, \quad (4.5)$$

where we have introduced the effective mass m^* according to its customary definition as the inverse of the band curvature at $\mathbf{k} = 0$. Substituting Eq. (4.4) back into the original Schrödinger equation, we find that the Bloch part of the wavefunction cancels and the envelope function must satisfy the modified Schrödinger equation

$$\left[-\frac{\hbar^2}{2m^*} \nabla^2 + V(\mathbf{r}) \right] \phi(\mathbf{r}) = (E - \varepsilon_n(0))\phi(\mathbf{r}), \quad (4.6)$$

with energy now measured from the band extremum [110]. Hence, we have reduced the complicated initial problem to finding single particle solutions, where the semiconductor heterostructure only enters statically through the spatial variation of the band gap.

In the most simplistic model, one can treat the carriers as particles in a box with infinite potential walls in all directions. A still simple and widely used approach [111, 112] is that of assuming a separable potential with an infinite square well in the growth direction (where the transition from the dot to the substrate material

degenerate bands at $\mathbf{k} = 0$. However, we shall not consider this complication here, assuming that there is no mixing of the different hole states in the valence band.

tends to be rather abrupt) and an infinite harmonic potential in the lateral direction. While this gives nice analytic solutions, it is clear that the predicted energy spectrum cannot be expected to reflect the dot's energy levels very well. However, refining the model by replacing the infinite potential with a square well and a parabolic potential of finite height V is not as straightforward as it sounds because it leads to regions around the 'corners' in which the overall potential adds up to multiples of V , thus requiring further perturbative corrections.

When those simple models are not sufficient, there exist much more sophisticated approaches for calculating the single particle electron and hole states of the dot, notably pseudopotential calculations [113] and Density Functional Theory (DFT) [114]. These approaches resolve the crystal structure rather than relying on the effective mass approximation and are known to provide a far more accurate density of states. However, there is a huge amount of computational cost involved, limiting the calculations to structures on the order of roughly a thousand atoms. Another drawback of these techniques is that the single particle wavefunctions, typically an expansion in the plane wave basis, do not lend themselves to further analytical exploration.

Let us turn back to a commonly used simple model and calculate the ground state wavefunction of an infinite parabolic potential of strength f ,

$$V(\mathbf{r}) = \frac{1}{2}f\mathbf{r}^2, \quad (4.7)$$

which is easily separable, giving rise to three independent one-dimensional harmonic oscillators. Their solutions are essentially given by Hermite polynomials [75], reducing to a simple Gaussian for the ground state solution. Thus we obtain for the

envelope function (in the ground state)

$$\phi(\mathbf{r}) = \left(\frac{1}{d\sqrt{\pi}} \right)^{\frac{3}{2}} \exp \left(-\frac{\mathbf{r}^2}{2d^2} \right), \quad (4.8)$$

with energy $E_0 = 3/2 \hbar\omega$, where $d = (\hbar/\sqrt{m^*f})^{1/2} = (\hbar/(m^*\omega))^{1/2}$. Obviously, the isotropic potential does not reflect the physical reality of ‘disc-shaped’ self-assembled QDs very well, but the generalisation to an anisotropic confinement potential is straightforward [110].

We conclude this section by considering the case where one electron is promoted from the valence band into the conductance band (for example by optical excitation) leaving a hole behind in the valence band; such an electron-hole pair is called an exciton. Excitons are trapped inside the dot and cannot wander around freely, since the exciton creation energy is typically much smaller than the bulk semiconductor band gap. We shall consider excitons in the strong confinement regime, where we can treat the electron and hole state separately. The total energy of such a ground state exciton E_X is then given by

$$E_X = E_{gap} + E_e + E_h - E_{Coulomb}, \quad (4.9)$$

where E_{gap} denotes the band gap of the dot material (which is smaller than the substrate band gap), and the single particle ground state energies $E_{e,h}$ are obtained by solving the Schrödinger equation for the electron and hole separately. In general, these solutions for the electron and hole will be different, owing to their different effective mass [106]. $E_{Coulomb}$ is the Coulomb attraction term due to the opposite charge of the exciton constituents, an expression for which can be found in Ref. [111].

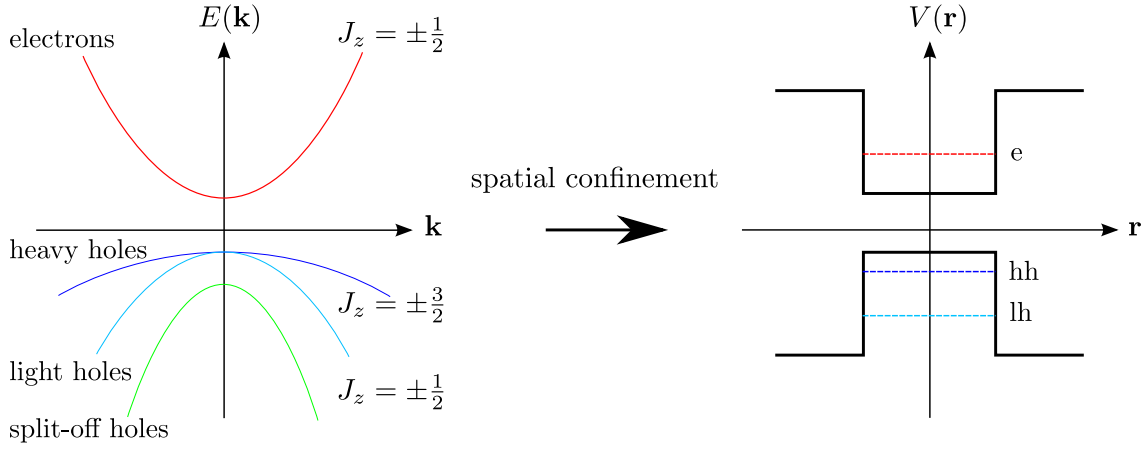


Figure 4.2: Schematic of the dispersion relation $E(\mathbf{k})$ of electrons and holes in the bulk material near the band edge. In a spatially confined geometry, discrete energy levels emerge and light and heavy holes are no longer degenerate because of their different effective mass. Crucially, spin is preserved and thus the confined heavy hole states retain $J_z = \pm 3/2$, while electrons and light holes possess spin projections of $J_z = \pm 1/2$ [115].

4.3 Optical properties and selection rules

Suppose an electron has been lifted from the valence to the conduction band. The overlap of electronic and hole envelope wavefunction then results in a dipole transition matrix element associated with the exciton, so that after some time (the exciton's natural lifetime), electron and hole recombine by emitting a photon. The nature of this process is very similar to the spontaneous photon emission of an excited atom. While a full treatment of the optical properties of QDs is rather involved and beyond the scope of this chapter, we shall introduce the selection rules of the dipole-allowed crystal ground state to single exciton transition.

Recall from Eq. (3.41) that the coupling of light to matter is directly proportional to the dipole operator $\mathbf{d}$ between the ground and excited state of the matter system. For the exciton, this dipole moment is determined by the wavefunction overlap of

the electron and hole as expressed by

$$\mathbf{d} = e \int d^3\mathbf{r} \psi_e^*(\mathbf{r}) \mathbf{r} \psi_h(\mathbf{r}), \quad (4.10)$$

where $\psi_{e,h}(\mathbf{r})$ are the independent electron and hole wavefunctions, respectively. This expression can be split up into the product of two integrals, where the first describes the overlap of the electron and hole envelope functions, and the second integral contains Bloch functions and corresponds to the intrinsic transition dipole moment of the bulk material [115].

In order to proceed, we need to look at the symmetry properties of the wavefunctions, which is contained in the Bloch functions. We have *s*-like orbital wavefunction symmetry in the conduction band [115], leading to electron states with the usual twofold spin degeneracy and angular momentum $J = 1/2$. On the other hand, the valence band possesses *p*-like orbital symmetry [115], so that there is one band with orbital angular momentum $L = 1$ and total angular momentum $J = 3/2$, and a second band with $L = 0$ and $J = 1/2$. This latter band is energetically shifted due to spin-orbit coupling and its states are fittingly known as split-off holes. The $J = 3/2$ band is subdivided into heavy ($J_z = \pm 3/2$) and light hole ($J_z = \pm 1/2$) states, which are degenerate at $\mathbf{k} = 0$ but diverge away from the band edge due to different band curvatures. In turn, this means that heavy and light holes have a different effective mass, so that their single particle states in the QD potential are not degenerate and heavy hole states lie closer to the band edge, as shown by Fig. 4.2.

In the following, we consider a *z*-direction that is parallel to the natural symmetry axis of the dot, i.e. the growth direction. Bearing in mind that photons carry unit angular momentum and that angular momentum must be conserved, the selection rules for dipole allowed transitions are now easy to understand for light propagating

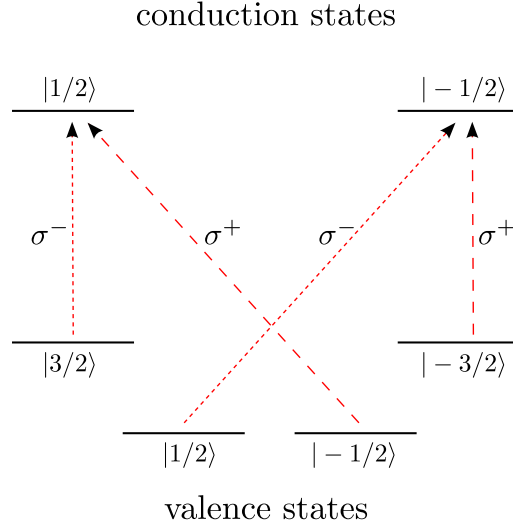


Figure 4.3: Dipole allowed transitions between lowest lying conduction and valence band states. The required photon polarisation to conserve angular momentum is also shown. The states $|i\rangle$ are labelled according to the angular momentum projection in the z -direction: $J_z = i$.

along this direction. For instance, a photon needs to supply $+1$ unit of angular momentum to promote an electron from the $J_z = -3/2$ valence band state to the $J_z = -1/2$ conduction band state and this can only be accomplished by σ^+ polarised light. Similarly, σ^- polarised light with angular momentum -1 can promote a $J_z = 3/2$ valence electron to the $J_z = 1/2$ conduction state. On the other hand, the transition $J_z = 3/2 \rightarrow J_z = -1/2$ is dipole-forbidden, as the required angular momentum cannot be delivered by a single photon. Fig. 4.3 gives an overview of all dipole-allowed transitions and the required laser polarisation to drive them. Finally, we note that an unoccupied $J_z = -3/2$ electron state in the valence band corresponds to a $J_z = 3/2$ hole, so speaking in terms of hole states, σ^+ light optically couples the $J_z = 3/2$ hole to the $J_z = -1/2$ electron and so on.

4.4 Using quantum dots as qubits

At this stage, we know enough about the physical properties of QDs to think about possible applications, for example using the states of the QD as a physical qubit. There are several natural ways of defining a qubit in a QD: spin qubits and excitonic or charge qubits. We shall outline these approaches and review their respective merits and shortcomings in the following.

The spin of an excess electron confined in the conduction band of a doped QD is a natural choice for a qubit, which can be defined as $|1\rangle \equiv |\uparrow\rangle$ and $|0\rangle \equiv |\downarrow\rangle$, where the ‘up’ and ‘down’ arrow denote the S_z spin projection. Because of weak environmental coupling, electron spins possess relatively long coherence times (T_2) up to a few microseconds [93, 116], and electron spin lifetimes (T_1) can even be on the order of many milliseconds [117].

A number of important experimental results have been achieved with spin qubits in lateral quantum dots: Rabi oscillations, a popular experimental indicator of coherent control, have been observed for a single electron spin in a quantum dot [118]. As for measurements, single-shot individual electron spin readout has been demonstrated through spin to charge conversion and subsequent detection with a quantum point contact [119], and by spin-dependent tunnel rates through the dot [120]. However, QD spin manipulation is not only restricted to electrically defined quantum dots. Ultrafast coherent optical control of electron spins (on the picosecond timescale) in self-assembled quantum dots have been reported by Refs. [121, 122], while Atatüre *et al.* have demonstrated a spin preparation fidelity in excess of 99.8% [123], as well as fast reliable measurements of spin states using Faraday rotations [124]. In addition, Greilich *et al.* [125] have performed optical measurements of spin coherence and Xu *et al.* [126] have achieved fast spin cooling using excitons.

An alternative way of encoding a qubit is to define it by the absence, $|0\rangle$, or

presence, $|1\rangle$, of a (ground state) exciton in the QD. In contrast to spin qubits, these excitonic qubits interact rather strongly with their environment and thus suffer from rapid decoherence, with excitonic lifetimes typically being shorter than a nanosecond [82]. The advantage of excitonic qubits is that they can be conveniently manipulated with optical laser pulses on the picosecond timescale [127]. Among the experimental achievements are excitonic Rabi flopping, which has now been observed by a number of groups [94, 95, 128, 129], and an entangling CROT gate between two excitonic qubits within a single QD has been accomplished by Li *et al.* [130]. Most recently, Robledo and co-workers have performed conditional logic with a two quantum dot system [131].

To summarise the previous paragraphs: spin qubits exhibit long coherence times and thus provide a good memory but fast manipulation remains a challenge, although significant experimental progress has recently been made. Excitonic qubits can be conveniently controlled and manipulated but suffer from more rapid decoherence. In this sense, the two encodings have complementary features and it would be desirable to combine the long coherence times of spin qubits with fast excitonic gating. Therefore, hybrid schemes that store the qubit in a localised electron spin for most of the time and only go to a selective excitonic representation while a gate is being performed, have been suggested as a way of marrying the advantages while avoiding the pitfalls of these two representations. A popular scheme for this relies on the spin-selective creation of an exciton, which can be achieved through angular momentum selection rules and Pauli blocking by optical excitation [132], an effect which has also been seen in experiments [133, 134]. This scheme has sparked a range of proposals for implementing single and two-qubit gates [132, 135, 136].

4.5 Excitonic decoherence mechanisms

We conclude this chapter on quantum dots by discussing the dominant decoherence mechanisms that afflict excitonic qubits: radiative decay and phonon-induced decoherence.

Radiative decay occurs when the electron in the conduction band and the hole in the valence band recombine; the rate for this process is the inverse excitonic lifetime. As we have stated before, radiative decay of the exciton is very similar to atomic spontaneous emission and can therefore be described with the same models, most notably various instances of the quantum optical master equation derived in Chapter 3. Having a much longer dephasing time than the prevalent phonon-induced processes, spontaneous emission is often not considered a primary concern for schemes based on coherent exciton control. Nonetheless, when hoping to meet the stringent error thresholds required for fault-tolerant QC, this problem certainly must be addressed [137, 138, 90].

Contrary to an isolated atom, a QD is embedded in the macroscopic solid state matrix of its substrate and the electronic state of the QD therefore couples to lattice vibrations [81]. There are three prevailing decoherence processes resulting from this interaction of the semiconductor QD's charge state with phonons: i) pure dephasing ii) phonon emission iii) phonon absorption. For fast carrier excitation, pure dephasing is the predominant process at low temperatures [139, 23, 140]. An abrupt change in the charge configuration suddenly changes the equilibrium lattice positions of surrounding ions and a packet of phonons is emitted as the lattice relaxation occurs. This corresponds to a transfer of information about the electronic state of the QD into the environment. Therefore, it can be interpreted as a kind of environmental 'which way' measurement of the charge qubit [141], resulting in a loss of coherence and forcing the system into a degraded mixed state. Pure dephasing is energy con-

serving and does not lead to a relaxation in the QD; it can also be interpreted as a series of virtual transitions through intermediate states but finally returning to the initial state [23]. It is an intrinsically non-Markovian process [112] and its influence has been studied extensively for Rabi oscillations [142, 143], absorption line shapes [140, 23] and in the context of the spin-boson model [139].

On the other hand, coherent control operations can take tens of picoseconds. The picosecond phonon memory time is then short compared to the system dynamics and we expect a Markovian approach to be valid, which only encompasses phonon-induced relaxation and cannot give rise to pure dephasing. Several theoretical studies confirm this intuition and show that pure dephasing can indeed be adiabatically eliminated if the coherent exciton creation is slow enough such that all lattice ions adiabatically follow their equilibrium positions [137, 112, 132, 143, 144].

Chapter 5

Optical spin manipulation in quantum dots

5.1 Introduction

In the previous chapter, we have seen that electron spins in quantum dots are promising candidates for quantum computation due to their long intrinsic decoherence times [12]. In combination with optical control of such spins via auxiliary exciton (electron-hole) states, the time required for gating operations could be very short compared to the decoherence time, leading to an extremely high ‘figure of merit’. As we have mentioned, this exciting possibility has been discussed in several proposals recently [135, 132, 145, 138, 112, 146].

However, direct exploitation of the excitonic degree of freedom, which we term *dynamic optical control*, may adversely affect the spin coherence: During the gate operation the quantum information is partially carried by the excitons, which are subject to more aggressive decoherence. It has been suggested that *adiabatic control* could avoid this problem, by ensuring that the qubit remains encoded in low-lying states throughout the process [132, 145]. In this chapter, we present calculations

for the adiabatic and dynamic form of a *single qubit phase gate* under the combined effect of the principal decoherence mechanisms, photon emission from excitonic recombination and acoustic phonon interaction. In contrast to previous work [132, 145], we shall derive a full master equation solution for the gate dynamics that simultaneously incorporates all decoherence channels.

Our principal interest will be in evaluating the performance of the adiabatic approach, and the Markovian master equation technique that we develop is tailored for this purpose. In order to provide a meaningful context for these results, we will also consider the performance of a simple dynamic approach in the same parameter regime, i.e. relatively weak driving. However, since the dynamic gate is not the primary focus of our study, we will neglect more advanced dynamic models and techniques such as pulse shaping [147], and also the effects of pure dephasing that may become significant in the case of ultrafast addressing [137].

5.2 Model

Consider a self-assembled QD that is doped such that one excess electron, the spin of which is the qubit, permanently occupies the lowest energy state of the conduction band. If the dot is irradiated with σ^+ polarised laser light, only one of the electron spin configurations is compatible with the creation of an additional exciton due to the Pauli blocking effect [132, 148, 135]. This effect, which has recently been observed experimentally [133, 134], relies on the Pauli exclusion principle for the lowest energy conduction band electron state (see Fig. 5.1). This state can be occupied by at most two electrons, each of opposite spin orientation, with the electron qubit permanently occupying one of the slots. In the valence band, the heavy and light hole energy levels split due to their differing effective masses. A σ^+ circularly polarised laser incident on the QD carries photons with angular momentum $l = +1$. Thus, if the qubit is in

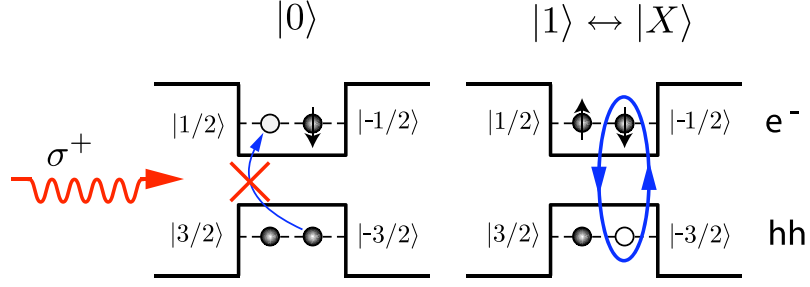


Figure 5.1: Pauli blocking effect: heavy holes occupy the lowest energy valence band states with spin $J_z = \pm 3/2$ whereas light holes have spin $J_z = \pm 1/2$ at a slightly higher energy. Therefore, σ^+ light cannot excite electron hole pairs if the qubit is in the state $|0\rangle$ with $J_z = -1/2$. On the other hand, if the qubit is in the $|1\rangle$ configuration the excitation is possible leading to a three particle trion $|X\rangle$ state.

state $|0\rangle$, with spin $m_z = -1/2$, promoting another electron to the conduction band is incompatible with the conservation of angular momentum and no transition can occur (see Chapter 4). Conversely, for a qubit in state $|1\rangle$ ($m_z = 1/2$) the transition is allowed and leads to the creation of a three-particle trion state $|X\rangle \equiv |\uparrow, \downarrow, \nabla\rangle$, where the arrows symbolise the spin projection of the electron, and the triangle of the valence band state ($|\nabla\rangle \equiv |-3/2\rangle$), respectively. Exploiting the selective coupling of $|1\rangle$ to $|X\rangle$ thus allows for exciton-mediated spin manipulation.

We assume that the qubit states are degenerate on the scale of the exciton creation energy and write the Hamiltonian of a QD and a classical laser field in the basis $\{|0\rangle, |1\rangle, |X\rangle\}$

$$H_S = \omega_0 |X\rangle\langle X| + \Omega \cos \omega_l t (|1\rangle\langle X| + |X\rangle\langle 1|), \quad (5.1)$$

where ω_l is the laser frequency and ω_0 the exciton creation energy. Ω is the laser-QD coupling strength, which is proportional to the dipole moment as well as the intensity of the laser beam. Furthermore, we set $\hbar = 1$ throughout this chapter. For a laser frequency ω_l close to resonance with ω_0 , we define $\Delta \equiv \omega_0 - \omega_l$ as the detuning and move into a frame rotating with the laser. After performing a rotating

wave approximation the Hamiltonian is:

$$H_S = \Delta|X\rangle\langle X| + \frac{\Omega}{2}(|1\rangle\langle X| + |X\rangle\langle 1|). \quad (5.2)$$

Hamiltonian (5.2) provides a valid description of the driven QD, regardless of whether a classical laser field or a quantum mechanical laser mode in a coherent state is assumed [80]. However, the eigenstates of the joint system in a fully quantum mechanical treatment contain the photon number N of the laser mode¹, giving rise to the following representation in the so-called ‘dressed basis’ [80]:

$$|-\rangle_N = \cos\theta|1, N+1\rangle - \sin\theta|X, N\rangle, \quad (5.3)$$

$$|+\rangle_N = \sin\theta|1, N+1\rangle + \cos\theta|X, N\rangle, \quad (5.4)$$

$$\theta = \frac{1}{2} \arctan\left(\frac{\Omega}{\Delta}\right). \quad (5.5)$$

In these quantised expressions, Ω depends on N , as we have seen in Chapter 3. The states $|-\rangle_N$ and $|+\rangle_N$ are conveniently grouped and referred to as a manifold $\mathcal{M}(N)$. In this picture, there is a ladder of manifolds, each separated from its neighbours by the energy of a laser photon ω_l [80]. Henceforth, we shall label the $|-\rangle_N$ state simply as $|-\rangle$ whenever information about the photon number in the laser mode is not needed.

Based on Hamiltonian (5.2), a $R_Z(\phi)$ gate ($|0\rangle \rightarrow |0\rangle$, $|1\rangle \rightarrow e^{i\phi}|1\rangle$) can be performed in two different ways. First, a resonant Rabi cycle allows one to achieve any angle of rotation provided control of the driving laser phase is given [146]. Second, slow switching of an off-resonance laser beam can be used to achieve adiabatic following of instantaneous system eigenstates. As depicted in Fig 5.2, population initially in $|1\rangle$ follows the $|-\rangle$ state as θ goes from zero to θ_{max} and returns back to

¹For clarity, we use the simpler number state rather than a coherent state, which suffices for this purpose [80].

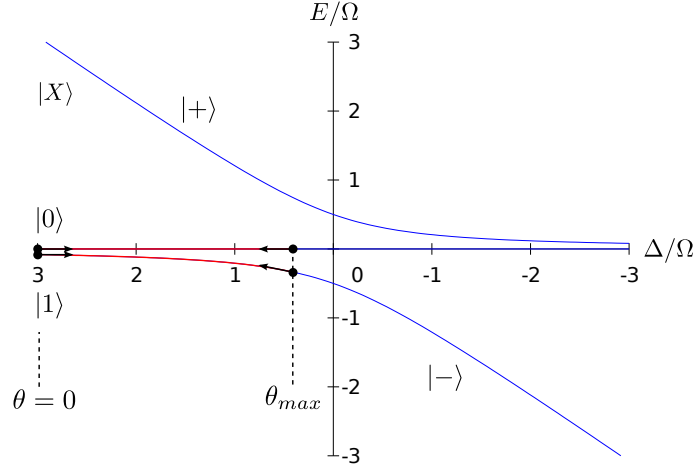


Figure 5.2: **Energy of the dressed states:** shown as a function of the detuning Δ in units of Ω . The $|-\rangle$ state tends to $|1\rangle$ and $|+\rangle$ to $|X\rangle$ for a large positive detuning. At resonance, $\Delta = 0$, the dressed states are an equal superposition of the bare basis states. The decoupled $|0\rangle$ state is also shown, its energy remaining constant. **Adiabatic following:** population in the $|0\rangle$ and $|1\rangle$ states follows the respective instantaneous eigenstates if the mixing angle θ changes sufficiently slowly. $|0\rangle$ is unaffected by the laser pulse but $|1\rangle$ goes into $|-\rangle$ and back. Due to the energy shift of the $|-\rangle$ state, a dynamical phase accumulates relative to $|0\rangle$.

$|1\rangle$ as θ goes back to zero. During this process, the energy difference between $|-\rangle$ and $|0\rangle$,

$$E_m = \frac{1}{2} \left(\Delta - \sqrt{\Delta^2 + \Omega^2} \right), \quad (5.6)$$

causes an accumulation of phase of $|1\rangle$ relative to $|0\rangle$. This kind of adiabatic following can be achieved by using a laser pulse with slowly changing Gaussian field amplitude $\Omega(t) = \Omega_0 \exp[-(t/\tau)^2]$ with a constant detuning Δ .

5.3 Radiative decay

Excitonic lifetimes up to a nanosecond have been reported [82]. Therefore, the problem of spontaneous photon emission has previously often been assumed to be insignificant compared with other decoherence channels [132, 145]. However, to

achieve adiabaticity, our quantum gate must be performed slowly and the finite excitonic lifetime becomes a relevant factor in limiting performance. Spontaneous emission then causes qubit dephasing, although it does not cause qubit relaxation since $|0\rangle$ is always uncoupled to the photon emission process.

Dynamic gates also suffer from such decoherence since any population in the excited state is susceptible to radiative decay at the rate of the inverse natural lifetime Γ_0 . Let Ξ denote the overall ‘number of expected decays’ for a square 2π -pulse that is defined by the integral of the excitonic population over the pulse duration. For undamped, resonant Rabi oscillations, the population of the excited state obeys $\langle |X\rangle\langle X| \rangle = \sin^2(\Omega/2t)$, so that

$$\Xi = \Gamma_0 \int_0^{2\pi/\Omega} \sin^2(\Omega t/2) dt = \frac{\pi}{\Omega} \Gamma_0. \quad (5.7)$$

In principle a stronger driving reduces Ξ . However, increasing the pulse amplitude is technically demanding and can lead to population leakage from the trion subspace. Moreover, pure dephasing due to the coupling to phonons becomes important in the limit of strong driving, prohibiting a high fidelity operation in this regime [137, 140].

In the adiabatic scheme, radiative transition rates between dressed states (Eqs. (5.3) and (5.4)) are required to describe spontaneous photon emission processes. These are obtained analogously to the case of an atom interacting with a laser pulse [80]. As can be seen in Fig 5.3, the total decay rate Γ_m from $|-\rangle$ into the adjacent manifold is given by the sum of two processes $|-\rangle_N \rightarrow |-\rangle_{N-1}$ and $|-\rangle_N \rightarrow |+\rangle_{N-1}$ [80],

$$\Gamma_m = \Gamma_0 \sin^2 \theta \cos^2 \theta + \Gamma_0 \sin^4 \theta = \Gamma_0 \sin^2 \theta. \quad (5.8)$$

Obviously, Γ_m decreases as θ gets smaller. On the other hand, a smaller θ entails a prolonged gating time. By using Eq. (5.5) and performing a series expansion in

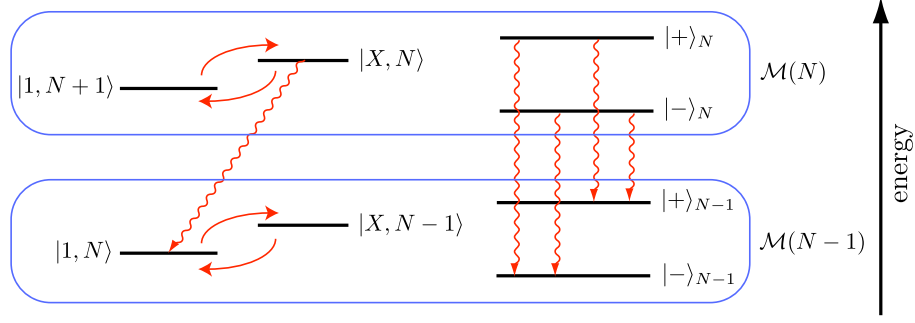


Figure 5.3: Radiative decay from manifold $\mathcal{M}(N)$ to $\mathcal{M}(N-1)$. **Left:** the uncoupled basis. The energy between the two states in each manifold is the detuning Δ and the solid arrows correspond to absorption and stimulated emission processes, whereas the wavy arrows denote spontaneous emission. **Right:** Allowed spontaneous emission transitions between the dressed states. The energetic splitting in each manifold is the effective Rabi frequency $\Omega' = \sqrt{\Delta^2 + \Omega^2}$ and the spacing between adjacent manifolds is the laser frequency ω_l . The left emission process is at frequency $\omega_l + \Omega'$, the two centre-lines emit at the frequency of the laser and the right at $\omega_l - \Omega'$; this is the Mollow triplet well known in quantum optics [149].

Ω/Δ around $\Omega/\Delta = 0$, we obtain

$$\Gamma_m = \frac{\Gamma_0}{4} \left(\frac{\Omega}{\Delta} \right)^2 + \mathcal{O} \left(\frac{\Omega}{\Delta} \right)^4. \quad (5.9)$$

A similar expansion for the energy shift E_m of the $|-\rangle$ state yields

$$E_m = \frac{\Omega}{4} \left(\frac{\Omega}{\Delta} \right) + \mathcal{O} \left(\frac{\Omega}{\Delta} \right)^3. \quad (5.10)$$

The time required for a $R_Z(\pi)$ operation is well approximated by $\tau = \pi/E_m$.² The expected number of decays during the gate is simply the product of time and decay rate, yielding

$$\Xi = \frac{\pi}{\Delta} \Gamma_0. \quad (5.11)$$

Hence Ξ decreases with increasing Δ at the cost a longer gating duration τ .

²Strictly, the pulse duration τ is defined by meeting the condition $\pi = \int_{-\infty}^{\infty} E_m(t) dt$. For a top-hat pulse with E_m this would hence simply give $\tau = \pi/E_m$. This expression also approximates time-dependent excitation well when using the maximal $E_m(t)$.

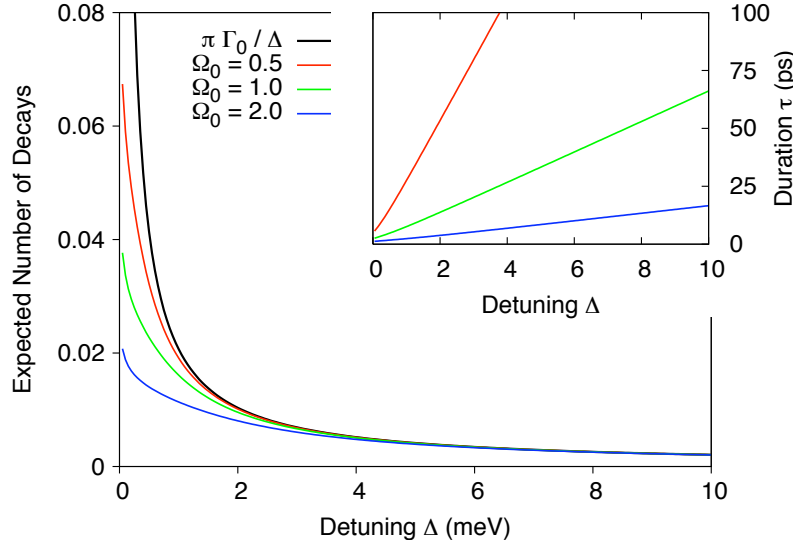


Figure 5.4: Any significant population of the excitonic state makes decay events inevitable. The expected number of decays (for $\Gamma_0 = 0.01 \text{ ps}^{-1}$) during an adiabatic $R_Z(\pi)$ gate is shown as a function of Δ . We depict the analytical prediction (black) along with full numerical solutions, for which Ω_0 is given in meV. The inset shows the corresponding pulse durations τ in ps.

We verify this result in Fig. 5.4, which compares the result Eq. (5.11) with a full numerical simulation, showing an excellent agreement whenever $\Delta \gtrsim 2\Omega$. Moreover, we have resolved the full dynamical evolution of the system's density matrix with the appropriate quantum optical ME (see Chapter 3),

$$\dot{\rho} = -i[H_S, \rho] + \Gamma_0 \left(\sigma_- \rho \sigma_+ - \frac{1}{2}(\sigma_+ \sigma_- \rho + \rho \sigma_+ \sigma_-) \right), \quad (5.12)$$

which reconfirms that further detuning the laser reduces radiative decay in the adiabatic scheme, while it takes a stronger coupling Ω to achieve the same for the dynamic gate. Taking the purity $\text{tr}(\rho^2)$ as a measure for decoherence, we present the results of this calculation in Fig. 5.5, and the inset shows the acquisition of phase in the basis of the qubit. The step-like behaviour of the dynamical gate can be understood as follows: population in $|1\rangle$ does not pick up phase and thus

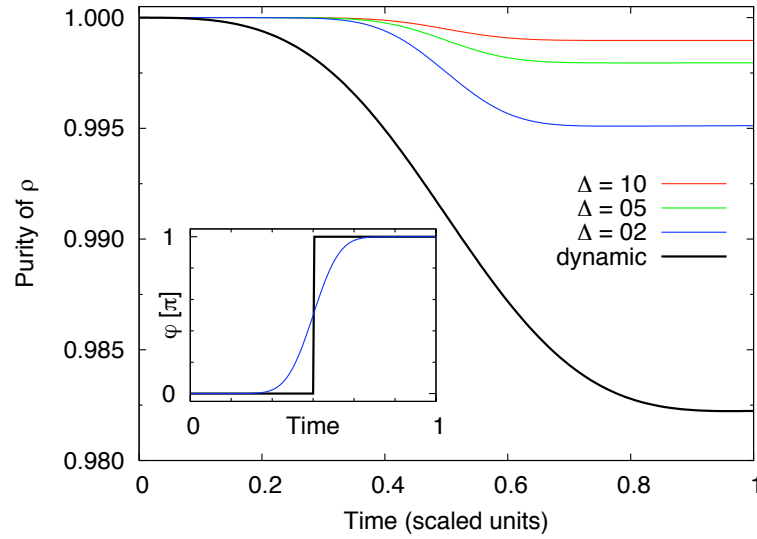


Figure 5.5: Purity of the system's density matrix during a $R_z(\pi)$ operation for an exciton lifetime of $100 \text{ ps} = 1/\Gamma_0$. The dynamic gate operates at $\Omega = 1 \text{ meV}$ and time is shown in units of $2\pi/\Omega$. Adiabatic gates are performed with $\Omega_0 = 1 \text{ meV}$ and for different Δ as shown in the figure. We have scaled the time axis by dividing by 6τ and adding 0.5 (thus mapping the actual integration interval of -3τ to 3τ to the scale of this figure). The inset shows the acquired phase in the basis of the qubit, note that all adiabatic curves coincide.

nothing happens while population is moved from $|1\rangle$ to $|X\rangle$ during the first half of the pulse. It is only the population having undergone a full Rabi cycle that possesses additional phase (the amount can be controlled by the phase of the driving laser [146]). Therefore, the phase between $|0\rangle$ and $|1\rangle$ changes abruptly at the moment when $|1\rangle$ is entirely depopulated and starts being repopulated as population from $|X\rangle$ returns.

5.4 Phonon interactions

Interactions with vibrational modes of the surrounding lattice cause twofold decoherence. First, they dephase the qubit in a similar way to radiative decay and second they can lead to gate failure through state relaxation. We shall see below that it

suffices to focus on deformation potential coupling [81, 140] to acoustic phonons as the main phonon-induced dephasing mechanism for slow excitonic processes, since it dominates over the much weaker piezoelectric coupling.

In addition to the electron spin qubit, the trion state $|X\rangle$ also consists of a strongly bound exciton, which couples to acoustic phonons. The interaction between charge carriers and acoustic phonons is generically given by summing over the displacement operator $(\hat{a}_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^\dagger)$ for each phonon mode $\mathbf{q}$ as follows (see Eq. (3.62) in Chapter 3)

$$H_{ep} = \sum_{\mathbf{q}} M_{\mathbf{q}} \hat{\varrho}(\mathbf{q}) (\hat{a}_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^\dagger). \quad (5.13)$$

Here, $M_{\mathbf{q}}$ is the coupling element,

$$M_{\mathbf{q}} = \sqrt{\frac{\hbar}{2\mu V \omega_{\mathbf{q}}}} C_{\mathbf{q}}, \quad (5.14)$$

where μ is the mass density, V the lattice volume and $\omega_{\mathbf{q}}$ the phonon frequency of mode $\mathbf{q}$. For the deformation potential, the coupling constant is $C_{\mathbf{q}} = D_{e/h} |\mathbf{q}|$ for electrons / holes, and it is $C_{\mathbf{q}} = iP$ for piezoelectric coupling. Meanwhile, $\hat{\varrho}(\mathbf{q})$ is the charge density operator [81]

$$\hat{\varrho}(\mathbf{q}) = \sum_{i,j} \hat{d}_j^\dagger \hat{d}_i \int d^3r e^{-i\mathbf{q}\cdot\mathbf{r}} \psi_j^\dagger(\mathbf{r}) \psi_i(\mathbf{r}), \quad (5.15)$$

with $\hat{d}_i^\dagger, \hat{d}_i$ the creation and annihilation operators of charge carrier i , and $\psi_i(\mathbf{r})$ the corresponding wavefunction. For strongly bound excitons the coupling elements are diagonal and given by the difference between electron and hole coupling elements [140]:

$$M_{\mathbf{q}} \hat{\varrho}(\mathbf{q}) = \sum_i (M_{\mathbf{q}}^e \mathcal{P}[\psi_i^e(\mathbf{r})] - M_{\mathbf{q}}^h \mathcal{P}[\psi_i^h(\mathbf{r})]) \hat{c}_i^\dagger \hat{c}_i, \quad (5.16)$$

where $\hat{c}_i^\dagger$ ($\hat{c}_i$) now denote excitonic creation (annihilation) operators and $\mathcal{P}[\psi^{e/h}]$ are

the form factor of the electron and hole wavefunction, respectively.

Introducing $g_{\mathbf{q}} \equiv (M_{\mathbf{q}}^e \mathcal{P}[\psi^e(\mathbf{r})] - M_{\mathbf{q}}^h \mathcal{P}[\psi^h(\mathbf{r})])$ as the effective excitonic coupling strength, we thus obtain for the ground state exciton on a single QD:

$$H_{ep} = |X\rangle\langle X| \sum_{\mathbf{q}} g_{\mathbf{q}} \left(\hat{a}_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^\dagger \right). \quad (5.17)$$

We proceed by transforming Eq. (5.17) into the diagonal basis of the system Hamiltonian (5.2) and by then writing it in the interaction picture with respect to both H_S and $H_B = \sum_{\mathbf{q}} \omega_{\mathbf{q}} \hat{a}_{\mathbf{q}}^\dagger \hat{a}_{\mathbf{q}}$, yielding,

$$\tilde{H}_I(t) = \sum_{\omega', \mathbf{q}} \left(P_{\omega'} e^{-i\omega' t} + P_{\omega'}^\dagger e^{i\omega' t} \right) g_{\mathbf{q}} \left(\hat{a}_{\mathbf{q}} e^{-i\omega_{\mathbf{q}} t} + \hat{a}_{-\mathbf{q}}^\dagger e^{i\omega_{\mathbf{q}} t} \right), \quad (5.18)$$

where $\omega' \in \{0, \Lambda\}$, $P_0 = \cos^2 \theta |+\rangle\langle +| + \sin^2 \theta |-\rangle\langle -|$ and $P_\Lambda = -\sin \theta \cos \theta |-\rangle\langle +|$. $\Lambda = \sqrt{\Omega^2 + \Delta^2}$ is the energy difference between the dressed states. Here, we have made the assumption that $\Omega(t)$ changes slowly compared with the bath relaxation time. Hence, we can use the instantaneous value of Λ for ω' , which is constant to a good approximation over the relevant integration interval in the derivation of the ME (see also Appendix 5.A at the end of this chapter).

The ME is derived in the usual way (see Chapter 3 or Ref. [76]) by integrating the von Neumann equation for the density matrix ϱ of the joint system and tracing over the phonon modes. This results in an integro-differential equation for the qubit density matrix ρ :

$$\dot{\rho} = - \int_0^t dt' \text{tr}_{ph} [\tilde{H}_I(t), [\tilde{H}_I(t'), \varrho(t')]]. \quad (5.19)$$

The Born-Markov approximation, as described in Chapter 3, is now performed. We reiterate the underlying assumptions: first, there is no back-action from the small system on the much larger bath, meaning the joint density matrix factorises at all

times $\varrho = \rho \otimes \rho_B$. Second, the bath relaxation is assumed to be rapid and so we replace $\rho(t')$ by $\rho(t)$. Further, we assume that system dynamics occurs on a timescale much faster than the decoherence processes, $J(\Lambda) \ll \Lambda$, allowing us to perform a RWA [76, 150], to obtain:

$$\dot{\rho} = J(\Lambda) \left([N(\Lambda) + 1] D[P_\Lambda] \rho + N(\Lambda) D[P_\Lambda^\dagger] \rho \right). \quad (5.20)$$

Here, $D[L]\rho \equiv L\rho L^\dagger - 1/2(L^\dagger L\rho + \rho L^\dagger L)$ is the dissipator of the ME and

$$N(\omega) = \frac{1}{\exp(\omega/k_B T) - 1} \quad (5.21)$$

describes the thermal occupation of the phonon modes. Finally, $J(\omega)$ is the phonon spectral density, a quantity which encapsulates the number of phonon modes available at energy $\hbar\omega$ for a specific coupling mechanism:

$$J(\omega) = 2\pi \sum_{\mathbf{q}} |g_{\mathbf{q}}|^2 \delta(\omega - \omega_{\mathbf{q}}). \quad (5.22)$$

The Lindblad operator P_0 has been dropped because the spectral density vanishes for $\omega = 0$. Consequently, the ME in the Born-Markov approximation provides a suitable description of phonon-assisted transitions but does not describe pure dephasing [140, 139] in the diagonal basis. As we have noted before, pure dephasing is an intrinsically non-Markovian process [112] that can be adiabatically eliminated if the system dynamics proceeds no faster than at a characteristic timescale of the order of roughly a 1 ps [137, 132, 112].

The explicit calculation of the spectral density $J(\omega)$ requires a microscopic model for the carrier wavefunctions. The precise wavefunction shape is not important³ so

³The dynamic gate uses small Λ , where $\mathcal{P}[\psi^{e/h}(\mathbf{r})] \approx 1$ while the adiabatic gate operates best beyond the spectral cut-off of phonon modes.

Phonon coupling material parameters	
Electron deformation potential D_e	14.6 eV
Hole deformation potential D_h	4.8 eV
Piezoelectric coupling constant P	1.45 eV/nm
Effective electron mass m_e	$0.067 m_0$
Effective hole mass m_h	$0.34 m_0$
Mass density μ	5.3 g/cm^3
Velocity of sound c_s	$4.8 \times 10^5 \text{ cm/s}$

Table 5.1: Material parameters for GaAs [140, 139] used in this chapter. m_0 is the free electron mass.

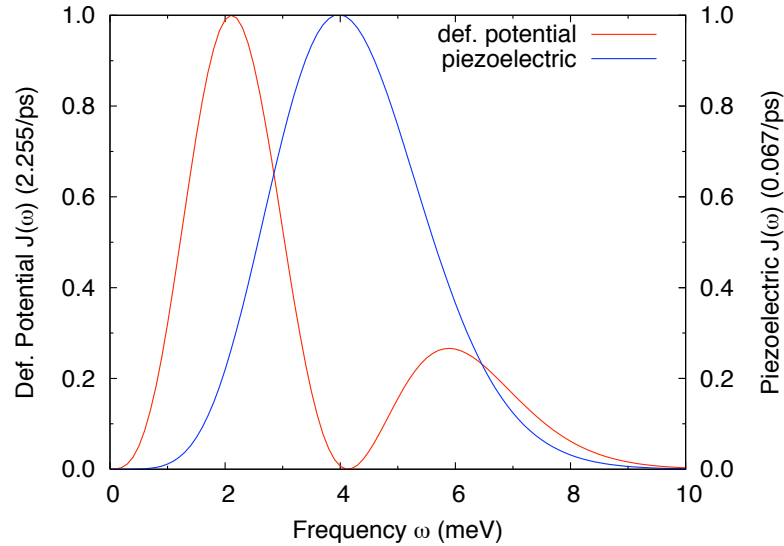


Figure 5.6: The spectral density of phonon modes for deformation potential Eq. (5.24) and piezoelectric coupling Eq. (5.25). Note the two different scales for the y -axes, and that the x -axis has been scaled to meV rather than inverse ps. The dip in the deformation potential curve arises from the assumption of a different effective mass for electron and hole on top of their different deformation potential coupling strengths.

we proceed by using ground state solutions to a harmonic confinement potential of strength 162 meV at 2.5 nm from the centre of the dot, which would be realistic for

an InGaAs dot embedded in a GaAs substrate [110]:

$$\psi_{e/h}(\mathbf{r}) = \left(\frac{1}{d_{e/h}\sqrt{\pi}} \right)^{\frac{3}{2}} e^{-\frac{r^2}{2d_{e/h}^2}}, \quad (5.23)$$

for which $d_{e/h} = (\hbar/\sqrt{m_{e/h}f})^{1/2}$ and $f = 8.3 \times 10^{-3} \text{ J/m}^2$ (see Chapter 4). Furthermore, we use the material parameters given in Table 5.1. Assuming a linear phonon dispersion $\omega_{\mathbf{q}} = c_s |\mathbf{q}|$, the spectral density then takes a superohmic form for the deformation potential,

$$J(\omega) = \frac{\omega^3}{2\pi\mu c_s^5} \left(D_e e^{-\omega^2/\omega_e^2} - D_h e^{-\omega^2/\omega_h^2} \right)^2, \quad (5.24)$$

and for piezoelectric coupling, we obtain an expression with the usual Ohmic prefactor

$$J(\omega) = \frac{P^2\omega}{2\pi\mu c_s^3} \left(e^{-\omega^2/\omega_e^2} - e^{-\omega^2/\omega_h^2} \right)^2. \quad (5.25)$$

The exponential cut-off terms on the right-hand side of Eqs. (5.24, 5.25) are related to the finite size of the QD and filter out high frequency phonons with wavelengths too short to interact with the dot; the cut-off frequencies are: $\omega_{e/h} = 2c_s/d_{e/h}$. Fig. 5.6 reveals that the deformation potential is indeed dominant, as expected from literature [139, 140]. This is primarily due to the fact that electrons and holes couple individually and with a different strength to phonons through the deformation potential, whereas only the difference in their wavefunctions contributes to the piezoelectric coupling. Therefore, we shall neglect piezoelectric coupling from now on, but we note that it is subject to the same cut-off frequencies as the deformation potential; whatever small effect there is due to it will consequently be suppressed in the same limit.

The difference between the deformation coupling constants D_e and D_h combined with different electron and hole cut-off frequencies gives rise to the dip in the de-

formation potential curve in Fig. 5.6. At about 4 meV it drops all the way to zero corresponding to a ‘sweet spot’ at which point the phonon couplings completely cancel out. Typically, self-assembled QDs are disc-shaped with a lateral width of 10–20 nm and a smaller height of around 5 nm in the growth direction. Accounting explicitly for this would introduce an angular dependency into the spectral density with the largest value of the cut-off frequency being in the growth direction, and having a value similar to that obtained using the isotropic potential model.⁴ Hence, the error introduced by the simple model above is likely to be an overestimation of the magnitude of the spectral density towards large frequencies, with no considerable change in the position of the cutoff. Furthermore, in the limit of very small frequency, the spectral density does not depend on the form of the wavefunctions for electron and hole at all because the Fourier transforms then give $\mathcal{P}[\psi^{e/h}(\mathbf{r})] \approx 1$.

By examining the structure of Eqs. (5.20) and (5.24) we conclude that the influence of phonon-induced decoherence becomes small as Λ approaches zero and can be exponentially suppressed for values of Λ beyond the spectral cut-off $\omega_{e/h}$. Outside the ultrafast regime only the first route of decreasing Λ via Ω lends itself to the dynamic scheme, but this is contrary to the requirement of avoiding radiative decay. On the other hand, operation beyond the cut-off is realisable for an adiabatic gate with a large enough detuning. Once more using the purity of the system’s density matrix, $\text{tr}(\rho^2)$, to characterise system decoherence, Fig. 5.7 shows that phonon decoherence can indeed be overcome with the adiabatic approach and that in general performance improves as the detuning increases.

⁴Accounting for the anisotropy would also slightly affect the position of the minima of the deformation potential spectral density in Fig. 5.6, which would cease to be identically zero after averaging over the angular dependence.

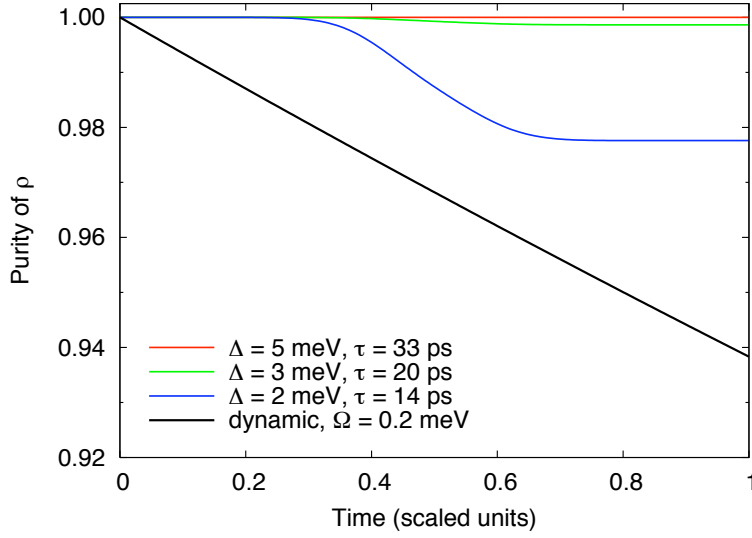


Figure 5.7: Phonons affect dynamic and adiabatic quantum operations differently. To characterise decoherence, we present the purity of the system's density matrix during an $R_z(\pi)$ operation for deformation potential coupling at $T = 5$ K. The dynamic gate uses $\Omega = 0.2$ meV and time is shown in units of $2\pi/\Omega$. Adiabatic gates are performed with $\Omega_0 = 1$ meV and for different Δ as shown in the figure. As in Fig. 5.5, the time axis is scaled to map the actual integration interval of -3τ to 3τ to the scale of this figure.

5.5 Landau-Zener transitions

Landau-Zener (LZ) transitions are non-adiabatic transitions between eigenstates approaching an anticrossing [151]; they are a source of error in the adiabatic scheme since they cause population to transfer to excited states, subjecting it to fast decoherence during and after the gate. LZ transitions can be suppressed by using longer gating times. Contrary to the adiabaticity conditions derived in Refs. [132] and [145], which are only valid for a linear sweep through resonance, we take a more general approach.

To derive an adiabaticity condition consider the effective 2LS Hamiltonian (5.2).

The transformation to the basis of instantaneous eigenstates,

$$U(\theta) = \begin{pmatrix} \cos \theta & -\sin \theta \\ \sin \theta & \cos \theta \end{pmatrix}, \quad (5.26)$$

is time-dependent. The Hamiltonian transforms as $\tilde{H} = U^\dagger H U + i (dU^\dagger/dt) U$, yielding

$$\tilde{H} = \lambda^- |-\rangle\langle -| + \lambda^+ |+\rangle\langle +| + \dot{\theta} (i|-\rangle\langle +| - i|+\rangle\langle -|), \quad (5.27)$$

where $\lambda^\pm = 1/2(\Delta \pm \sqrt{\Delta^2 + \Omega^2})$ denote the instantaneous eigenenergies of the states $|-\rangle$ and $|+\rangle$. The off-diagonal terms give rise to LZ transitions, which are therefore suppressed if

$$|\dot{\theta}| \ll |\lambda^+ - \lambda^-|. \quad (5.28)$$

Using Eq. (5.5) for θ , we obtain a general condition for adiabaticity:

$$\left| \frac{\dot{\Omega}\Delta - \Omega\dot{\Delta}}{2(\Delta^2 + \Omega^2)^{3/2}} \right| \ll 1. \quad (5.29)$$

For $\Omega = \Omega_0 \exp[-(t/\tau)^2]$ and constant Δ , we deduce that inequality (5.29) can be satisfied by demanding

$$\Omega_0/\Delta^2 \ll \tau. \quad (5.30)$$

Adiabatic following is therefore always achieved in the limit $\Omega \ll \Delta$, where $\tau \propto \Delta$ over a broad range of parameter space.

5.6 Overall gate fidelity

In order to bring together the results so far, we now calculate the fidelity of the $R_Z(\pi)$ gate with a combined ME describing both spontaneous photon emission and phonon-

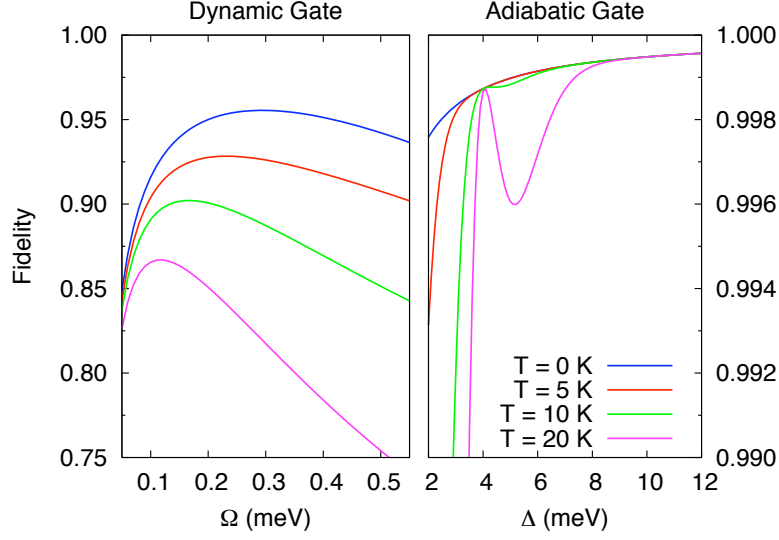


Figure 5.8: Comparison of the dynamic versus the adiabatic gate for a $R_Z(\pi)$ operation by showing the overall gate fidelity. **Left:** fidelity of the dynamic operation as a function of the coupling strength Ω . **Right:** fidelity of the adiabatic operation for a fixed $\Omega_0 = 1$ meV as a function of the detuning.

induced processes. The gate is performed on the input state $|\psi_+\rangle = (|0\rangle + |1\rangle)/\sqrt{2}$ and ideally produces $|\psi_-\rangle = (|0\rangle - |1\rangle)/\sqrt{2}$ as its output state. The fidelity is defined as $\mathcal{F} = \langle \psi_- | \rho | \psi_- \rangle$, where ρ is the density matrix of the system after the gate has finished. Again, we use the material parameters of GaAs and assume a radiative decay rate of $\Gamma_0 = 0.01$ ps $^{-1}$.

The left panel of Fig. 5.8 shows the fidelity of the dynamic gate as a function of Ω for different temperatures. Towards small values of Ω , the fidelity is limited by the finite excitonic lifetime, whereas phonon-induced processes become important for larger values of Ω . The highest fidelity here is approximately 0.95 at absolute zero and decreases rapidly at finite temperatures.

For much higher values of Ω beyond the phonon spectral cutoff (~ 10 meV), we would expect the fidelity to improve rapidly, neglecting the effects of pure dephasing. However, as outlined above, our approach is not suited to studying the regime of ultrafast driving, and we refrain from showing actual values at large Ω .

The fidelity of the adiabatic gate as a function of Δ for a fixed value of $\Omega_0 = 1$ meV is shown in the right panel of Fig. 5.8. Towards larger values of Δ , $\mathcal{F}$ increases and asymptotically approaches unity as radiative decay is more effectively suppressed. Phonon-induced processes are temperature dependent and predominantly occur for small values of Δ . The intermediate peak visible at higher temperatures is related to the dip in the spectral density of the deformation potential, as shown in Fig. 5.6.

5.7 Conclusion

In this chapter, we have performed a comprehensive Markovian decoherence study of an exciton-mediated spin phase gate in quantum dots. Within an adiabatic scheme all principal sources of decoherence can be effectively suppressed in the same limit of weak off-resonant laser driving, and a phase gate fidelity of 0.99 or better may be possible even at finite temperature. In contrast, for similar driving amplitudes, a dynamic gate with typical operating parameters suffers strong decoherence that leads to a significantly lower gate fidelity.

Importantly, these predictions for both the dynamic and the adiabatic gates could be tested with a current experimental setup, as experiments would only involve a single QD and one laser, which is well within the experimental state-of-the-art [129]. Such a demonstration would provide an important step towards implementing more general gates based on adiabatic approaches [138, 152, 90].

Additional to our main result of predicting gate fidelities, we also make another interesting observation. We have seen that the structure of the phonon spectral density has the potential to be rather rich. The adiabatic phase gate, if it were performed in an experiment, would allow a direct mapping of phonon spectral density through the inverse shape of the gate fidelity. Although similar in spirit, this

would go beyond the predicted revival of Rabi oscillations once the effective Rabi frequency is larger than the cut-off [153, 154].

5.A Appendix: interaction picture transformation

The system interaction picture operators of Eq. (5.18) are strictly valid when the system's eigenfrequencies are constant in time, as is the case for the dynamic control scheme. In the following paragraph, we give a qualitative argument for using the same interaction picture operators within the adiabatic approach, which has a time-varying system Hamiltonian due to $\Omega(t)$. In Appendix 7.A of this thesis, we shall revisit the same approximation with a more formal justification.

The transformation of Eq. (5.17) to the interaction picture as in Eq. (5.18) would be strictly correct if the system frequency Λ were constant. However, we require Λ to be time-dependent, albeit on a slow timescale that still achieves adiabatic following. Nonetheless, as long as $\dot{\Lambda}t \ll \Lambda$, we can assume that Λ is essentially constant at any given moment of time, such that a system operator $P_{\omega'}$ is well described by $P_{\omega'}e^{i\omega't}$ as its interaction picture representation (note that the relevant duration t in this comparison is that of the memory time of the bath and not the duration of the gate). Since a Markovian ME only depends on the current time, it is thus reasonable and consistent to use this instantaneous form of the interaction picture operator.

Chapter 6

Robust entangling operation in coupled quantum dots

6.1 Introduction

In this chapter, we perform a detailed study of the principal decoherence channels of an exciton-mediated CPHASE gate acting on two electron spin qubits in coupled quantum dots: radiative decay of the excitonic state, exciton-phonon interactions, and Landau-Zener transitions between laser-dressed states.

Universal QC requires arbitrary single qubit operations and at least one entangling two-qubit gate [3]. We have discussed a z -rotation of a QD spin in the previous chapter. Several other schemes for spin rotations in QDs have been proposed based on stimulated Raman adiabatic passage [155, 112, 138]. Decoherence processes have been thoroughly studied in these cases [112, 90, 152], leading to the prediction that high fidelity single qubit operations should be feasible.

Excitonic transitions may also enable ultrafast optical spin manipulation in coupled nanostructures. Promisingly, excitonic interactions between different QDs have been observed experimentally [156, 157, 158], providing the essential prerequisite for

an entangling *two-qubit* gate. Like in Chapter 5, we here consider a hybrid scheme that proposes to store the qubit in a long-lived localised electronic spin state [53] (with a T_2 time of up to three microseconds [116]), moving selectively to an excited state spin-exciton (trion) representation (which typically relaxes within a nanosecond [82]) only while a gate is actually being performed [132].

In many physical systems, the easiest entangling operation to implement is a controlled-phase (CPHASE) gate that leaves three of the four possible two-qubit computational basis states unaltered, while generating a phase of π on the fourth (i.e. $|00\rangle \rightarrow |00\rangle$, $|01\rangle \rightarrow |01\rangle$, $|10\rangle \rightarrow |10\rangle$, $|11\rangle \rightarrow -|11\rangle$) [3] (see also Chapter 2). Various schemes to realise such a gate exist for spins in coupled QDs [132, 135, 159], of which adiabatic optical control is a particularly promising example as it naturally suppresses excited state population throughout the operation [132, 145]. The technique relies on varying the intensity and frequency of a laser (i.e. the system's external control parameters) sufficiently slowly to achieve adiabatic following of the instantaneous system eigenstates throughout the gating operation. This approach overcomes light-heavy hole mixing in the QD valence band [132, 145] and is also thought to be robust against phonon-induced pure dephasing [132].

In this chapter, we shall simultaneously account for the *principal* decoherence channels of radiative recombination and carrier-phonon interactions by deriving a Markovian master equation (ME) for the laser-driven two-dot system. We will use the ME to assess the performance of the adiabatic approach within experimentally accessible parameter regimes. We also derive conditions to avoid non-adiabatic Landau-Zener (LZ) transitions between eigenstates [160, 161, 162, 151], an effect that would otherwise pose an additional error source. The results will be put in context by comparison with a simple dynamic approach based on resonant Rabi flopping.

6.2 Model

We consider a realistic state-of-the-art system: Two adjacent self-assembled QDs (I and II respectively) with distinct heavy and light hole levels, such as those grown on a GaAs substrate [163], which are usually arranged in stacks [102]. We consider small QDs with a strong confinement potential that dominates over any intra-dot Coulomb interactions. They are each doped such that a single excess electron, the spin of which embodies the qubit, permanently occupies the lowest energy state of the conduction band. The qubit basis is defined as $|0\rangle \equiv |\downarrow\rangle \equiv |-1/2\rangle$ and $|1\rangle \equiv |\uparrow\rangle \equiv |1/2\rangle$, where $|m_z\rangle$ is the spin projection, with $m_z = \pm 1/2$. The z direction also defines the quantisation axis for the hole spins.

In order to couple the two spin qubits we exploit the fact that trion-trion inter-dot Coulomb interactions are expected to be relatively strong [148, 135], such that we can address the system optically through spin-selective exciton creation [132, 148, 135]. Since we have already encountered this Pauli blocking effect in Chapter 5, we refer back to that earlier description and the illustration given in Fig. 5.1. Once more, we here only consider heavy hole valence band states (see Ref. [145] for detailed calculations of the effects of light-heavy hole mixing). In this case, if a dot is irradiated with σ^+ polarised laser light only one of the electron spin configurations is compatible with the creation of an additional exciton, and this resulting three particle state is called a trion and denoted $|X\rangle$. Hence, the logical $|0\rangle$ state of each dot is completely unaffected by the laser pulse, while the logical $|1\rangle$ state optically couples to $|X\rangle$.

Coulomb interactions between trions on the two adjacent dots mediate a coupling of the two spin qubits, as can be seen by considering the full system Hamiltonian. We assume that the qubit states are degenerate and write the Hamiltonian of the

two QDs and a single classical laser field in the basis $\{|0\rangle, |1\rangle, |X\rangle\}$ ($\hbar = 1$) as

$$\begin{aligned}
H(t) &= \omega_0(|X\rangle\langle X| \otimes \mathbf{1} + \mathbf{1} \otimes |X\rangle\langle X|) \\
&+ \Omega \cos \omega_l t (|1\rangle\langle X| \otimes \mathbf{1} + \mathbf{1} \otimes |1\rangle\langle X| + \text{H.c.}) \\
&+ V_{XX}|XX\rangle\langle XX| + V_F(|1X\rangle\langle X1| + \text{H.c.}).
\end{aligned} \tag{6.1}$$

where H.c. denotes the Hermitian conjugate, $\mathbf{1}$ is the identity operator, and ω_0 is the exciton creation energy (assumed the same for both dots). Here, Ω represents the coupling between the QD transition and the laser mode and ω_l is the laser frequency; both of these may be time-dependent quantities. The dots are Coulomb-coupled by virtual photon exchange (Förster interaction) of strength V_F [164], and by a biexcitonic dipolar coupling V_{XX} [148]. We consider typical inter-dot separations of 5 – 10 nm, close enough for both V_F and V_{XX} to be on the meV scale. We also assume that we are in a regime where tunnelling processes are suppressed [111].

To lowest nonzero order, the Förster coupling element is equivalent to the interaction of two point dipoles situated on dots I and II [165],

$$V_F = \frac{C}{\epsilon_r R^3} \left(\langle \mathbf{r}_I \rangle \cdot \langle \mathbf{r}_{II} \rangle - \frac{3}{R^2} (\langle \mathbf{r}_I \rangle \cdot \mathbf{R}) (\langle \mathbf{r}_{II} \rangle \cdot \mathbf{R}) \right), \tag{6.2}$$

where $R = |\mathbf{R}|$ with $\mathbf{R}$ being the vector connecting the centre of the two dots, $\langle \mathbf{r}_{I/II} \rangle$ is the position operator between the electron and hole on either dot I/II respectively, ϵ_r is the dielectric constant, and C the usual Coulomb term $C = e^2/4\pi\epsilon_0$. The sign of V_F is determined by the relative orientation of $\langle \mathbf{r}_I \rangle$ and $\langle \mathbf{r}_{II} \rangle$ with respect to $\mathbf{R}$. Ultimately, the orientation of $\langle \mathbf{r}_{I/II} \rangle$ depends on the crystal structure [165] and is hard to predict. However, for two identical dots in the same crystal structure, $\langle \mathbf{r}_I \rangle$ and $\langle \mathbf{r}_{II} \rangle$ should be parallel and have the same magnitude, such that their joint orientation to $\mathbf{R}$ is the sole factor determining whether V_F is positive or negative.

While the sign of V_F is an immutable property of any given two-dot system, both positive and negative values of V_F should be possible in principle by a suitable geometrical arrangement.

It can be seen from Eq. (6.1) that the Hamiltonian decouples into four non-interacting subspaces [135]: $\mathcal{H}_0 = \{|00\rangle\}$, $\mathcal{H}_1 = \{|01\rangle, |0X\rangle\}$, $\mathcal{H}_{1'} = \{|10\rangle, |X0\rangle\}$ and $\mathcal{H}_2 = \{|11\rangle, |1X\rangle, |X1\rangle, |XX\rangle\}$; this is a direct result of the Pauli-blocking effect. To implement the CPHASE operation, we need to achieve a net phase shift of π on the input state $|11\rangle$, and are therefore primarily interested in the dynamics of $\mathcal{H}_2$. When we need to consider dynamics in the two level subspaces $\mathcal{H}_1$ and $\mathcal{H}_{1'}$ we can appeal to the results of Chapter 5, which analyses an isomorphic Hamiltonian structure.

Proceeding as in [135, 68], we first transform $\mathcal{H}_2$ into the basis of its eigenstates in the absence of driving ($\Omega = 0$): $|11\rangle$, $|\psi_+\rangle = (|1X\rangle + |X1\rangle)/\sqrt{2}$, $|\psi_-\rangle = (|1X\rangle - |X1\rangle)/\sqrt{2}$ and $|XX\rangle$, giving

$$\begin{aligned} H_2(t) &= (\omega_0 - V_F)|\psi_-\rangle\langle\psi_-| + (\omega_0 + V_F)|\psi_+\rangle\langle\psi_+| \\ &+ \sqrt{2}\Omega \cos \omega_l t (|11\rangle\langle\psi_+| + |\psi_+\rangle\langle XX| + \text{H.c.}) \\ &+ (2\omega_0 + V_{XX})|XX\rangle\langle XX|. \end{aligned} \quad (6.3)$$

We now move to a frame rotating with the laser frequency ω_l , detuned from the $|11\rangle \leftrightarrow |\psi_+\rangle$ transition by an amount $\Delta = \omega_0 + V_F - \omega_l$. Within the RWA, Eq. (6.3) becomes

$$\begin{aligned} H'_2 &= (\Delta - 2V_F)|\psi_-\rangle\langle\psi_-| + \Delta|\psi_+\rangle\langle\psi_+| + (2\Delta - 2V_F + V_{XX})|XX\rangle\langle XX| \\ &+ \frac{\Omega}{\sqrt{2}}(|11\rangle\langle\psi_+| + |\psi_+\rangle\langle XX| + \text{H.c.}). \end{aligned} \quad (6.4)$$

Ref. [135] describes how to perform a dynamic CPHASE operation by applying a

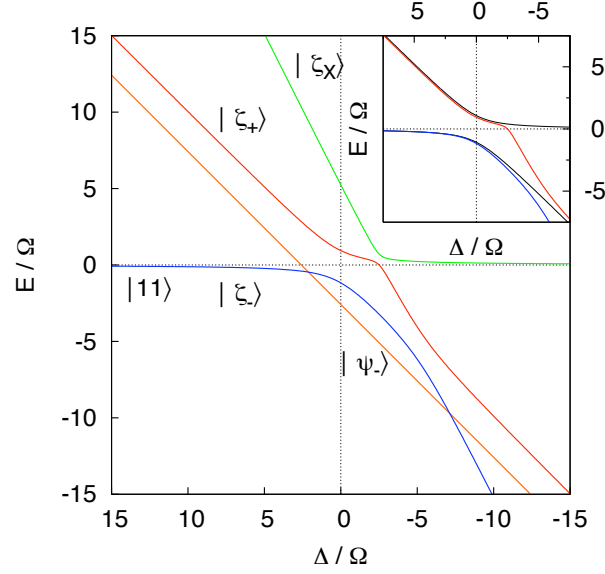


Figure 6.1: Dressed states of subsystem $\mathcal{H}_2$. Parameters are: $V_{XX} = 5$ meV, $V_F = 0.85$ meV, and $\Omega = 1$ meV. The inset shows the states $|\zeta_-\rangle$ and $|\zeta_+\rangle$ around the origin. Also shown are the dressed states obtained if $|XX\rangle$ is neglected. For positive Δ , these approximate $|\zeta_-\rangle$ and $|\zeta_+\rangle$ well.

resonant 2π laser pulse to the $|11\rangle \leftrightarrow |\psi_+\rangle$ transition (i.e. with $\Delta = 0$), generating the required π phase shift on state $|11\rangle$. In this case, certain conditions on the driving strength Ω must be satisfied in order to suppress unwanted transitions to $|XX\rangle$ and transitions within $\mathcal{H}_1$ and $\mathcal{H}_1'$ [135]. However, the gate may also be operated adiabatically without those same constraints by slowly switching an off-resonant laser beam, forcing the system to follow its instantaneous eigenstates [145]. The energy difference between states $|11\rangle$ and $|\psi_+\rangle$ then allows for a phase accumulation on the $|11\rangle$ state, as can be seen by analysing the eigenstates in more detail.

The three levels $|11\rangle$, $|\psi_+\rangle$ and $|XX\rangle$ are coupled by the laser in Eq. (6.4). Moving to the diagonal basis results in three dressed states, which we shall label $\{|\zeta_-\rangle, |\zeta_+\rangle, |\zeta_X\rangle\}$, each of which are superpositions of the three bare basis states weighted according to certain mixing angles that are complicated functions of the system parameters. Nevertheless, we can greatly simplify matters by working with

an approximate 2LS that describes the dynamics to a high degree of accuracy in the interesting parameter regime, where $|\zeta_X\rangle$ is in fact irrelevant. As shown in the inset of Fig. 6.1, for positive $V_F = 0.85$ meV, $V_{XX} = 5$ meV, and *positive* Δ , the energies of states $|\zeta_-\rangle$ and $|\zeta_+\rangle$ are very well approximated by taking

$$\begin{aligned} |\zeta_-\rangle &\approx \cos \Theta |11\rangle - \sin \Theta |\psi_+\rangle, \\ |\zeta_+\rangle &\approx \sin \Theta |11\rangle + \cos \Theta |\psi_+\rangle, \end{aligned} \quad (6.5)$$

where $\Theta = (1/2) \arctan(\sqrt{2}\Omega/\Delta)$. The agreement improves further for larger Δ or smaller driving Ω_0 , which we shall demonstrate is desirable for the adiabatic scheme. Choosing a negative Förster coupling $V_F < 0$ pushes the avoided crossing between $|\zeta_+\rangle$ and $|\zeta_X\rangle$ even further to the right (at the same time shifting the dark state $|\psi_-\rangle$ in Fig. 6.1 upwards). Therefore, we can safely assume that the perturbation caused by $|XX\rangle$ is negligible over a wide parameter range.

Consider shining a laser on the QD with a temporal evolution of $\Delta(t)$ and $\Omega(t)$ that slowly and continuously changes Θ from 0 to some value Θ_{max} , and back to 0 again. Since $|11\rangle$ and $|\zeta_-\rangle$ coincide for $\Theta = 0$, any population in state $|11\rangle$ will adiabatically follow the instantaneous eigenstate of $|\zeta_-\rangle$ and return to $|11\rangle$ at the end of the pulse. Hence, at the end of the pulse, $|11\rangle$ has picked up a phase $\exp(-i\phi_{11})$ relative to $|00\rangle$, where

$$\phi_{11} = \int_0^T dt E_{\zeta_-}(t) = \frac{1}{2} \int_0^T dt \left(\Delta(t) - \sqrt{\Delta^2(t) + 2\Omega^2(t)} \right), \quad (6.6)$$

to a good approximation. Here, T is the pulse duration and $E_{\zeta_-}(t)$ the energy of $|\zeta_-\rangle$ relative to $|00\rangle$. Of course, the states $|01\rangle$ and $|10\rangle$ also accumulate phase relative

to $|00\rangle$ according to

$$\phi_{01,10} = \frac{1}{2} \int_0^T dt \left(\Delta'(t) - \sqrt{\Delta'^2(t) + \Omega^2(t)} \right), \quad (6.7)$$

where $\Delta'(t) = \Delta(t) - V_F$ is the detuning in the $\mathcal{H}_1$ subspace. To achieve a gate locally equivalent to the CPHASE, we demand that

$$\phi_{00} - \phi_{01} - \phi_{10} + \phi_{11} = \pi \quad (6.8)$$

following the operation. This constrains the control parameters $\Delta(t)$, $\Omega(t)$, and T . It is possible, although by no means necessary, to achieve adiabatic following with a chirped laser pulse, although the pulse must always start and finish off resonance [132, 145]. However, for simplicity and practicality, we restrict the following discussion to a pulse with constant detuning and a Gaussian pulse envelope:

$$\Delta(t) \equiv \Delta, \quad (6.9)$$

$$\Omega(t) = \Omega_0 e^{-(t/\tau)^2}. \quad (6.10)$$

Having described, in some detail, the internal dynamics of the driven system we now turn to the impact of the external environment on the CPHASE operation.

6.3 Spontaneous photon emission

The lifetime of excitons in a QD can be as long as a nanosecond [82], while coherent control should be possible on the picosecond timescale [166]. However, in order to avoid phonon-induced pure dephasing [137] and to maintain adiabaticity it can be advantageous to perform operations much more slowly, such that exciton lifetime

becomes an important source of decoherence.

Any population in an excited state of the system is susceptible to radiative decay at the rate of the inverse natural lifetime Γ_0 , due to coupling with the vacuum radiation field. As discussed above, we need only consider the behaviour of an effective 2LS to capture such effects in $\mathcal{H}_2$. We write the Hamiltonian of this 2LS as $H_{\text{2eff}} = \Delta|\psi_+\rangle\langle\psi_+|$ (in the basis $\{|11\rangle, |\psi_+\rangle\}$). In the absence of the dot-laser coupling term, the energies of the combined QD and laser mode states $\{|11, N+1\rangle, |\psi_+, N\rangle\}$ differ by the detuning $\Delta \equiv \omega_0 - \omega_l + V_F$. For convenience, we group these states into a manifold $\mathcal{M}(N)$. Likewise, we group the pairs $\{|11, N\rangle, |\psi_+, N-1\rangle\}$, $\{|11, N+2\rangle, |\psi_+, N+1\rangle\}$ etc. into a ladder of manifolds differing in energy by steps ω_l . Introducing the dot-laser coupling mixes the bare eigenstates, again defining a dressed basis

$$|-\rangle_N = \cos\Theta|11, N+1\rangle - \sin\Theta|\psi_+, N\rangle, \quad (6.11)$$

$$|+\rangle_N = \sin\Theta|11, N+1\rangle + \cos\Theta|\psi_+, N\rangle, \quad (6.12)$$

with $\Theta = (1/2) \arctan(\sqrt{2}\Omega/\Delta)$ as before, and we use the subscript N to denote the manifold $\mathcal{M}(N)$. From this definition, it is clear that for $\Theta \neq 0$ both dressed states have some excitonic character and may decay into the adjacent manifold below, $\mathcal{M}(N-1)$ (see Fig. 5.3 in Chapter 5 for a similar scenario). Therefore, adiabatic following of the state $|\zeta_-\rangle \approx |-\rangle$ does not leave the operation immune to radiative decay. On the contrary, if the pulse is tuned all the way to resonance, $\Theta = \pi/4$, half the population will be susceptible to recombination.¹

To estimate the impact of spontaneous emission on the gate fidelity we need to consider the decay rate Γ_- from the $|-\rangle$ state, which is given by the sum of two

¹Analogous to direct resonant excitation where half the population remains in state $|11\rangle$ and half in state $|\psi_+\rangle$ on average.

processes: $|-\rangle_N \rightarrow |-\rangle_{N-1}$ and $|-\rangle_N \rightarrow |+\rangle_{N-1}$ with a total decay rate

$$\Gamma_- = \sqrt{2}\Gamma_0 \sin^2 \Theta \cos^2 \Theta + \sqrt{2}\Gamma_0 \sin^4 \Theta = \sqrt{2}\Gamma_0 \sin^2 \Theta, \quad (6.13)$$

where Γ_0 is defined as the single dot spontaneous emission rate. We note that Γ_- decreases as Θ becomes smaller, suggesting that keeping Θ small during the gating operation should be advantageous. Rewriting Eq. (5.8) as

$$\Gamma_- = \frac{\Gamma_0}{\sqrt{2}} \left(1 - \frac{1}{\sqrt{1 + (\sqrt{2}\Omega/\Delta)^2}} \right) \quad (6.14)$$

and expanding in a Taylor series around $\Omega/\Delta = 0$ for weak excitation ($\Theta \ll \pi/2$), yields

$$\Gamma_- = \frac{\Gamma_0}{\sqrt{2}} \left(\frac{\Omega}{\Delta} \right)^2 + \mathcal{O} \left(\frac{\Omega}{\Delta} \right)^4. \quad (6.15)$$

Hence, we realise that $\Gamma_- \sim \Delta^{-2}$ to leading order for fixed Ω .

The CPHASE gate requires that the phase accumulated on $|11\rangle$ exceeds that on $|01\rangle$ and $|10\rangle$ by π , the build-up being due essentially to V_F . During the pulse, $|11\rangle$ follows $|\zeta_-\rangle$ with an energy well approximated by $E_{\zeta_-} = (\Delta - \sqrt{\Delta^2 + 2\Omega^2})/2$, whereas $|01\rangle$ and $|10\rangle$ follow the dressed eigenstates in their respective subspaces, each of which has an energy $E_D = (\Delta - V_F - \sqrt{(\Delta - V_F)^2 + \Omega^2})/2$. Again, in the limit of a large detuning, we Taylor expand E_{ζ_-} around $\Omega/\Delta = 0$, yielding

$$E_{\zeta_-} = \frac{\Omega}{2} \left(\frac{\Omega}{\Delta} \right) + \mathcal{O} \left(\frac{\Omega}{\Delta} \right)^3. \quad (6.16)$$

Similarly, expanding E_D around $\Omega/(\Delta - V_F) = 0$ gives

$$E_D = \frac{\Omega}{4} \left(\frac{\Omega}{\Delta - V_F} \right) + \mathcal{O} \left(\frac{\Omega}{\Delta - V_F} \right)^3. \quad (6.17)$$

Therefore, assuming Ω is fixed for the moment and that phase accumulates as $\phi = \delta E t$, we obtain to leading order

$$\delta E = E_{\zeta_-} - 2E_D = -\frac{\Omega^2 V_F}{2\Delta(\Delta - V_F)}, \quad (6.18)$$

for the relevant energy shift during the CPHASE operation. For $\Delta \gg V_F$ the denominator is clearly dominated by Δ^2 , such that the required gating time t would simply take the form $t = \pi/(E_{\zeta_-} - 2E_D) \sim \Delta^2/V_F$, in the case of a square pulse. Of course, the adiabatic gate relies on a Gaussian pulse profile, yet the numerical data presented in the inset of Fig. 6.2 illustrates that the required Gaussian pulse duration τ also scales quadratically with Δ . Comparing with $\Gamma_- \sim \Delta^{-2}$ from before, we see that any decrease in the decay rate Γ_- by means of a larger detuning Δ is compensated for by the longer duration of the operation. Hence, contrary to the naive expectation, in this parameter regime applying a farther-detuned laser pulse does not improve the robustness of the adiabatic CPHASE gate to spontaneous emission.

To verify this simple analysis we perform a numerical simulation of the laser-driven CPHASE operation with a Gaussian pulse envelope as described by Eqs. (6.9) and (6.10) accounting for radiative decay with a standard quantum optical ME for the density matrix ρ of the system,

$$\dot{\rho} = -i[H_S, \rho] + \sum_i \left(L_i \rho L_i^\dagger - \frac{1}{2}(L_i^\dagger L_i \rho + \rho L_i^\dagger L_i) \right), \quad (6.19)$$

where L_i are the Lindblad operators as described below: Radiative recombination affects each dot separately in $\mathcal{H}_1$ and $\mathcal{H}_{1'}$, and the Lindblad operator describing spontaneous emission in both subspaces is therefore the same as it would be in the single dot case, $L_1 = \sqrt{\Gamma_0}|01\rangle\langle 0X|$ and $L_{1'} = \sqrt{\Gamma_0}|10\rangle\langle X0|$, respectively. In

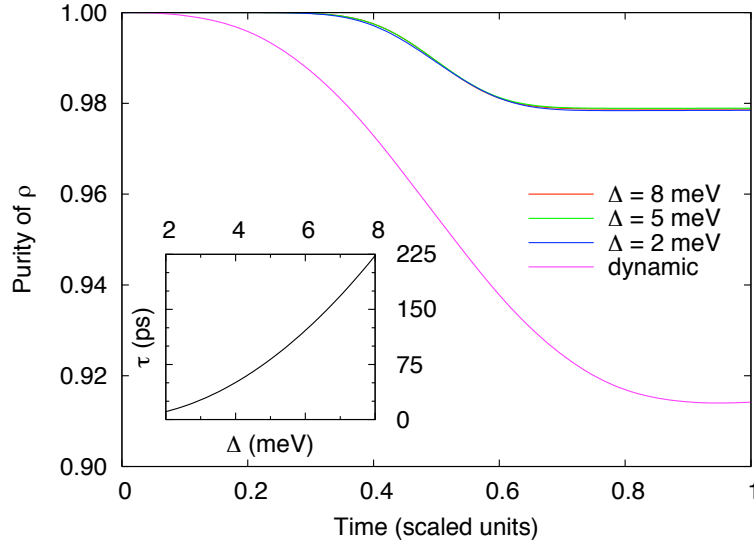


Figure 6.2: Purity of the density matrix for a CPHASE gate with $\Gamma_0 = 1/100 \text{ ps}^{-1}$, $V_{XX} = 5 \text{ meV}$ and $V_F = +0.85 \text{ meV}$. The dynamic gate uses $\Omega = 0.1 \text{ meV}$, whereas the adiabatic gates are performed at $\Omega_0 = 1 \text{ meV}$ with different Δ as indicated in the plot. The purity of the adiabatic operation does not depend on the detuning despite the substantial differences in the pulse duration, shown in the inset (see text for an explanation).

the larger subspace $\mathcal{H}_2$ transformation to the diagonal basis gives a single effective Lindblad operator, $L_2 = (\sqrt{2}\Gamma_0)^{1/2}|11\rangle\langle\psi_+|$, acting with respect to the Hamiltonian of Eq. (6.4).² Any thermally induced emission and absorption processes are neglected as the bosonic thermal occupancy $N(\omega_0)$ is extremely small even at room temperature.

Based on Eq. (6.19), we now present a comparison of the adiabatic CPHASE gate with the dynamic operation proposed in Ref. [135] and outlined in Section 6.2. As the figure of merit, we show the purity, $\text{tr}(\rho^2)$, of the full 9LS density matrix, including all decoupled subsystems, taking a general input state of the form $|\phi\rangle = (|00\rangle + |01\rangle + |10\rangle + |11\rangle)/2$. Figure 6.2 shows the results obtained for a natural excitonic lifetime of 0.1 ns. The adiabatic curves are plotted for $\Omega_0 = 1 \text{ meV}$

²No differentiation between positive and negative values of V_F is necessary throughout this section as the radiative decoherence channel is insensitive to the sign.

and for various Δ . All curves nearly coincide, clearly showing that the purity is indeed largely independent of Δ . Nevertheless, we note that the adiabatic gate retains a purity much closer to unity than the dynamic gate, the speed of which is limited by the conditions set out in Ref. [135].

6.4 Interaction with phonons

In contrast to an isolated atom, a QD is embedded in the solid state matrix of its substrate and its electronic state therefore couples to the phonon modes of the lattice (see Chapter 3) causing two-fold decoherence. First, we recall that pure dephasing – the dominant process at very low temperatures – is entirely eliminated when the coherent system excitation is sufficiently slow [139, 23, 140]. To study the second decoherence mechanism of phonon-assisted transitions, we derive a Markovian ME from first principles for the subspace $\mathcal{H}_2$. Since we are not interested in the regime of ultrafast driving (for the reason given above), we may neglect optical phonons, which are separated by a large energy gap of 30 meV or more [140], and concentrate on coupling to longitudinal acoustic phonons as the dominant phonon decoherence mechanism. For completeness, we start with the generic charge carrier phonon interaction Hamiltonian [81]

$$H_{ep} = \sum_{\mathbf{q}} M_{\mathbf{q}} \hat{\rho}(\mathbf{q}) \left(a_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^{\dagger} \right), \quad (6.20)$$

with coupling elements $M_{\mathbf{q}}$,

$$M_{\mathbf{q}} = \sqrt{\frac{\hbar}{2\mu V \omega_{\mathbf{q}}}} C_{\mathbf{q}}. \quad (6.21)$$

As before, μ is the mass density, V the lattice volume and $\omega_{\mathbf{q}}$ the phonon frequency, with wavevector $\mathbf{q}$. The coupling constant is $C_{\mathbf{q}} = D|\mathbf{q}|$ for the deformation potential and $C_{\mathbf{q}} = iP$ for piezoelectric coupling. The charge density operator $\hat{\rho}(\mathbf{q})$ is evaluated analogously as for Eq. (5.16), yielding

$$M_{\mathbf{q}}\hat{\rho}(\mathbf{q}) = \sum_i (M_{\mathbf{q}}^e \mathcal{P}[\psi_i^e(\mathbf{r})] - M_{\mathbf{q}}^h \mathcal{P}[\psi_i^h(\mathbf{r})]) \hat{c}_i^\dagger \hat{c}_i. \quad (6.22)$$

Here, $\hat{c}_i^\dagger$ ($\hat{c}_i$) denote excitonic creation (annihilation) operators and $\mathcal{P}[\psi^{e/h}]$ are the form factor of the electron and hole wavefunction, respectively. Once more neglecting the biexcitonic level $|XX\rangle$,³ the term (6.22) involves only single exciton levels, and Eq. (6.20) turns into

$$H_I = \sum_{\mathbf{q}} \left(g_{\mathbf{q}}^{0X} c_{0X}^\dagger c_{0X} + g_{\mathbf{q}}^{X0} c_{X0}^\dagger c_{X0} \right) (\hat{a}_{\mathbf{q}} + \hat{a}_{-\mathbf{q}}^\dagger), \quad (6.23)$$

where $g_{\mathbf{q}}^i = M_{\mathbf{q}}^e \mathcal{P}[\psi_i^e(\mathbf{r})] - M_{\mathbf{q}}^h \mathcal{P}[\psi_i^h(\mathbf{r})]$. If the wavefunctions $\psi_{0X}(\mathbf{r})$ and $\psi_{X0}(\mathbf{r})$ are of identical form, albeit centred at different positions $\pm \mathbf{d}$, we obtain by the shift property of the Fourier transform,

$$g_{\mathbf{q}}^{0X} = e^{+i\mathbf{q}\mathbf{d}} (M_{\mathbf{q}}^e \mathcal{P}[\psi^e(\mathbf{r})] - M_{\mathbf{q}}^h \mathcal{P}[\psi^h(\mathbf{r})]), \quad (6.24)$$

$$g_{\mathbf{q}}^{X0} = e^{-i\mathbf{q}\mathbf{d}} (M_{\mathbf{q}}^e \mathcal{P}[\psi^e(\mathbf{r})] - M_{\mathbf{q}}^h \mathcal{P}[\psi^h(\mathbf{r})]). \quad (6.25)$$

Meanwhile, in the absence of $|XX\rangle$ the two-dot system Hamiltonian Eq. (6.4) reduces to

$$H_2'' = (\Delta - 2V_F)|\psi_-\rangle\langle\psi_-| + \Delta|\psi_+\rangle\langle\psi_+| + \frac{\Omega}{\sqrt{2}}(|11\rangle\langle\psi_+| + \text{H.c.}), \quad (6.26)$$

³On resonance, this assumption requires $|V_{XX} - 2V_F| \gg |\Omega|$. However, for a larger detuning, Ω can be larger as well, and we have checked numerically that excitations to the $|XX\rangle$ level remain well below 10^{-5} for all parameters used in the following.

and the free Hamiltonian of a common bath of phonons reads

$$H_B = \sum_{\mathbf{q}} \omega_{\mathbf{q}} \hat{a}_{\mathbf{q}}^\dagger \hat{a}_{\mathbf{q}}. \quad (6.27)$$

We now diagonalise Eq. (6.26) and write H_I in the resulting basis $\{|\zeta_-\rangle, |\zeta_+\rangle, |\psi_-\rangle\}$, with respective eigenenergies $(\Delta - \sqrt{2\Omega^2 + \Delta^2})/2$, $(\Delta + \sqrt{2\Omega^2 + \Delta^2})/2$ and $\Delta - 2V_F$. Moving to the interaction picture with respect to $H_2'' + H_B$, the system operators are ordered by their instantaneous frequencies (see Appendix 5.A), giving

$$\begin{aligned} \tilde{H}_I(t) &= \frac{1}{2} \sum_{\omega' \in \{0, \Lambda, \Upsilon, \Xi\}} \left(P_{\omega'} e^{-i\omega' t} + P_{\omega'}^\dagger e^{i\omega' t} \right) \\ &\quad \times \sum_{\mathbf{q}} \left(g_{\mathbf{q}}^{0X} \pm g_{\mathbf{q}}^{X0} \right) \left(\hat{a}_{\mathbf{q}} e^{-i\omega_{\mathbf{q}} t} + \hat{a}_{-\mathbf{q}}^\dagger e^{i\omega_{\mathbf{q}} t} \right), \end{aligned} \quad (6.28)$$

where $\pm \equiv +$ for $\omega' \in \{0, \Lambda\}$ and $-$ for $\omega' \in \{\Upsilon, \Xi\}$. Here,

$$P_0 = \frac{1}{2} (\cos^2 \Theta |\zeta_+\rangle \langle \zeta_+| + \sin^2 \Theta |\zeta_-\rangle \langle \zeta_-| + |\psi_-\rangle \langle \psi_-|), \quad (6.29)$$

$$P_\Lambda = -\frac{1}{2} \sin 2\Theta |\zeta_-\rangle \langle \zeta_+|, \quad (6.30)$$

$$P_\Upsilon = \cos \Theta |\psi_-\rangle \langle \zeta_+|, \quad (6.31)$$

$$P_\Xi = -\sin \Theta |\zeta_-\rangle \langle \psi_-|, \quad (6.32)$$

with frequencies

$$\Lambda = \sqrt{2\Omega^2 + \Delta^2}, \quad (6.33)$$

$$\Upsilon = 2V_F - \frac{1}{2} (\Delta - (\sqrt{2\Omega^2 + \Delta^2})), \quad (6.34)$$

$$\Xi = \frac{1}{2} (\Delta + (\sqrt{2\Omega^2 + \Delta^2}) - 2V_F). \quad (6.35)$$

In this form, the phonon operators Eqs. (6.31, 6.32) are only appropriate for the detuned adiabatic gate with $\Delta > 2|V_F|$. Otherwise, care must be taken to ensure

all frequencies are greater than zero. Around resonance and for $V_F > 0$, Ξ switches sign (see Fig. 6.1) and we need to use $P_\Xi^\dagger$ with associated frequency $|\Xi|$ instead. On the other hand, for $V_F < \Omega/\sqrt{2}$, Υ switches sign requiring the redefinition of P_Υ to $P_\Upsilon^\dagger$ with associated frequency $|\Upsilon|$.

A master equation for the system evolution is now derived in the usual way [76, 77] by integrating the von Neumann equation for the joint density matrix of the system and bath and tracing over the phonon modes. The Born-Markov approximation is performed [76, 77] (valid in the weak-coupling regime, as explained in Chapter 3). If the system dynamics occurs on a timescale much faster than relaxation due to interactions with the bath, we can perform a RWA⁴ to arrive at an interaction picture ME in Lindblad form [76, 150]

$$\begin{aligned} \dot{\rho} = & J_+(\Lambda) \left([N(\Lambda) + 1] D[P_\Lambda] \rho + N(\Lambda) D[P_\Lambda^\dagger] \rho \right) \\ & + \sum_{\omega' \in \{\Upsilon, \Xi\}} J_-(\omega') \left([N(\omega') + 1] D[P_{\omega'}] \rho + N(\omega') D[P_{\omega'}^\dagger] \rho \right), \end{aligned} \quad (6.36)$$

with the dissipator $D[L]\rho \equiv L\rho L^\dagger - 1/2(L^\dagger L\rho + \rho L^\dagger L)$. Here,

$$N(\omega) = (\exp(\omega/k_B T) - 1)^{-1}$$

describes the thermal occupation of the phonon modes and $J_\pm(\omega)$ are the spectral densities given by

$$J_+(\omega) = 2\pi \sum_{\mathbf{q}} |g_{\mathbf{q}}^{0X} + g_{\mathbf{q}}^{X0}|^2 \delta(\omega - \omega_{\mathbf{q}}), \quad (6.37)$$

$$J_-(\omega) = 2\pi \sum_{\mathbf{q}} |g_{\mathbf{q}}^{0X} - g_{\mathbf{q}}^{X0}|^2 \delta(\omega - \omega_{\mathbf{q}}). \quad (6.38)$$

⁴The RWA requires $\Lambda, \omega' \gg J_\pm(\Lambda), J_\pm(\omega')$ the diagonal Lindblad-type ME only being strictly valid in this limit. Fortunately, the RWA assumption is indeed justified when $\Omega_0 < 1$ and $\Delta = 0$ as in the dynamic case or $\Delta \gtrsim \omega_e/h$ as will be used in the adiabatic case.

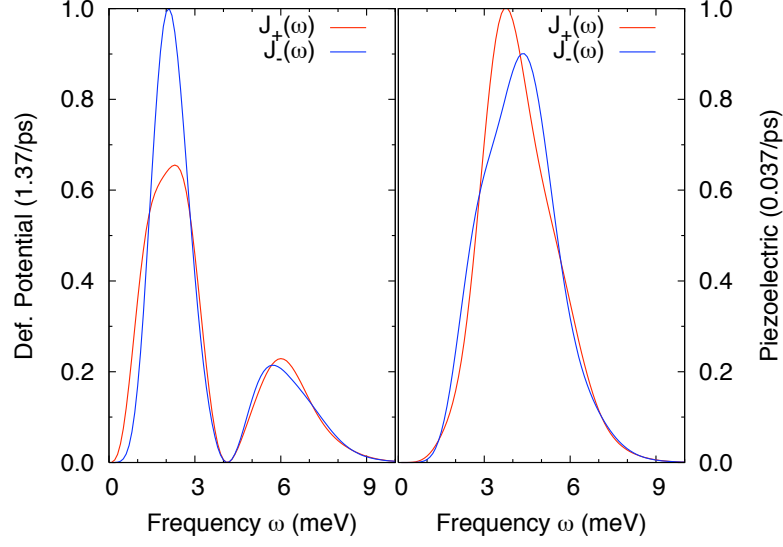


Figure 6.3: Normalised spectral densities $J_{\pm}(\omega)$: left, for deformation potential, and right, for piezoelectric coupling. Note the different y -axis scaling for each of the panels.

Note that the Lindblad operator P_0 has been dropped since the spectral density vanishes in the limit of $\omega = 0$, meaning the ME does not capture non-Markovian pure dephasing [139, 140] in the dressed basis, as expected.

For the calculation of $J_{\pm}(\omega)$ we choose the simple case of an isotropic harmonic confinement potential $V(\mathbf{r}) = f\mathbf{r}^2$ for each dot. The excitonic ground state therefore has the Gaussian wavefunction of Eq. (4.8),

$$\psi_{e/h}(\mathbf{r}) = \left(\frac{1}{d_{e/h}\sqrt{\pi}} \right)^{\frac{3}{2}} e^{-\frac{r^2}{2d_{e/h}^2}}, \quad (6.39)$$

where the width of the wavefunction envelopes is given by $d_{e/h} = (\hbar/\sqrt{m_{e/h}f})^{1/2}$, thus depending on the different effective masses $m_{e/h}$ for electrons and holes. We use a confinement potential realistic for InGaAs dots by choosing $f = 8.3 \times 10^{-3} \text{ J/m}^2$ such that electrons and holes are subject to a harmonic potential strength of 162 meV at $r = 2.5 \text{ nm}$ from the centre of the dot. Furthermore, we assume a dot centre-to-centre distance of $R = 7 \text{ nm}$, fulfilling the assumption of negligible wavefunction

overlap. Crucially, the dots are then still close enough to sustain a significant Förster interaction and dipolar shift [111]. Assuming a linear and isotropic phonon dispersion, $\omega_{\mathbf{q}} = c_s |\mathbf{q}|$, we then obtain for the deformation potential

$$J_{\pm}(\omega) = \frac{\omega^3}{4\pi\mu c_s^5} \left(1 \pm \text{sinc} \frac{\omega}{\omega_p} \right) \left(D_e e^{-\omega^2/\omega_e^2} - D_h e^{-\omega^2/\omega_h^2} \right)^2, \quad (6.40)$$

where $\omega_p = c_s/R$. Both spectral densities are obviously of superohmic form [84], with high-frequency cutoff terms $\omega_{e,h} = 2c_s/d_{e,h}$ related to the finite QD size. These terms filter out phonons with wavelengths too short to interact with the dots. For piezoelectric coupling we get spectral densities given by

$$J_{\pm}(\omega) = \frac{P^2\omega}{4\pi\mu c_s^3} \left(1 \pm \text{sinc} \frac{\omega}{\omega_p} \right) \left(e^{-\omega^2/\omega_e^2} - e^{-\omega^2/\omega_h^2} \right)^2. \quad (6.41)$$

All four spectral functions are plotted in Fig. 6.3 for the material parameters given in Table 5.1 from Section 5.4. From Fig. 6.3 we identify deformation potential coupling as the dominant phonon-decoherence mechanism (as one expects from the literature [139, 140]) and neglect piezoelectric coupling in the following. The dip between the two peaks in the left-hand side of Fig. 6.3 is a consequence of the difference between D_e and D_h combined with the slightly different electron and hole cut-off frequencies.⁵

Solving Eq. (6.36) allows us to characterise the performance of both adiabatic and dynamic operation of the CPHASE gate in the presence of the phonon bath. We find that the dynamic gate, with positive coupling $V_F = 0.85$ meV, fares poorly even at zero temperature due to phonon emission processes. In this case, $|\psi_{-}\rangle$ is the lowest energy eigenstate at resonance such that relaxation from both $|\zeta_{-}\rangle$ and $|\zeta_{+}\rangle$ into the dark state $|\psi_{-}\rangle$ is possible. By the definitions of Υ and Ξ in Eqs. (6.34) and

⁵For anisotropic dot wavefunctions we would still expect a dip, though it would not go all the way to zero. See Section 5.4 for a more detailed discussion of this feature.

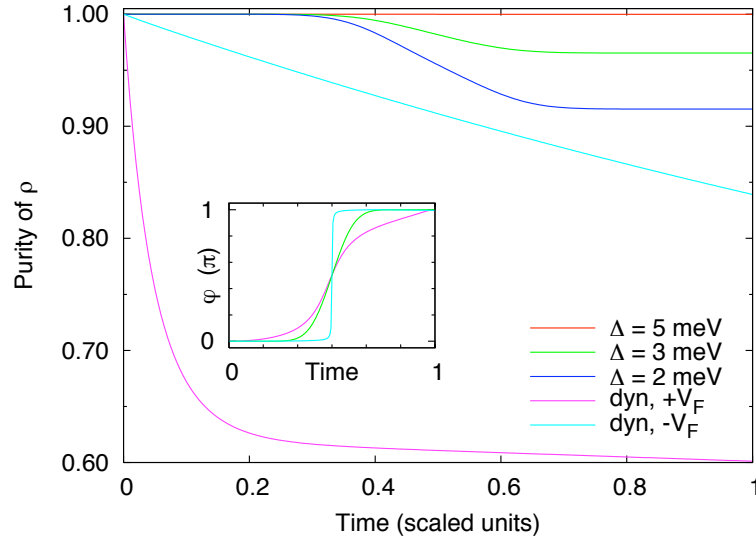


Figure 6.4: The purity of the full density matrix during a CPHASE operation at $T = 5$ K. The system is coupled to a phonon bath as described by the ME (6.36). For the dynamic operation, we show results for $V_F = \pm 0.85$ meV, as indicated by the labels “dyn, $+V_F$ ” and “dyn, $-V_F$ ”. The phase build-up of the dynamic gate (shown in the inset) should be step-like but is considerably smeared out in the case of the positive V_F , indicative of the fact that this approach does not work (see text).

(6.35), it is clear that a reduction of both these frequencies simultaneously to values much smaller than $2V_F$ is impossible. The result is that phonon emission processes, being roughly proportional to $J_{\pm}(2V_F)$, always operate at a fast rate, eventually transferring all population initially in $|11\rangle$ into $|\psi_{-}\rangle$. However, if the Förster coupling is negative, with $|V_F| > \Omega/\sqrt{2}$, the upwards shift of $|\psi_{-}\rangle$ is sufficiently large to lift it above both $|\zeta_{-}\rangle$ and $|\zeta_{+}\rangle$ at resonance and the dynamic scheme then works relatively well at low temperatures. In contrast, in the adiabatic approach phonon emission is always prevented for $\Delta > 2|V_F|$ because $|\zeta_{-}\rangle$ is then the ground state of the system, yielding a perfect gating operation at zero temperature. In fact, if V_F is negative this is even true irrespective of its particular value. At finite temperature we expect the adiabatic performance to improve with increasing detuning due to the rapid reduction in spectral density for large frequencies.

In Fig. 6.4, we plot the purity of the full 9LS density matrix at $T = 5$ K for both the dynamic and adiabatic approaches. The purity of the dynamic gate with positive Förster coupling drops quickly to a value of around 0.62, corresponding to the expected state where all population has been transferred into $|\psi_{-}\rangle$. Of course, decoherence also occurs in $\mathcal{H}_1$ and $\mathcal{H}_{1'}$, and this is included in the calculation as illustrated by the slow and steady decline of the curve as time progresses. As expected, the performance of the adiabatic gate improves dramatically with increasing Δ by pushing both Λ and Ξ beyond the spectral cutoff,⁶ meaning $J(\Lambda)$ and $J(\Xi)$ decay exponentially. Physically, this implies that phonons with an energy matching the transitions between the relevant system eigenstates no longer interact with the dot because of their short wavelengths. This advantageous regime sets in at an energy below 10 meV, allowing us safely to assume a linear phonon dispersion and ignore optical phonon modes.

6.5 Landau-Zener transitions

Non-adiabatic LZ transitions [160, 161, 162] between system eigenstates are a further error source in the adiabatic scheme and can only be avoided by varying the control parameters more slowly. There is, of course, a limit as to how slowly things can be done, since typically many operations need to be performed within the lifetime of the electron spin.

Unfortunately, the exact theory for the final LZ transition amplitude cannot generally be applied to driven qubits, and the Schrödinger equation needs to be solved numerically [151]. Using such an approach, Fig. 6.5 illustrates that some of the population undergoing LZ transitions during the pulse returns into $|-\rangle$ state once the pulse has finished. Nevertheless, these temporary transitions are still undesirable

⁶ Υ does not increase with larger Δ , but this turns out to be unimportant as P_{Υ} couples states which, ideally, remain unpopulated.

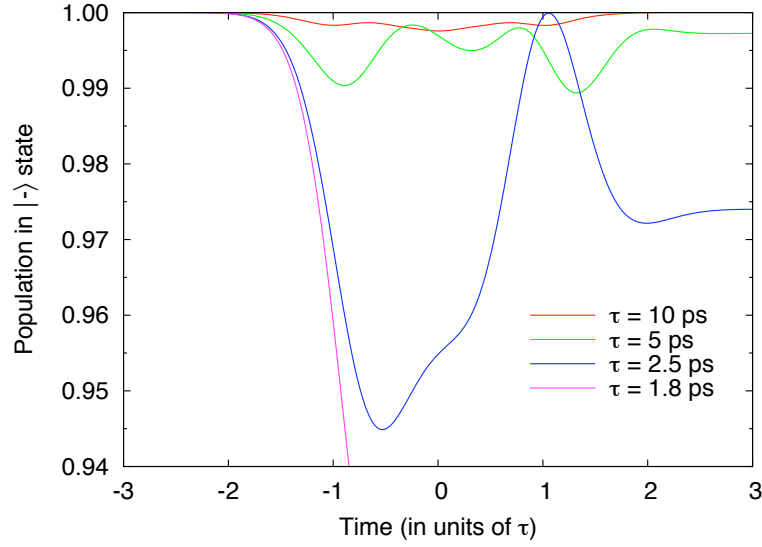


Figure 6.5: The population remaining in the $|-\rangle$ ($\approx |\zeta_{-}\rangle$) state of the $\mathcal{H}_2$ subspace is shown for different pulse durations τ of a laser pulse $\Omega(t) = \Omega_0 \exp[-(t/\tau)]$ and $\Omega_0 = \Delta = 1 \text{ ps}^{-1}$. In this case the adiabaticity condition (6.45) requires $\tau \gg \sqrt{2} \text{ ps}$. The final population leakage for the $\tau = 1.8 \text{ ps}$ curve is roughly 8.5 percent and off the scale of this plot.

as they make it difficult to accurately predict the final phase achieved during the adiabatic operation. Furthermore, any population transferred to the excited state is much more susceptible to decoherence processes.

To derive an adiabaticity condition for the CPHASE gate we again consider the 2LS approximation to Eq. (6.4), valid for large positive detuning. In fact, in this regime the level spacing between $|\zeta_{-}\rangle$ and the upper dressed state $|\zeta_X\rangle$ is significantly larger than that between $|\zeta_{-}\rangle$ and $|\zeta_{+}\rangle$. Furthermore, the coupling between $|\zeta_{-}\rangle$ and $|\zeta_X\rangle$ originates from the coupling of $|\psi_{+}\rangle$ to $|XX\rangle$, but at large detuning $|\zeta_{-}\rangle$ only contains a small admixture of $|\psi_{+}\rangle$. Therefore, the probability of LZ transitions from $|\zeta_{-}\rangle$ to $|\zeta_X\rangle$ is doubly small and it is sufficient to consider only those between $|\zeta_{-}\rangle$ and $|\zeta_{+}\rangle$.

We can now follow the same procedure already used in Section 5.5 by performing (the time-dependent) transformation to the basis of the approximate instantaneous

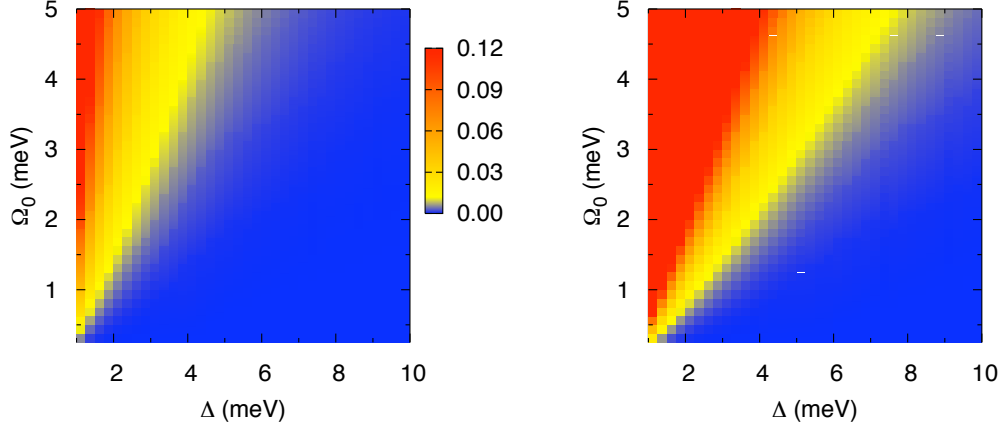


Figure 6.6: Comparison of adiabaticity conditions. **Left:** the left-hand side of inequality (6.44) is plotted as a function of detuning and coupling strength. The pulse duration τ is chosen to generate a phase shift of π . The blue colour denotes a region where LZ transitions are adiabatically suppressed, i.e. where the LHS of (6.44) is much smaller than one. Towards the yellow region, it approaches values of $1/100$ and the approximation starts to break down. The red colour denotes a value of $1/10$ or more and adiabatic following can no longer be expected. **Right:** the same for the LHS of Eq. (6.45) divided by τ . We observe the same qualitative behaviour as in the left part of the plot, however, this simplified form of the adiabatic condition is more stringent than the previous one, confirming that it guarantees adiabatic following, but is not necessarily required for it.

eigenstates $|\zeta_{-}\rangle$ and $|\zeta_{+}\rangle$, now yielding

$$\tilde{H} = \lambda^{-}|\zeta_{-}\rangle\langle\zeta_{-}| + \lambda^{+}|\zeta_{+}\rangle\langle\zeta_{+}| + \dot{\Theta}(i|\zeta_{-}\rangle\langle\zeta_{+}| + \text{H.c.}), \quad (6.42)$$

where $2\Theta = \arctan(\sqrt{2}\Omega/\Delta)$, and where $\lambda^{\pm} = 1/2(\Delta \pm \sqrt{\Delta^2 + 2\Omega^2})$ denote the approximate instantaneous eigenenergies. As before, adiabatic following requires that the magnitude of the off-diagonal terms must be much smaller than the energy

difference between the eigenstates:

$$|\dot{\Theta}| \ll |\lambda^+ - \lambda^-|.^7 \quad (6.43)$$

By inserting Θ and $\lambda^\pm$ we obtain to the following general adiabaticity condition:

$$\left| \frac{\dot{\Omega}\Delta - \Omega\dot{\Delta}}{\sqrt{2}(\Delta^2 + 2\Omega^2)^{3/2}} \right| \ll 1. \quad (6.44)$$

If the temporal evolution of Ω and Δ is known, this condition can be brought into the form $F(\Delta_0, \Omega_0) \ll \tau$, where $F(\Delta_0, \Omega_0)$ is a time-independent relation of known parameters and τ gives the characteristic time of the applied pulse. For instance, in the particular case considered here (i.e. $\Omega = \Omega_0 \exp(-t^2/\tau^2)$ and constant Δ) an upper bound for adiabaticity is found to be

$$\sqrt{2}\Omega_0/\Delta^2 \ll \tau. \quad (6.45)$$

Fig. 6.6 shows that the adiabaticity condition of Eq. (6.45) is indeed more stringent than that of Eq. (6.44). Referring back to Section 5.5, we recall that the same condition also holds in the subspaces $\mathcal{H}_1$ and $\mathcal{H}_{1'}$ with the simple change $\sqrt{2}\Omega \rightarrow \Omega$.

Clearly, LZ transitions do not occur in limit in which phonon transitions are suppressed, i.e. $\Delta \gg \Omega_0$, thus allowing us to circumvent both error sources simultaneously.

6.6 CPHASE gate fidelity

Bringing together the results of the previous sections, we now calculate the overall fidelity of the CPHASE operation, obtained from a numerical solution to a

⁷We note that this condition can be shown to be equivalent to the more familiar condition $\langle \zeta_- | \frac{d}{dt} | \zeta_+ \rangle \ll |\lambda^+ - \lambda^-|$ [151].

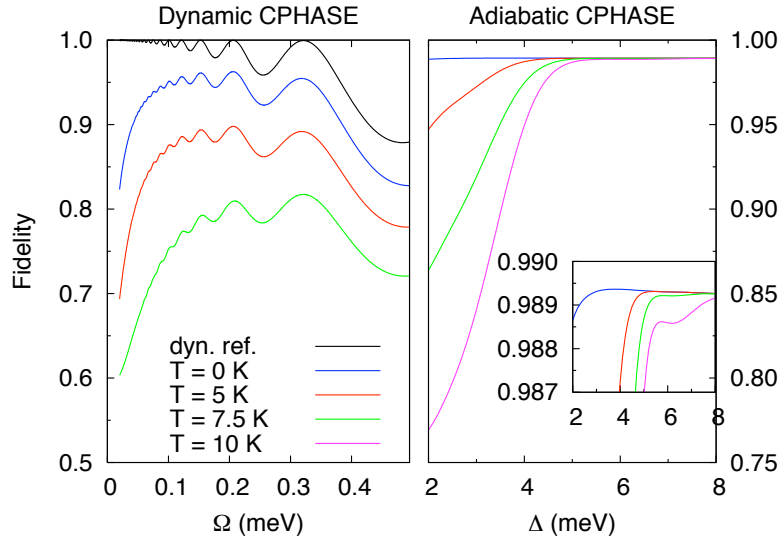


Figure 6.7: Fidelity of the CPHASE gate. **Left:** fidelity of the dynamic operation for a negative $V_F = -0.85$ meV. The dynamic reference curve gives the fidelity without additional decoherence (see text). **Right:** fidelity of the adiabatic operation for fixed $\Omega_0 = 1$ meV. The inset shows the same curves on a different scale to resolve more subtle features.

ME that includes both spontaneous emission as well as phonon-induced processes (and, of course, allows for LZ transitions). We take an input state given by $|\phi_i\rangle = (|00\rangle + |01\rangle + |10\rangle + |11\rangle)/2$, which, after the CPHASE operation, should ideally produce an output state $|\phi_f\rangle = (|00\rangle + |01\rangle + |10\rangle - |11\rangle)/2$. In practice, for both the adiabatic and dynamic approach some phase is picked up in the $\mathcal{H}_1$ and $\mathcal{H}_{1'}$ subspaces, which needs to be ‘unwound’ using two single qubit operations. However, we here only consider the genuine two-qubit part of the operation, i.e. meeting the condition of Eq. (6.8). We therefore define the fidelity as $\mathcal{F} = \langle \phi'_f | \rho | \phi'_f \rangle$, where $|\phi'_f\rangle$ accommodates the additional phases on $|10\rangle$ and $|01\rangle$, and ρ is the full density matrix of the system after the gate has finished, including all detrimental environmental effects. As before, we use the material parameters of Table 5.1 and assume a single dot radiative decay rate of $\Gamma_0 = 0.01$ ps⁻¹.

The results are plotted in Fig. 6.7. The left-hand side shows the fidelity of

the dynamic gate with $V_F = -0.85$ meV as a function of the coupling strength Ω , and for different temperatures. A laser resonant with the optical transition of $\mathcal{H}_2$ will inevitably lead to detuned Rabi oscillations in both $\mathcal{H}_1$ and $\mathcal{H}_{1'}$, affecting the fidelity unless the Rabi periods of all subspaces are commensurate. This degradation of the fidelity is included and, as expected, the effect is oscillatory and becomes more pronounced as Ω increases [135] (to further illustrate this point, we have omitted all other sources of decoherence for the curve labelled “dyn. ref.”). The fidelity is limited by the finite excitonic lifetime towards small values of Ω , while to the right of the plot phonon-induced processes become more important. The best fidelity achieved here is roughly 0.95 at absolute zero, and decreases even further at finite temperature.

The contrasting behaviour of the adiabatic scheme is shown in right-hand side of Fig. 6.7. Here, the gate fidelity is plotted as a function of the detuning Δ , for a fixed value of $\Omega_0 = 1$ meV. In agreement with the conclusions of the previous sections, the fidelity can be substantially increased by simply applying a suitably large detuning to the driving laser. In this limit, the fidelity is restricted by spontaneous emission, as the detrimental effects of LZ and phonon-induced transitions are confined to relatively small values of Δ . Nonetheless, for $\Delta \gtrsim 5$ meV the fidelity is greater than 0.98 for all temperatures shown and would improve even further for a smaller spontaneous emission rate (the rate used here translates into a rather pessimistic excitonic lifetime of 0.1 ns). For interest, the inset shows an intermediate peak only visible at higher temperatures, which is related to the dip in the spectral density of the deformation potential (see Fig. 6.3).

6.7 Conclusion

To summarise, we have performed a realistic decoherence study of both the adiabatic and dynamic approaches to exciton-mediated spin manipulation in coupled QDs. We have shown that while dynamic gates suffer from rapid decoherence at finite temperatures (to an extent that may prohibit fault-tolerant QC), performing off-resonant adiabatic manipulations allows us to greatly suppress decoherence during the entangling operations. On the other hand, the adiabatic scheme looks more promising. Since no upper bound is imposed on the gate duration by radiative decay, a detuned and slow adiabatic operation is suitable for alleviating the adverse effects of both phonon-induced decoherence and LZ transitions. In this case, it is the finite spin coherence time that sets a bound on the possible gate duration. However, the adiabatic gates we consider operate on timescales of around 100 ps, much shorter than typical spin coherence times [93, 116] and only about an order of magnitude greater than the ‘fast’ dynamic gates. We therefore conclude that adiabatic optical manipulation is a remarkably robust method for entangling spin qubits embodied in semiconductor nanostructures.

Moreover, similar adiabatic schemes would also work for a different form of coupling in the $\mathcal{H}_2$ subspace, since any coupling will shift the energy levels and therefore lead to the accumulation of a non-trivial phase. Importantly, the adiabatic entangling protocol presented in this chapter is not far from the realm of experimental capability, as only a single (unchirped) control laser with a suitably shaped pulse envelope is required, given that a suitable coupled nanostructure can be identified.

Chapter 7

Phonon-exciton processes in quantum dots

Understanding the interaction between electronic states in solids and lattice vibrations is fundamentally interesting. In this chapter, we derive a weak-coupling Markovian master equation for describing the system dynamics of an excitonic two level system coupled to a reservoir of phonons. In contrast to previous studies, we also include ‘phase markers’ in the derivation, allowing us to simultaneously monitor the number of excitations in the environment. As a second part, we employ the system part of such a Markovian master equation to experimental data, finding an excellent agreement over a large parameter space and for a range of temperatures.

7.1 Introduction

The coherent dynamics of the driven two level system is of utmost importance not only for the field of quantum information processing but also in the wider context of understanding the physics of many real-world quantum systems.

In solid state systems, the crystal ground state and a trapped electron hole pair

(exciton) define a two level system which can be controlled optically. However, such an excitonic qubit is not isolated and the coupling to the lattice dynamics of the surrounding semiconductor material is known to be significant [140, 167]. Therefore, we must expect substantial deviations from the dynamics of an idealised two level system (that can often be realised by atomic systems). To a very good approximation, the normal phonon modes of the lattice can be described as independent bosons [81], and the coupling of a quantum 2LS to such a bosonic bath has been extensively studied in literature, e.g. in Leggett's seminal dissertation on the 'dissipative two level system' [84], and also in Refs. [86, 85].

Whereas previous theoretical treatments have fully discarded all information about the state of the environment [140, 132, 168, 153], we here present a technique allowing us to track the number of phonon excitations as a distinguishing feature of the work presented in the first part of this chapter.

7.2 Derivation of the Master Equation

7.2.1 The Hamiltonian

Consider a self-assembled quantum dot illuminated by a laser beam with frequency ω_l , which is nearly resonant with the crystal ground state to exciton transition. In a frame rotating with the laser frequency and within the RWA, the system is governed by the Hamiltonian ($\hbar = 1$).

$$H_S(t) = \frac{\epsilon}{2}\sigma_z + \frac{\Omega(t)}{2}\sigma_x, \quad (7.1)$$

where the detuning ϵ describes the energy difference between the basis states in the rotating frame, $\Omega(t)$ is the time-dependent Rabi frequency coupling the two basis states, and where σ_x and σ_z are the usual pseudo-spin operators defined with

respect to the quantum dot ground state $|g\rangle$ and single-exciton state $|e\rangle$.

Let us further assume that this excitonic qubit is weakly coupled to a bath of phonons, resulting in the total Hamiltonian:

$$H(t) = H_S(t) + H_B + H_I, \quad (7.2)$$

where

$$H_B = \sum_{\mathbf{q}} \omega_{\mathbf{q}} \hat{a}_{\mathbf{q}}^\dagger \hat{a}_{\mathbf{q}} \quad (7.3)$$

is the free Hamiltonian of the phonon modes with $\hat{a}_{\mathbf{q}}^\dagger$ and $\hat{a}_{\mathbf{q}}$ being the creation and annihilation operators of a phonon in mode $\mathbf{q}$ with frequency $\omega_{\mathbf{q}}$. The exciton phonon interaction term is (see Chapter 3)

$$H_I = \sigma_z \sum_{\mathbf{q}} g_{\mathbf{q}} (\hat{a}_{\mathbf{q}}^\dagger + \hat{a}_{\mathbf{q}}), \quad (7.4)$$

where the $g_{\mathbf{q}}$ are coupling constants. The transformation into the interaction picture with respect to $H_S(t)$ and H_B is then achieved with the unitary operator

$$U(t) = T \exp \left(-i \int_{t_0}^t d\tau H_S(\tau) \right) e^{-iH_B(t-t_0)}, \quad (7.5)$$

where T is the time ordering operator and we have chosen the Schrödinger and the interaction picture to coincide at the arbitrarily chosen initial time $t_0 = 0$. Using this time evolution operator, we can formally write down the transformed interaction Hamiltonian as

$$\tilde{H}_I(t) = \tilde{\sigma}_z(t) \sum_{\mathbf{q}} g_{\mathbf{q}} (\hat{a}_{\mathbf{q}}^\dagger e^{i\omega_{\mathbf{q}}t} + \hat{a}_{\mathbf{q}} e^{-i\omega_{\mathbf{q}}t}), \quad (7.6)$$

where $\tilde{\sigma}_z(t)$ denotes the transformed σ_z operator. Since $H_S(t)$ generally does not commute with itself at different times, it is difficult to write down an explicit ex-

pression for $\tilde{\sigma}_z(t)$ at this stage, but this problem will be addressed further down and in Appendix 7.A.

7.2.2 Phonon excitation counting statistics

Before deriving the master equation, let us introduce the formalism for monitoring phonon excitations in the quantum dot's solid state environment. Mathematically, this is achieved by a gauge transformation, which adds phase markers to the interaction Hamiltonian; crucially, these do not alter the system's dynamics. The technique presented below is adapted from the literature on observing charge transport in the quantum regime described in Refs. [169, 170, 171], and is also related to the problem of photon counting [77].

Quantum mechanics predicts probabilities rather than giving access to the precise state of a system in configuration space. While this prevents us from counting the exact number of phonon excitations in the environment, we can instead obtain a probability distribution. The probability of having emitted n phonons into the environment can be formally expressed with the help of a projection operator P_n acting on the joint density matrix ϱ of the system and environment [170]:

$$p_n = \text{tr}(P_n \varrho). \quad (7.7)$$

Equivalently, the part of the wavefunction corresponding to n excitations can be tagged with the phase marker $e^{in\lambda}$, an operation that is conveniently written in terms of a gauge transformation operator $\mathcal{E}_\lambda$ [172, 171],

$$\mathcal{E}_\lambda = e^{i\lambda \sum_{\mathbf{q}} \hat{n}(\mathbf{q})}, \quad (7.8)$$

where $\hat{n}(\mathbf{q}) = \hat{a}_{\mathbf{q}}^\dagger \hat{a}_{\mathbf{q}}$ is the number operator. The projection to a state with n exci-

tations is then simply achieved by filtering out the component with the appropriate fixed phase. Hence, we may write the projection operator as an integration over the phase as proposed by Anderson [172]

$$P_n = \frac{1}{2\pi} \int_0^{2\pi} d\lambda e^{-in\lambda} \mathcal{E}_\lambda. \quad (7.9)$$

Importantly, the inverse relation also holds:

$$\mathcal{E}_\lambda = \sum_n e^{in\lambda} P_n. \quad (7.10)$$

Knowing how to project onto a state with a given number of environmental phonon excitations, we shall discuss how to express a change from n initial excitations at time zero to $n - m$ excitations at a later time t . In the Heisenberg picture, this conditional probability is given by [171]

$$p(n - m, t | n, 0) = \text{tr} (P_n(0) \varrho P_n(0) P_{n-m}(t)). \quad (7.11)$$

However, when we are only interested in the change of the number of excitations m , the initial number n is unimportant. Hence, summing over n gives

$$p_m(t) = \sum_n \text{tr} (P_n \varrho P_n U^\dagger(t) P_{n-m} U(t)), \quad (7.12)$$

where we have expressed the time evolution of $P_{n-m}(t)$ with the time evolution operator $U(t)$. Using Eq. (7.9) for each of the projection operators results in three integrals, one of which drops out owing to the identity $\sum_n e^{in\gamma} = 2\pi\delta(\gamma)$, yielding

$$p_m(t) = \frac{1}{(2\pi)^2} \int d\gamma_1 d\gamma_2 e^{-im(\gamma_1 + \gamma_2)} \text{tr} (\mathcal{E}_{\gamma_1} \varrho \mathcal{E}_{\gamma_2} U^\dagger(t) \mathcal{E}_{-\gamma_1 - \gamma_2} U(t)). \quad (7.13)$$

To simplify this expression further, we exploit the cyclicity of operators under the trace, as well as the following properties of the gauge transformation operator:

$\mathcal{E}_{-\gamma_1-\gamma_2} = \mathcal{E}_{-\gamma_2}\mathcal{E}_{-\gamma_1}$ and $\mathcal{E}_\gamma = \mathcal{E}_{-\gamma}^\dagger$. Thus, we obtain

$$p_m(t) = \frac{1}{(2\pi)^2} \int d\gamma_1 d\gamma_2 e^{-im(\gamma_1+\gamma_2)} \text{tr} \left(\varrho U_{-\gamma_2}^\dagger(t) U_{\gamma_1}(t) \right), \quad (7.14)$$

with the following definition of the gauge transformed time evolution operator $U_\lambda(t) = \mathcal{E}_\lambda^\dagger U(t) \mathcal{E}_\lambda$. Performing a variable substitution $\lambda = \gamma_1 + \gamma_2$ and $2\Lambda = \gamma_1 - \gamma_2$, we can rewrite Eq. (7.14) as

$$p_m(t) = \frac{1}{2\pi} \int_0^{2\pi} d\lambda e^{-im\lambda} p_\lambda(t), \quad (7.15)$$

where $p_\lambda(t)$ is the counting generating function of Refs. [171, 170], defined as

$$p_\lambda(t) = \frac{1}{2\pi} \int_0^{2\pi} d\Lambda \text{tr} \left(\varrho U_{\Lambda-\frac{\lambda}{2}}^\dagger(t) U_{\Lambda+\frac{\lambda}{2}}(t) \right). \quad (7.16)$$

When the initial density matrix $\varrho_0 = \varrho(0)$ describes a diagonal excitation state (rather than a superposition of different excitation numbers),¹ we may instead use a simpler variant of the generating function [170, 169]:

$$p_\lambda(t) = \text{tr} (\varrho_\lambda(t)), \quad (7.17)$$

where we have introduced the gauge transformed density matrix:

$$\varrho_\lambda(t) = U_{\frac{\lambda}{2}}(t) \varrho_0 U_{-\frac{\lambda}{2}}^\dagger(t). \quad (7.18)$$

¹We shall henceforth label the initial state of the environment as possessing zero excitations, meaning that no phonons have yet been absorbed or emitted into the environment by the quantum dot. Note that this does not imply that the environment must be in its ground state, though.

7.2.3 Gauge transformed density matrix master equation

In order to derive a phonon master equation, we must first establish the appropriate von Neumann equation of the gauge transformed density matrix. The first time-derivative of Eq. (7.18) is (in the interaction picture)

$$\dot{\varrho}_\lambda(t) = \dot{U}_{\frac{\lambda}{2}}(t)\varrho_0 U_{-\frac{\lambda}{2}}^\dagger(t) + U_{\frac{\lambda}{2}}(t)\varrho_0 \dot{U}_{-\frac{\lambda}{2}}^\dagger(t) \quad (7.19)$$

$$= -i \left(\mathcal{E}_{\frac{\lambda}{2}}^\dagger \tilde{H}_I U(t) \mathcal{E}_{\frac{\lambda}{2}} \varrho_0 U_{-\frac{\lambda}{2}}^\dagger(t) - U_{\frac{\lambda}{2}}(t) \varrho_0 \mathcal{E}_{-\frac{\lambda}{2}}^\dagger \tilde{H}_I U^\dagger(t) \mathcal{E}_{-\frac{\lambda}{2}} \right). \quad (7.20)$$

By using the commutation relations $P_{n-1}\hat{a}_{\mathbf{q}} = \hat{a}_{\mathbf{q}}P_n$ and $P_{n+1}\hat{a}_{\mathbf{q}}^\dagger = \hat{a}_{\mathbf{q}}^\dagger P_n$ and the gauge transformation operator representation of Eq. (7.10), we can interchange the order of $\mathcal{E}_{\pm\frac{\lambda}{2}}^{(\dagger)}$ and $\tilde{H}_I$ to arrive at the gauge transformed interaction picture von Neumann equation

$$\dot{\varrho}_\lambda(t) = -i \left(\tilde{H}_\lambda(t) \varrho_\lambda(t) - \varrho_\lambda(t) \tilde{H}_{-\lambda}(t) \right), \quad (7.21)$$

where the interaction Hamiltonian (7.6) has acquired phase markers as a consequence of the operator reordering:

$$\tilde{H}_\lambda = \tilde{\sigma}_z(t) \sum_{\mathbf{q}} g_{\mathbf{q}} (e^{-i\frac{\lambda}{2}} \hat{a}_{\mathbf{q}}^\dagger e^{i\omega_{\mathbf{q}}t} + e^{i\frac{\lambda}{2}} \hat{a}_{\mathbf{q}} e^{-i\omega_{\mathbf{q}}t}) = \tilde{\sigma}_z(t) B_\lambda(t). \quad (7.22)$$

We proceed by formally integrating Eq. (7.21) and substituting the resulting expression back into the right-hand side of the von Neumann equation. After tracing over the environment, we obtain for the reduced density matrix of the system ρ_λ

$$\begin{aligned} \dot{\rho}_\lambda(t) = -\text{tr}_E \int_0^t ds \quad & (H_\lambda(t) H_\lambda(s) \varrho_\lambda(s) - H_\lambda(t) \varrho_\lambda(s) H_{-\lambda}(s) \\ & + \varrho_\lambda(s) H_{-\lambda}(s) H_{-\lambda}(t) - H_\lambda(s) \varrho_\lambda(s) H_{-\lambda}(t)). \end{aligned} \quad (7.23)$$

The Born-Markov approximation is now performed (see Chapter 3 or Refs. [76, 77] for details). The Born approximation assumes that it is justified to write the joint density matrix as a product at all times $\varrho_\lambda(s) = \rho_\lambda(s) \otimes \rho_E$. Within the Markov approximation, we replace $\rho(s)$ by $\rho(t)$, and take the upper integration limit to infinity.² With $B_\lambda(t)$ from Eq. (7.22), the ME then reads

$$\begin{aligned} \dot{\rho}_\lambda(t) = & - \int_0^\infty ds \quad \langle B_\lambda(s) B_\lambda(0) \rangle \tilde{\sigma}_z(t) \tilde{\sigma}_z(t-s) \rho_\lambda(t) \\ & - \langle B_{-\lambda}(0) B_\lambda(s) \rangle \tilde{\sigma}_z(t) \rho_\lambda(t) \tilde{\sigma}_z(t-s) \\ & + \langle B_{-\lambda}(0) B_{-\lambda}(s) \rangle \rho_\lambda(t) \tilde{\sigma}_z(t-s) \tilde{\sigma}_z(t) \\ & - \langle B_{-\lambda}(s) B_\lambda(0) \rangle \tilde{\sigma}_z(t-s) \rho_\lambda(t) \tilde{\sigma}_z(t). \end{aligned} \quad (7.24)$$

To simplify the further algebraic evaluation, we express the operator $\tilde{\sigma}_z(t)$ in terms of instantaneous system eigenoperators, justified in Appendix 7.A, yielding

$$\tilde{\sigma}_z(t) = \sum_{\omega \in \{0, \Lambda\}} (e^{-i\omega t} P_\omega + e^{i\omega t} P_\omega^\dagger), \quad (7.25)$$

where $\Lambda = \sqrt{\Omega^2 + \Delta^2}$ now denotes the spacing between the instantaneous system eigenstates $|-\rangle = \cos \theta |g\rangle - \sin \theta |e\rangle$ and $|+\rangle = \cos \theta |e\rangle + \sin \theta |g\rangle$ of Hamiltonian (7.1) with $2\theta = \arctan \Omega/\epsilon$. In this basis, $P_0 = \cos 2\theta (|-\rangle\langle -| - |+\rangle\langle +|)/2$ and $P_\Lambda = \sin 2\theta |-\rangle\langle +|$.

By introducing the Hermitian operator $\tilde{\mathcal{P}}$,

$$\tilde{\mathcal{P}} = 2P_0 + e^{-i\Lambda t} P_\Lambda + e^{i\Lambda t} P_\Lambda^\dagger, \quad (7.26)$$

²Note that the integration variable s has also been substituted by $t-s$ before the integration limit is extended.

and abbreviating

$$\tilde{\mathcal{L}} = e^{-i\Lambda t} P_\Lambda, \quad (7.27)$$

we obtain the following interaction picture master equation after some straightforward algebra:³

$$\begin{aligned} \dot{\rho}_\lambda &= \Gamma_\downarrow \left(e^{-i\lambda} \left(\tilde{\mathcal{L}} \rho_\lambda \tilde{\mathcal{P}}^\dagger + \tilde{\mathcal{P}} \rho_\lambda \tilde{\mathcal{L}}^\dagger \right) - \tilde{\mathcal{P}}^\dagger \tilde{\mathcal{L}} \rho_\lambda - \rho_\lambda \tilde{\mathcal{L}}^\dagger \tilde{\mathcal{P}} \right) \\ &+ \Gamma_\uparrow \left(e^{i\lambda} \left(\tilde{\mathcal{P}}^\dagger \rho_\lambda \tilde{\mathcal{L}} + \tilde{\mathcal{L}}^\dagger \rho_\lambda \tilde{\mathcal{P}} \right) - \tilde{\mathcal{P}} \tilde{\mathcal{L}}^\dagger \rho_\lambda - \rho_\lambda \tilde{\mathcal{L}} \tilde{\mathcal{P}}^\dagger \right), \end{aligned} \quad (7.28)$$

where the rates are given by $\Gamma_\downarrow = J(\Lambda) (N(\Lambda) + 1) / 2$ and $\Gamma_\uparrow = J(\Lambda) N(\Lambda) / 2$ when ρ_E is in a thermal state. Further,

$$J(\omega) = 2\pi \sum_{\mathbf{q}} |g_{\mathbf{q}}|^2 \delta(\omega - \omega_{\mathbf{q}}) \quad (7.29)$$

is the spectral density of phonon modes and $N(\omega) = (\exp(\beta\omega) - 1)^{-1}$ denotes the thermal occupancy of a phonon mode with frequency ω .

Upon going back to the Schrödinger picture, the operators $\tilde{\mathcal{P}}$ and $\tilde{\mathcal{L}}$ lose their time-dependent phase factors, turning into $\mathcal{P} = \sigma_z$ and $\mathcal{L} = P_\Lambda$, respectively. Using the density matrix relations⁴

$$\rho_n = \frac{1}{2\pi} \int_0^{2\pi} d\lambda e^{-in\lambda} \rho_\lambda, \quad \rho_\lambda = \sum_m e^{im\lambda} \rho_m, \quad (7.30)$$

we transform the ME (7.28) to a set of coupled differential equations for the evolution

³As in previous ME derivations, we neglect frequency renormalisations due to principal value terms.

⁴The first of these is essentially Eq. (7.15) applied to the excitation-specific system density matrix, $\rho_n(t) = \text{tr}_E(\varrho(t)P_n)$, while the second is the inverse relation (for a diagonal initial state [171]).

of excitation-specific system density matrices:

$$\begin{aligned} \dot{\rho}_n = -i[H_S(t), \rho_n] &+ \Gamma_{\downarrow} (\mathcal{L}\rho_{n+1}\mathcal{P}^{\dagger} + \mathcal{P}\rho_{n+1}\mathcal{L}^{\dagger} - \mathcal{P}^{\dagger}\mathcal{L}\rho_n - \rho_n\mathcal{L}^{\dagger}\mathcal{P}) \\ &+ \Gamma_{\uparrow} (\mathcal{P}^{\dagger}\rho_{n-1}\mathcal{L} + \mathcal{L}^{\dagger}\rho_{n-1}\mathcal{P} - \mathcal{P}\mathcal{L}^{\dagger}\rho_n - \rho_n\mathcal{L}\mathcal{P}^{\dagger}). \end{aligned} \quad (7.31)$$

When summing over all ρ_n of Eq. (7.31), the resulting ME is – as expected – fully equivalent to Eq. (3.23) from Section 3.2.1 evaluated with Eq. (7.4) as the interaction Hamiltonian.

Further, a ME in diagonal Lindblad form (as used in the previous chapters) is obtained from Eq. (7.31) by setting $\mathcal{P} = \mathcal{L}$, thereby performing the RWA in the derivation of the ME that we have encountered in Chapter 3:

$$\begin{aligned} \dot{\rho}_n = -i[H_S(t), \rho_n] &+ \Gamma_{\downarrow} \left(2P_{\Lambda}\rho_{n+1}P_{\Lambda}^{\dagger} - P_{\Lambda}^{\dagger}P_{\Lambda}\rho_n - \rho_nP_{\Lambda}^{\dagger}P_{\Lambda} \right) \\ &+ \Gamma_{\uparrow} \left(2P_{\Lambda}^{\dagger}\rho_{n-1}P_{\Lambda} - P_{\Lambda}P_{\Lambda}^{\dagger}\rho_n - \rho_nP_{\Lambda}P_{\Lambda}^{\dagger} \right). \end{aligned} \quad (7.32)$$

The RWA is expected to be valid whenever $\Lambda \gg J(\Lambda)$ [76, 150]. However, we find that it often provides a good description outside this strict limit, as it comprises all the diagonal (energy conserving) terms, while neglecting the less important off-diagonal processes, which give rise to a small renormalisation of the effective Rabi frequency.

7.3 Phonon absorption and emission

We shall now apply our master equation (7.31) to an excitonic qubit optically driven by a pulse of constant intensity, or with a Gaussian temporal intensity profile. Initially, the system is in the ground state at $t = 0$ with zero excitations in the environment: $\rho_n(0) = \delta_{n,0}|g\rangle\langle g|$. As system parameters, we use $\Omega_0 = 1 \text{ ps}^{-1}$ ($\approx 2/3 \text{ meV}$)

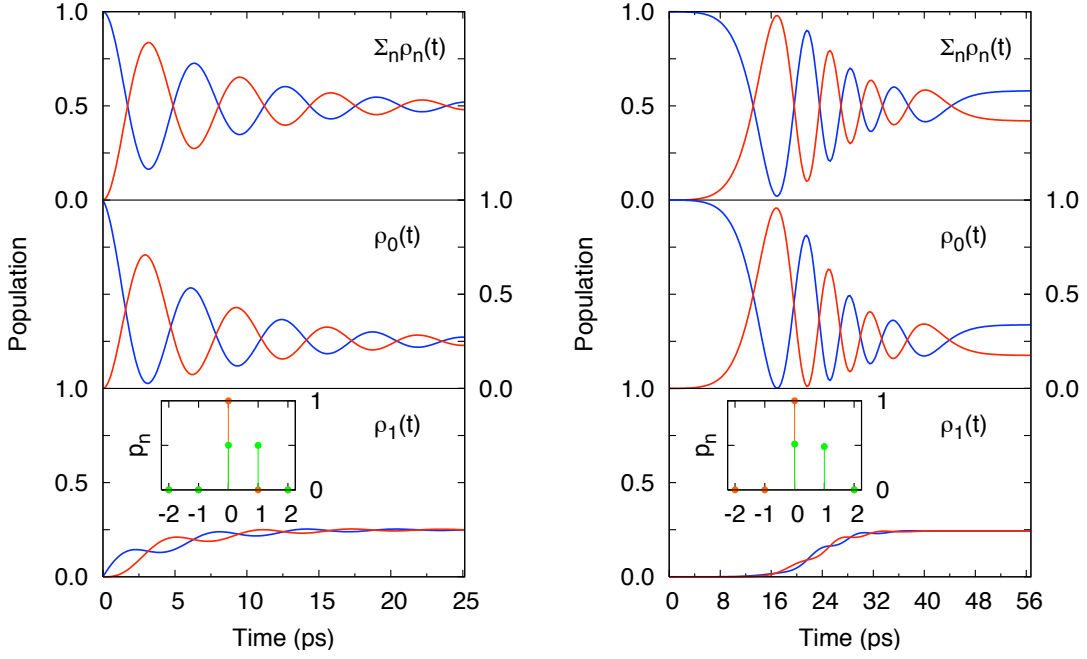


Figure 7.1: Population in $|g\rangle$ (blue) and $|e\rangle$ (red) of different excitation specific density matrices up to a final pulse area of 8π for resonant excitation at $T = 0$ K. Left for rectangular excitation and right for a Gaussian pulse centred around $t_0 = 16\sqrt{\pi}$ ps ≈ 28 ps ($\tau = 4\sqrt{\pi}$ ps). The insets show the initial (orange) and final (green) p_n distribution as a function of n . See text for further parameters.

and $\Omega(t) = \Omega_0$ for the rectangular pulse, whereas $\Omega(t) = \Omega_0 \exp(-(t - t_0)^2/\tau^2)$ for the Gaussian pulse. For simplicity, we define the spectral density phenomenologically: $J(\omega) = \alpha\omega^3 \exp(-\omega^2/\omega_c^2)$, where α describes the effective deformation potential coupling strength and ω_c is the high frequency phonon cut-off. For relatively weak driving with a peak Rabi frequency Λ well below both the electron and the hole cut-off, we can neglect the exponential cut-off term altogether. Setting $\alpha = 1/4$ ps² yields a coupling strength that is consistent with the magnitude of the GaAs deformation potential interaction reported in the literature [23, 140].

Figure 7.1 shows the damping of resonant Rabi oscillations of $\rho(t) = \sum_n \rho_n(t)$, $\rho_0(t)$ and $\rho_1(t)$ at zero temperature, where only phonon emission is possible. The population transfer from ρ_0 to ρ_1 is reminiscent of an exponential saturation curve.

The small ripples are a consequence of the off-diagonal Lindblad terms of Eq. (7.31); they disappear when the RWA is performed.

Even though the laser beam carries a continuous supply of energy, no more than one phonon is emitted, which becomes apparent when analysing the structure of Eq. (7.32): The system is initialised in the state $|g\rangle\langle g| = (|-\rangle + |+\rangle)(\langle -| + \langle +|)/2$, i.e. in an equal superposition of system eigenstates. The Lindblad operator P_Λ induces a transition from $|+\rangle$ to $|-\rangle$ while $P_\Lambda^\dagger$ raises population from $|-\rangle$ to $|+\rangle$. Once a decay process has happened, we find the system in the (instantaneous) $|-\rangle$ state, meaning it cannot decay again, unless the system Hamiltonian, and with it the eigenbasis, undergo a sufficient change, such that the $|-\rangle$ state at time t features a $|+\rangle$ component of the new eigenbasis at a later time $t + \delta t$. However, for resonant excitation, the system eigenstates are independent of time – regardless of the pulse shape. Conversely, for a detuned pulse, the eigenbasis is time-dependent unless the (optical) excitation is constant.

Provided the excitation is sufficiently long, the population ratios tend to a Boltzmann distribution, as is obvious from the phonon emission rate proportional to $N(\Lambda) + 1$ and the absorption rate proportional to $N(\Lambda)$,

$$\lim_{t \rightarrow \infty} \frac{\text{tr}(\rho_0(t)|+\rangle\langle +|)}{\text{tr}(\rho_1(t))} = \lim_{t \rightarrow \infty} \frac{\text{tr}(\rho_{-1}(t))}{\text{tr}(\rho_0(t)|-\rangle\langle -|)} = \frac{N(\Lambda)}{N(\Lambda) + 1} = e^{-\beta\Lambda}. \quad (7.33)$$

This fully explains the $T = 0$ K situation shown in Figure 7.1: all population that is initially in the $|+\rangle$ state of ρ_0 relaxes to the $|-\rangle$ state of ρ_1 , while the initial population in $|-\rangle$ remains in ρ_0 .

Let us now consider the case of detuned, time-dependent excitation with off-resonant Gaussian pulses. Generally speaking, the initial state $|g\rangle$ consists of a growing component of $|-\rangle$ at the expense of $|+\rangle$ as the detuning increases. Consequently, phonon absorption begins to dominate over emission, even at low temper-

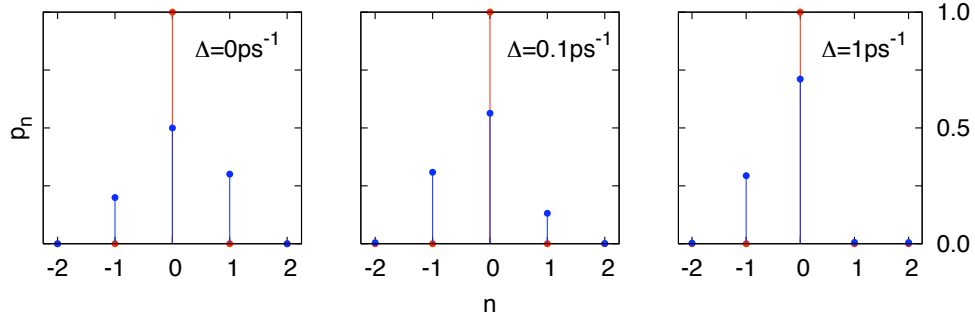


Figure 7.2: Initial (red) and final (blue) distribution of $\text{tr}(\rho_n)$ for different amounts of detuning. As before, $\Omega_0 = 1 \text{ ps}^{-1}$ with a pulse duration $\tau = 4\sqrt{\pi} \text{ ps}$ that corresponds to a pulse area of 8π on resonance.

ature. Figure 7.2 shows the final $p_n = \text{tr}(\rho_n)$ distribution for different amounts of detuning at $T = 10 \text{ K}$. The leftmost plot shows the resonant case, i.e. an equal superposition of $|-\rangle$ and $|+\rangle$, where phonon emission is more probable than absorption. However, even for a relatively small value of $\Delta = \Omega_0/10$, there is a higher net absorption than emission rate, while practically no phonon emission occurs in the right plot of Figure 7.2. This is because at $\Delta \gtrsim \Omega_0$, and beyond, we achieve adiabatic following of the $|-\rangle$ state (unless τ is very short, see Chapter 5), such that emission becomes impossible. However, a larger detuning also reduces the $\sin 2\theta$ prefactor of the $P_\Lambda^\dagger$ operator, ultimately suppressing all phonon-induced transitions.

Perhaps surprisingly, the detuned time-dependent excitation barely populates ρ_2 or ρ_{-2} , in spite of the fact the instantaneous eigenstates are now functions of time; we have checked numerically that the population in ρ_2 (ρ_{-2}) is more than two orders of magnitude smaller than that of ρ_1 (ρ_{-1}). This reflects the fact that the system dynamics is essentially determined by the peak Rabi frequency Ω_0 , where the ratio of (optical) coupling strength to detuning is largest and where the ‘mixing angle’ θ only changes slowly.

So far, we have been concerned with the case where the only perturbation to the system is caused by the coupling to the phonon bath. For the final part of this

section, we introduce additional phenomenological noise operators, such as pure dephasing and an alternative (radiative) relaxation channel. We model these processes with an additional Lindblad dissipator [76],

$$\Gamma \left(L\rho L^\dagger - \frac{1}{2}(L^\dagger L\rho + \rho L^\dagger L) \right), \quad (7.34)$$

added to the right-hand side of Eq. (7.31). The Lindblad operators $L = \sigma_z$ and $L = \sigma_-$ do not preserve the system's eigenstates; consequently, under their action, phonon-assisted transitions become possible in any of the ρ_n subspaces. Remarkably, this leads to a dynamic equilibrium, with phonon-assisted transitions occurring after the transient and coherent evolution of $\rho = \sum_n \rho_n$ has subsided. The counting statistics technique developed in this chapter is well-suited for illustrating this behaviour: Fig. 7.3 presents the p_n distribution at different points of time for both dephasing and decay, in both cases showing a broadening distribution with a shifting peak.

The pure dephasing σ_z operator randomises the phase between $|g\rangle$ and $|e\rangle$, thus balancing the population of $|-\rangle$ and $|+\rangle$. Consequently, once the steady state has been reached, phonon emission always occurs with a faster rate than absorption, and the distribution is therefore shifted in the direction of increasing n , meaning the average number of emitted phonons increases. On the other hand, the distribution can move in either direction for a relaxation process from $|e\rangle$ to $|g\rangle$. For $\Delta = 0$, $|g\rangle$ consists of an equal superposition of $|-\rangle$ and $|+\rangle$, while it contains a larger $|-\rangle$ component for $|\Delta| > 0$. Under these latter circumstances, it is possible for phonon absorption to permanently dominate over emission, shifting the distribution in the direction of decreasing n , as shown in the upper right panel of Fig. 7.3. In this case, thermal energy is removed from the QD's bulk surroundings and released into the wider environment by spontaneous photon emission.

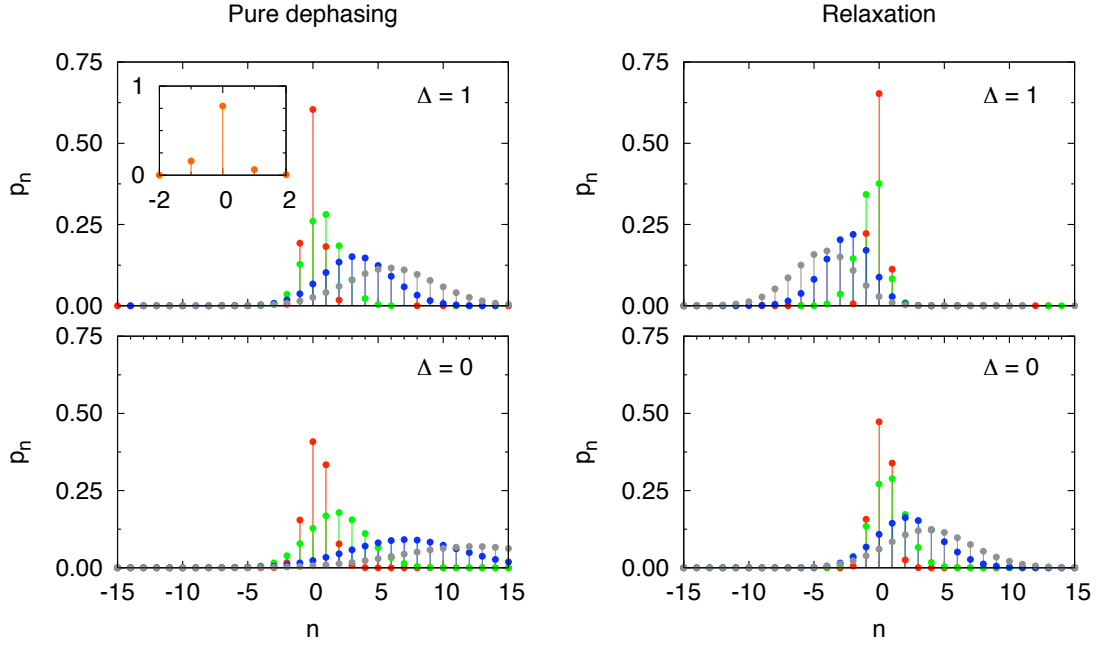


Figure 7.3: The distribution of p_n at different points of time in the presence of additional noise. The constant excitation pulse uses $\Omega_0 = 1 \text{ ps}^{-1}$ and a detuning Δ as indicated in the plots (in ps^{-1}). The noise rates are fixed at $\Gamma = 0.1 \text{ ps}^{-1}$. The colour-coding refers to time as follows: red = 1.2, green = 10, blue = 40, grey = 70, all in units of full Rabi cycles, $2\pi/\sqrt{\Omega_0^2 + \Delta^2}$. The upper left inset shows that phonon absorption is still more likely at very short times ($t = 0.4$) before the σ_z -Lindblad operator becomes effective.

To quantify this observation, we proceed by calculating the rate of phonon emission or absorption for the more interesting case of radiative relaxation. This is given by the following expression [171]

$$\begin{aligned} \langle \dot{n}(t) \rangle &= \frac{d}{dt} \sum_n n p_n = i \frac{d}{d\lambda} \text{tr}(\dot{\rho}_\lambda) \Big|_{\lambda=0} \\ &= 2 \text{tr} (\Gamma_\downarrow \mathcal{L} \rho(t) \mathcal{L}^\dagger - \Gamma_\uparrow \mathcal{L}^\dagger \rho(t) \mathcal{L}), \end{aligned} \quad (7.35)$$

where $\rho(t)$ is obtained by integrating Eq. (7.28) in the Schrödinger picture (after the RWA, i.e. setting $\mathcal{P} = \mathcal{L}$), and inclusive of the Lindblad dissipator (7.34) with $L = \sigma_-$.

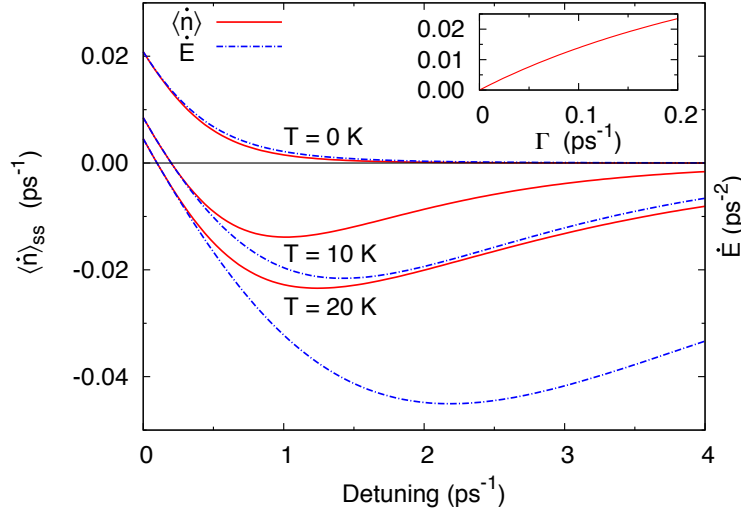


Figure 7.4: The rate of phonon-induced transitions (red) and the rate of energy transfer (blue), both as a function of the detuning. The rates shown here are the steady state values of Eq. (7.35) for a decay rate $\Gamma = 0.1 \text{ ps}^{-1}$. A positive sign indicates phonon emission whereas a negative corresponds to phonon absorption. The inset shows $\langle \dot{n} \rangle_{ss}$ for fixed $\Delta = 1 \text{ ps}^{-1}$ as a function of Γ at $T = 10 \text{ K}$.

Fig. 7.4 presents the steady-state value $\langle \dot{n} \rangle_{ss}$ of Eq. (7.35) as a function of the detuning. As expected, net phonon absorption becomes only possible at finite temperature for off-resonant excitation. It is a natural question to ask at which rate energy is transferred to or from the surroundings of the system. Considering the quantity $\dot{E} = \sqrt{\Omega^2 + \Delta^2} \langle \dot{n} \rangle_{ss}$ shows that the number of absorbed phonons decreases for a larger detuning, yet their energy is greater, shifting the Δ that achieves optimal heat transfer. The inset of Fig. 7.4 shows that the process is limited by the radiative decay time Γ of the system. A saturation only occurs when Γ becomes faster than the phonon-mediated transitions, but this regime would require an unrealistic optical lifetime of the order of a picosecond or less, invalidating our Markovian ME model.

For context, we estimate the achievable cooling rate for realistic parameters in SI units. Using $\Omega_0 = \Delta = 1 \text{ ps}^{-1}$ gives a phonon energy of $\Lambda = 1.49 \times 10^{-22} \text{ J}$. With approximately 0.02 absorbed phonons per picosecond (at $T = 20 \text{ K}$), we obtain a

theoretical energy transfer rate of roughly 3×10^{-12} J/s. While this sounds small, it could still induce a temperature change of the order of one Kelvin per second in a micrometre cube of GaAs.⁵ However, seeing a cooling effect in practice would be extremely challenging, as the required laser illumination is likely to heat the sample at a much faster rate.

As an aside, we note that the results presented here are largely independent of the superohmic structure of the spectral density. In fact, we get qualitatively very similar results for an Ohmic or a flat spectral density.⁶ In particular, the spectral density's prefactor determines the overall magnitude of the process, while its functional dependence on ω mainly affects the decay of the tails of the curves in Fig. 7.4.

To summarise, we have adapted a counting statistic approach from the context of charge transfer to an excitonic system coupled to an environment consisting of normal phonon modes. Using this technique, we glean information about how a dissipative, driven 2LS acts as a heat pump, removing energy from its environment by means of repeated phonon absorption in conjunction with spontaneous photon emission. In the next part of this chapter, we shall apply a similar master equation (but without phase markers) to experimental data.

7.4 Temperature dependence of phonon-induced damping

The cause of the experimentally observed intensity-dependent damping of optically driven excitonic qubits in QDs has been a subject of intense debate. Several mechanisms have been put forward: excitation of hybridised wetting layer states [173, 174],

⁵Using a mass density of $\mu = 5.3 \times 10^3$ kg/m³ and a specific heat of around 350 J/(kg K) gives a heat capacity of 1.85×10^{-12} J/K.

⁶Provided that $\omega_c \gtrsim 2$, including the cut-off only leads to a faster decay of the tail.

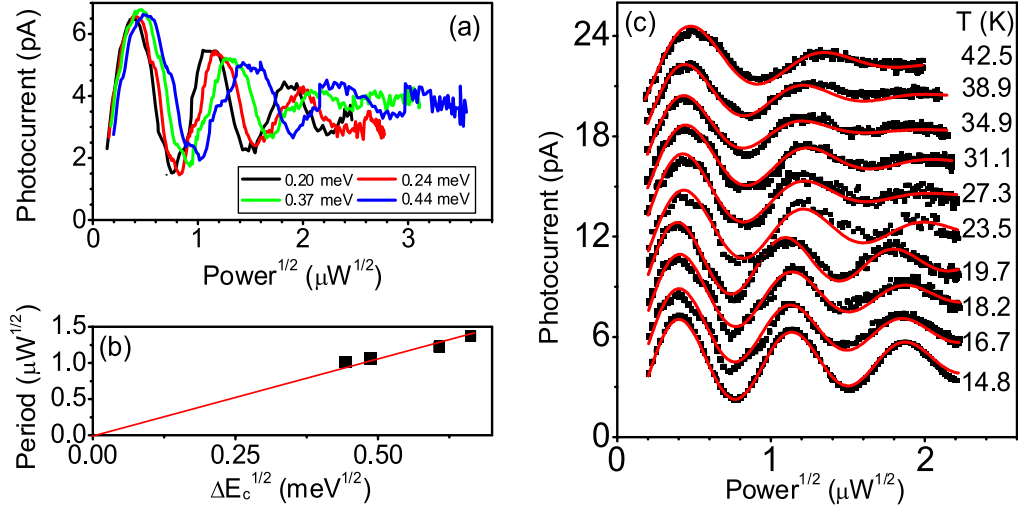


Figure 7.5: Photocurrent versus the square root of incident power (proportional to the pulse area) for resonant excitation. **(a)** Rabi rotations for pulses with different spectral FWHM ΔE_c . The envelope is independent of ΔE_c , but the Rabi period is proportional to $\sqrt{\Delta E_c}$ as shown in **(b)**. **(c)** Temperature dependence of measured Rabi oscillations (black squares) and fits to the theory (red lines). Figure adapted from Ref. [175].

multi-exciton excitation and acoustic phonon mediated dephasing [142, 153]. Here, we present a theoretical model for the damping due to longitudinal acoustic phonons. Importantly, we find that the temperature dependence of this model is in agreement with measured data, in contrast to the competing model of wetting layer induced dephasing.

Fig. 7.5 shows the measured Rabi rotations in a InGaAs/GaAs dot embedded in the intrinsic GaAs region of an n-i-Schottky diode at a bias of 0.6 V, where the signal is measured as the current produced by charge carriers tunnelling out of the QD on a timescale exceeding 60 ps. Thus, the photocurrent is directly proportional to the final excitonic population after the much shorter control pulse has finished. For full details of the measurement and experimental setup see Refs. [176, 175].

We shall now develop a theoretical model, beginning with the familiar Hamiltonian of the driven 2LS coupled to a bath of phonons. The QD crystal ground-state

$|0\rangle$ to single-exciton state $|X\rangle$ is driven by a control laser of Rabi frequency

$$\Omega(t) = (\Theta/2\tau\sqrt{\pi})\exp(-(t/2\tau)^2),$$

which is detuned from resonance by $\Delta \equiv \omega_0 - \omega_l$. Here, ω_0 is the exciton creation energy ($\hbar = 1$) and ω_l the laser frequency. In a frame rotating at frequency ω_l , and after a RWA on the driving field, the QD exciton Hamiltonian is

$$\begin{aligned} H_C &= \Delta|X\rangle\langle X| + \frac{\Omega(t)}{2}(|0\rangle\langle X| + |X\rangle\langle 0|) \\ &= \frac{\Delta}{2}(\mathbf{1} + \sigma_z) + \frac{\Omega(t)}{2}\sigma_x, \end{aligned} \quad (7.36)$$

with the standard definition of the Pauli operators σ_x and σ_z , and where $\mathbf{1}$ is the identity. The QD charge configuration interacts with a reservoir of acoustic phonons of wavevector $\mathbf{q}$, with Hamiltonian $H_B = \sum_{\mathbf{q}} \omega_{\mathbf{q}} \hat{b}_{\mathbf{q}}^\dagger \hat{b}_{\mathbf{q}}$, in the regime of linear dispersion, $\omega_{\mathbf{q}} = c_s |\mathbf{q}|$, with speed of sound c_s . The dominant deformation potential coupling of a localised exciton to the phonon bath is given by (see Eq. (5.13) or Chapter 3)

$$\begin{aligned} H_I &= |X\rangle\langle X| \sum_{\mathbf{q}} g_{\mathbf{q}} (\hat{b}_{\mathbf{q}}^\dagger + \hat{b}_{\mathbf{q}}) \\ &= \frac{1}{2}(\mathbf{1} + \sigma_z) \sum_{\mathbf{q}} g_{\mathbf{q}} (\hat{b}_{\mathbf{q}}^\dagger + \hat{b}_{\mathbf{q}}) \equiv \hat{S} \hat{R}, \end{aligned} \quad (7.37)$$

where $\hat{b}_{\mathbf{q}}^\dagger$ ($\hat{b}_{\mathbf{q}}$) are the creation (annihilation) operators for phonons of frequency $\omega_{\mathbf{q}}$.

The deformation potential coupling strengths are [81]

$$g_{\mathbf{q}} = \frac{q(D_e \mathcal{P}[\psi^e(\mathbf{r})] - D_h \mathcal{P}[\psi^h(\mathbf{r})])}{\sqrt{2\mu\hbar\omega_{\mathbf{q}}V}}, \quad (7.38)$$

where μ is the mass density of the host material, V the lattice volume, $D_{e(h)}$ the

respective bulk electron (hole) coupling constant, and $\mathcal{P}[\psi^{e(h)}]$ denotes the form factor of the electron (hole) wavefunction (see Eq. (5.16)). The total Hamiltonian of the QD and phonon system is therefore written as $H = H_C + H_B + H_I$.

To derive a master equation governing the driven-dot dynamics we now transform the Hamiltonian into the interaction picture with respect to its non-interacting part, $H_0 = H_C + H_B$:

$$H_I(t) = S_I(t) \sum_{\mathbf{q}} g_{\mathbf{q}} \left(\hat{b}_{\mathbf{q}}^{\dagger} e^{i\omega_{\mathbf{q}} t} + \hat{b}_{\mathbf{q}} e^{-i\omega_{\mathbf{q}} t} \right). \quad (7.39)$$

For resonant excitation $\Delta = 0$, the system part of the interaction Hamiltonian is most conveniently expressed in the diagonal basis of H_C , i.e. in the basis $|-\rangle = (|0\rangle - |X\rangle)/\sqrt{2}$ and $|+\rangle = (|0\rangle + |X\rangle)/\sqrt{2}$, as

$$S_I(t) = \frac{1}{2} \left(\mathbf{1} - e^{-i \int_{t_0}^t \Omega(t') dt'} \tilde{\sigma}_- - e^{i \int_{t_0}^t \Omega(t') dt'} \tilde{\sigma}_+ \right), \quad (7.40)$$

where $\tilde{\sigma}_+ = |+\rangle\langle-|$ and $\tilde{\sigma}_- = |-\rangle\langle+|$ are the raising and lowering operators of the diagonal basis. As the time dependence of the driving pulse envelope $\Omega(t)$, and the resulting time integral in Eq. (7.40), complicate the subsequent derivation of the master equation [177], we replace them by the instantaneous value $\tilde{\Omega}$ of the Rabi frequency, justified when $\Omega(t)$ changes only slowly compared to the bath relaxation time (as outlined in Appendix 7.A). This condition is satisfied in the experiments: the 14 ps FWHM of the laser pulses is large compared with the expected 1 ps relaxation time of the acoustic phonon bath [82, 167, 153]. Hence, we approximate $S_I(t)$ as

$$S_I(t) = \frac{1}{2} \left(\mathbf{1} - e^{-i\tilde{\Omega}t} \tilde{\sigma}_- - e^{i\tilde{\Omega}t} \tilde{\sigma}_+ \right). \quad (7.41)$$

We are now in a position to derive the dot-phonon master equation by integrating the von Neumann equation for the joint system-bath density matrix χ , then tracing over the phonon modes, which results in an integro-differential equation for the

reduced exciton density matrix ρ ,

$$\dot{\rho} = - \int_0^t ds \operatorname{tr}_{ph} ([H_I(t), [H_I(s), \chi(s)]]). \quad (7.42)$$

The Born-Markov approximation (see Chapter 3) is now performed, allowing us to factorise $\chi(t) = \rho(t) \otimes \rho_B$, where ρ_B describes the phonon bath in thermal equilibrium. Furthermore, we replace $\rho(s)$ by $\rho(t)$ in Eq. (7.42), based on the assumption that the timescale over which the excitonic system varies appreciably is large compared with the phonon bath relaxation time. Thus, we arrive at the time local Redfield equation,

$$\dot{\rho} = - \int_0^t ds' \operatorname{tr}_{ph} ([H_I(t), [H_I(t-s'), \rho(t) \otimes \rho_B]]), \quad (7.43)$$

where we have also made the substitution $s = t - s'$. Taking the upper integration limit to infinity and transforming back into the Schrödinger picture, we obtain the following master equation in the excitonic basis after some algebra:⁷

$$\begin{aligned} \dot{\rho} &= -\frac{i\Omega(t)}{2}[\sigma_x, \rho] - \frac{\pi}{8}J(\Omega(t))\coth\left(\frac{\Omega(t)}{2k_B T}\right)[\sigma_z, [\sigma_z, \rho]] \\ &= -\frac{i\Omega(t)}{2}[\sigma_x, \rho] - \frac{K(\Omega(t))}{4}[\sigma_z, [\sigma_z, \rho]], \end{aligned} \quad (7.44)$$

where we have reintroduced the time-dependence of $\Omega(t)$, since the relevant timescale in Eq. (7.44) is no longer that of the bath correlations, but of the system dynamics. Further, we neglect small frequency shifts and terms which do not influence the population dynamics, as we only measure the excitonic population in the experiment.

⁷A detailed account of the algebra can be found in the Supplemental Material [178] of Ref. [175].

The term $J(\omega)$ in Eq. (7.44) denotes the spectral density of phonon modes,

$$J(\omega) = \sum_{\mathbf{q}} |g_{\mathbf{q}}|^2 \delta(\omega - \omega_{\mathbf{q}}) = \frac{(D_e - D_h)^2 \omega^3}{4\pi^2 \mu c^5}, \quad (7.45)$$

under the assumption that $\mathcal{P}[\psi^{e,h}(\mathbf{r})] \approx 1$, as is appropriate for relatively weak driving below the cut-off frequency.

The exciton coherence $\rho_{0X} = \langle 0|\rho|X \rangle$ evolves according to Eqs. (7.44) and (7.45)

$$\dot{\rho}_{0X} = i \frac{\Omega(t)}{2} (1 - 2\rho_{XX}) - (\Gamma_2^* + K(\Omega)) \rho_{0X}, \quad (7.46)$$

where $\rho_{XX} = \langle X|\rho|X \rangle$ is the population of the excitonic state, in turn obeying

$$\dot{\rho}_{XX} = i \frac{\Omega(t)}{2} (\rho_{X0} - \rho_{0X}), \quad (7.47)$$

and Γ_2^* is a phenomenological, *field-independent* rate accounting for any additional pure dephasing processes. The dominant phonon contribution to the dynamics is a driving-dependent dephasing term

$$K(\Omega) = \frac{(D_e - D_h)^2 \Omega^3}{8\pi \mu c_s^5} \coth \frac{\Omega(t)}{2k_B T},$$

which we approximate by $K(\Omega) \approx K_2 \Omega^2(t)$ in the regime $k_B T > \Omega/2$. Consequently, the excitation induced dephasing is described by a characteristic timescale K_2 that is proportional to temperature, with a gradient A that depends only on bulk material parameters and is independent of the dot details:

$$K_2 = \frac{(D_e - D_h)^2}{4\pi \mu c_s^5} k_B T \equiv AT. \quad (7.48)$$

The red curves of Fig. 7.5 (c) present the best fits of this model to the data,

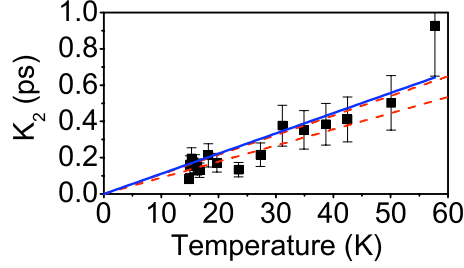


Figure 7.6: Linear dependence of K_2 on temperature. **Dashed red lines:** range of values calculated for bulk GaAs, with some uncertainty in the effective deformation coupling strength $|D_e - D_h| = 8.5 \pm 0.4$ eV [179]; further parameters: $\mu = 5.37$ g/cm³, $c_s = 5.11$ nm/ps [140]. **Blue line:** best fit of data to $K_2 = AT$. Figure taken from Ref. [175].

where the only fitting parameters are K_2 , the effective dipole, and Γ_2^* , which is comparatively small and leads to a loss of contrast that is nearly independent of the pulse area [180]. The data is in close quantitative agreement with the model, confirming that LA phonons constitute the dominant source of dephasing. Fig. 7.6 shows the K_2 parameter as a function of temperature, obtained with the same numerical fitting technique [175]. The linear growth of K_2 as a function of T is clearly visible, and we extract the gradient $A = 11 \pm 1$ fs/K from the measured data. This is consistent with first principles calculations of the host material GaAs, suggesting that considering the host bulk phonon modes is a good approximation, in agreement with Ref. [181].

In summary, the predictions of the acoustic phonon model lead to an excellent agreement with the measured excitation induced damping of Rabi oscillations, providing strong evidence for the role of acoustic phonons as the principal dephasing mechanism of s-shell excitons in self-assembled quantum dots. Moreover, the very good level of agreement between theory and experiment confirms the validity of the phonon models used in other parts of this thesis.

7.A Appendix: validity of instantaneous interaction picture operators

We here provide a justification for employing instantaneous system operators in the derivation of the ME. As a starting point for the discussion, consider Eq. (7.24) with all references to the gauge transformation removed, as this is not relevant in the following:

$$\dot{\rho} = - \int_0^\infty ds \langle B(s)B(0) \rangle (\tilde{\sigma}_z(t)\tilde{\sigma}_z(t-s)\rho - \tilde{\sigma}_z(t-s)\rho\tilde{\sigma}_z(t)) + \text{H.c.}, \quad (7.49)$$

where the Hermitian conjugate refers to the entire integrand. Transforming back to the Schrödinger picture, we obtain

$$\dot{\rho} = -i[H_S(t), \rho] - \int_0^\infty ds \langle B(s)B(0) \rangle (\sigma_z\tilde{\sigma}_z(t-s, t)\rho - \tilde{\sigma}_z(t-s, t)\rho\sigma_z) + \text{H.c.}, \quad (7.50)$$

where

$$\tilde{\sigma}_z(t-s, t) = U(t, 0)U^\dagger(t-s, 0)\sigma_z U(t-s, 0)U^\dagger(t, 0) = U^\dagger(t-s, t)\sigma_z U(t-s, t), \quad (7.51)$$

by the properties $U(t_1, t_0) = U(t_1, t')U(t', t_0)$ and $U^\dagger(t_1, t_0) = U(t_0, t_1)$ of the time evolution operator [75]. Here, $U(t, t_0)$ is only the system part of Eq. (7.5),

$$U(t, t_0) = T \exp \left(-i \int_{t_0}^t d\tau H_S(\tau) \right). \quad (7.52)$$

The Markovian approximation relies on the bath correlation functions in Eq. (7.50) decaying fast on the timescale of the system dynamics. Furthermore, this presupposes that the system Hamiltonian itself must not change appreciably during the

bath memory time, allowing us to approximate it as constant over the relevant integration interval. Therefore, we may Taylor expand the exponent of Eq. (7.52) to first order and replace the $U(t-s, t)$ in Eq. (7.51) by

$$U(t-s, t) \approx e^{iH_S(t)s}. \quad (7.53)$$

Incidentally, this step also makes the time ordering operator T redundant. Rewriting $\tilde{\sigma}_z(t-s, t)$ in the instantaneous eigenbasis with respect to $H_S(t)$ (denoted by the ‘ $\wedge$ ’ symbol),

$$\sigma_z = \cos 2\theta \hat{\sigma}_z + \sin 2\theta \hat{\sigma}_- + \sin 2\theta \hat{\sigma}_+, \quad (7.54)$$

each eigenoperator then has a simple time-dependence (where s is the time variable),

$$\tilde{\sigma}_z(t-s, t) = \cos 2\theta \hat{\sigma}_z + e^{+i\Lambda s} \sin 2\theta \hat{\sigma}_- + e^{-i\Lambda s} \sin 2\theta \hat{\sigma}_+. \quad (7.55)$$

Substituting this into Eq. (7.50) and going through the algebra yields

$$\begin{aligned} \dot{\rho} = -i[H_S(t), \rho] &+ \Gamma_{\downarrow} \sin 2\theta (\hat{\sigma}_- \rho \sigma_z + \sigma_z \rho \hat{\sigma}_+ - \sigma_z \hat{\sigma}_- \rho - \rho \hat{\sigma}_+ \sigma_z) \\ &+ \Gamma_{\uparrow} \sin 2\theta (\hat{\sigma}_+ \rho \sigma_z + \sigma_z \rho \hat{\sigma}_- - \sigma_z \hat{\sigma}_+ \rho - \rho \hat{\sigma}_- \sigma_z). \end{aligned} \quad (7.56)$$

Bearing in mind that $\mathcal{L} = P_{\Lambda}(t) = \sin 2\theta \hat{\sigma}_-$ and $\mathcal{P} = \sigma_z$, this recovers Eq. (7.31) exactly, as expected.

With this alternative derivation of Eq. (7.31), we have shown that a clear separation of timescales between bath relaxation and system dynamics – of the kind already demanded for performing the Markov approximation – justifies using instantaneous interaction picture operators in Eq. (7.25).

Chapter 8

Remote entangling of spins via an optically active mediator

The control and physical representation of entanglement lie at the heart of quantum computing. In this chapter, we shall analyse a theoretical scheme for a remote entangling operation of two spin qubits with possible applications in a range of systems, notably quantum dots, molecular systems, or defects in crystals. Our model consists of two spin qubits with no direct mutual interaction, both of which are selectively coupled to an optically active intermediary. Based on this scenario, we shall first extend the analysis of a previously studied gate and show that an entangling operation between the spins is possible even when the intermediary coupling strengths are unequal. Next, we shall develop an alternative adiabatic control procedure; we will find that this approach is similarly robust and has the additional benefit of having even less restrictive system requirements.

8.1 Introduction

To establish the proper background of the work described in this chapter, let us begin with a short review of recent progress towards the practical implementation of quantum information processing with spin qubits in the solid state. Additional to finding a well-protected and long-lived quantum two level system as the carrier of information, it is necessary to design quantum gates to precisely control both the individual dynamics of each qubit, and their correlated motion. These two classes of operation are usually treated separately, and define the single and two qubit gates we have encountered in Chapter 2. Together, these gates form a universal set that can encode any quantum algorithm.

Moreover, any practical quantum computer must meet the DiVincenzo criteria outlined in Chapter 2 and thus provide significantly more than just a universal set of gates. These requirements have led to a wealth of research on solid state implementations of quantum computers. In particular, an electron or nucleus with spin half constitutes a perfectly defined qubit. However, in solid state systems there is often uncertainty about the positions of and interactions between such spins, which makes control difficult. In this chapter we shall demonstrate that it is nonetheless perfectly possible to devise very accurate quantum gates even when the parameters describing the qubits have a degree of randomness.

Many papers have discussed how spins could be used as qubits in various materials [12, 49]. It is often the case that spins with the long coherence times that are so desirable for storing quantum information, are hard to control directly. However, they can be accessed indirectly through other states with shorter lifetimes, and manipulated more quickly. For example, different defects in semiconductors can be chosen with properties conducive to playing different roles: A defect with an electron spin in a quiet environment can be used to represent that quantum information,

and interactions can be provided by optically active defects with shorter decoherence times [50]. Alternatively, excitons in quantum dot systems can be excited to provide coupling between dot based spin qubits [132, 135, 182], that can sometimes be mediated by an optical cavity [53, 64]. In Chapter 5 of this thesis we have described an example of such an approach (without a cavity, in this instance). Further, in NMR quantum computing, the interaction between nuclear spins is provided by the electrons [3, 17]. A summary of those approaches can be found in Table 8.1.

More recently, several key experiments have been performed that demonstrate many of the ingredients that are needed for the operation of these control schemes. For example, the spin of an NV^- centre in diamond can be initialised, read and manipulated optically [183], and can be coupled to other nearby electron spins coherently, and this coherence can be manipulated optically [51, 184]. The motion of carbon-13 nuclear spins can also be detected optically in the NV^- system [185], and it has even been possible to map the NV^- electron spin state onto a nearby ^{13}C nucleus, and get it back again [186]. In semiconductor quantum dots the coherence of electron spins has been optically controlled [116] and initialised [126] and tunnel coupling between two electron spins in neighbouring quantum dots has been detected optically [131]. Remote spin coupling through a spin bath has been demonstrated in a lithographic quantum dot system [187]. Electron spin states have been used to manipulate nuclear spin qubits in other systems as well [47, 188], in a time shorter than needed for direct addressing.

Here, we consider explicitly a system for entangling two such long lived spin qubits, via a third, central electron spin of a different species that might have a much shorter decoherence time. This central spin can be optically excited, and in the excited state the electron wavefunction typically has a greater spatial extent than in the ground state, a scenario which is illustrated in Fig. 8.1. If this larger wavefunction overlaps with the two neighbouring spins, it gives rise to an exchange

Qubit	Control	Method	Reference
Electron donor spin	Electron donor spin	Optical excitation	Ref. [50]
QD electron spin	Exciton	Optical excitation	Ref. [132, 135, 182]
Nuclear donor spin	Electron spin	Electric Field	Ref. [49]
Molecular nuclear spin	Electron spin	RF pulse	Refs. [3, 17]

Table 8.1: Summary of proposals for generating pairwise entanglement of qubits with the help of controlled intermediaries. Different approaches will be optimised by choosing different material systems.

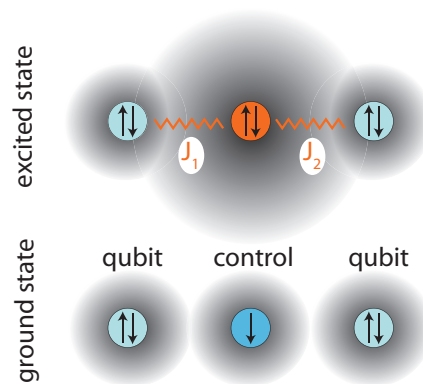


Figure 8.1: In the optical ground state of the central control qubit, the wavefunctions of the three species do not overlap and there is no spin-spin exchange interaction. When the central control is optically excited its wavefunction has a larger extent and so activates the spin-spin coupling.

coupling. This possibility was first raised by Stoneham *et al.* [50] who introduced a scheme for entangling deep donor spins in silicon, where various defects could be used for the different spin species (for example, Mg^+ , Se^+ , or Bi^+ [50, 189] are good candidates). In contrast to the previous work, we shall here study a more general situation in which there is only limited control over the various coupling parameters in the problem, and show that it is still possible to obtain a highly entangling gate operation.

8.2 Model

Let us first introduce a general model for the system we are considering. It has the following Hamiltonian, in the usual notation using Pauli pseudo-spin operators (all with eigenvalues ± 1):

$$H = E_Q \sigma_z^Q + E_C \sigma_z^C + E_{Q'} \sigma_z^{Q'} + \omega_0 |e\rangle \langle e| + |e\rangle \left(J_1 (\boldsymbol{\sigma}^Q \cdot \boldsymbol{\sigma}^C - \alpha \sigma_z^Q \cdot \sigma_z^C) + J_2 (\boldsymbol{\sigma}^{Q'} \cdot \boldsymbol{\sigma}^C - \alpha \sigma_z^{Q'} \cdot \sigma_z^C) \right) \langle e|, \quad (8.1)$$

Q and Q' label the two qubit spins; C is the central (control) spin which has two degrees of freedom: one is its spin $\boldsymbol{\sigma}^C$ and the other is its orbital state which we restrict to the space spanned by states $\{|g\rangle, |e\rangle\}$. We assume an allowed optical transition of energy ω_0 between $|g\rangle$ and $|e\rangle$, but that Q and Q' do not couple directly to an optical field. Each of the E_j gives the Zeeman splitting of spin j in an external magnetic field of strength B ($E_j = \mu_j B$). $\boldsymbol{\sigma}^Q = (\sigma_x, \sigma_y, \sigma_z)$ is the spin operator of spin Q and J_1, J_2 is the exchange coupling between spins Q or Q' and C respectively, which is only present when C is in the excited state $|e\rangle$. For $\alpha = 0$ it takes an isotropic Heisenberg form, and for $\alpha = 1$ it represents an XY type coupling. We assume that the control-qubit coupling strength when the control is in the $|g\rangle$ state is negligible in comparison with the coupling when the control is in the $|e\rangle$ state. Calculations in Ref. [190] show that this is indeed the case for donors in silicon, where the $|e\rangle$ coupling can be two to four orders of magnitude larger. We also ignore direct donor-donor coupling, which again is valid for the silicon donor system. For suitable spatial configuration with donors separated by around 25 nm, direct donor-donor coupling can be as low as 4.73×10^{-3} GHz whereas the control mediated coupling strength is still up to 157 GHz [191, 192].

In order to control the interaction, a laser is applied with frequency ω_l , and

so introduces an oscillatory term into the Hamiltonian. The oscillation can be removed by transforming into a frame rotating at ω_l and making a rotating wave approximation, whereupon we write:

$$H = E_Q \sigma_z^Q + E_C \sigma_z^C + E_{Q'} \sigma_z^{Q'} + \Delta |e\rangle \langle e| + \frac{\Omega(t)}{2} (|e\rangle \langle g| + |g\rangle \langle e|) + |e\rangle \left(J_1 (\boldsymbol{\sigma}^Q \cdot \boldsymbol{\sigma}^C - \alpha \sigma_z^Q \cdot \sigma_z^C) + J_2 (\boldsymbol{\sigma}^{Q'} \cdot \boldsymbol{\sigma}^C - \alpha \sigma_z^{Q'} \cdot \sigma_z^C) \right) \langle e|, \quad (8.2)$$

where $\Delta \equiv \omega_0 - \omega_l$ is the laser detuning from the transition and $\Omega(t)$ is the (generally time-dependent) Rabi frequency.

A general state of the three spin system $|\phi\rangle$ is given by a superposition of the spin basis states $|\{\uparrow, \downarrow\}\rangle_Q \otimes |\{\uparrow, \downarrow\}\rangle_C \otimes |\{\uparrow, \downarrow\}\rangle_{Q'}$, where the arrows represent the spin up or down projection along the z -quantisation axis. For convenience, we adopt the usual qubit notation $|Q C Q'\rangle$ with Q, C, Q' being either 0 for the ‘down’ or 1 for the ‘up’ projection of the respective spin qubit.

We shall be concerned with the situation in which we initialise the system in $|g\rangle$ and where the control spin C is prepared in the state $|0\rangle$. We then allow the system to evolve under laser excitation until there is a ‘revival’ such that C returns to the state $|0\rangle$. We will show that the remaining two qubits Q and Q' can become entangled by such an operation. In the following, we will explore two contrasting scenarios. First, a fast ‘dynamic’ optical excitation in which the system is excited suddenly by a pulsed laser, then allowed to evolve for a time before sudden de-excitation. Second, we will look at an ‘adiabatic’ approach in which the laser intensity and/or frequency is changed slowly and continuously such that the system follows its instantaneous eigenstates.

8.3 Dynamic excitation

Consider a laser that is resonant with the $|g\rangle \leftrightarrow |e\rangle$ transition (i.e. $\Delta = 0$). If a rectangular pulse is applied for a time $t_l = \pi/\Omega$ all population is transferred from $|g\rangle$ to $|e\rangle$, thus activating the spin couplings. After waiting for a specified amount of time this interaction is deactivated again using an identical pulse. For the dynamical approach to work, we require a system where $\alpha = 1$, i.e. that the Ising part is removed from the Heisenberg interaction and we are left with an XY coupling. We will discuss the reason for this at the end of this section. Hamiltonians not satisfying $\alpha = 1$ are not amenable to the dynamic method and the more general adiabatic approach discussed later must be used.

Let us assume that the optical excitation is fast in comparison to the subsequent spin dynamics, which are described by the restricted Hamiltonian $\langle e|H|e\rangle$:

$$H_e = E_Q\sigma_z^Q + E_C\sigma_z^C + E_{Q'}\sigma_z^{Q'} + \left(J_1(\sigma_x^Q \cdot \sigma_x^C + \sigma_y^Q \cdot \sigma_y^C) + J_2(\sigma_x^{Q'} \cdot \sigma_x^C + \sigma_y^{Q'} \cdot \sigma_y^C) \right). \quad (8.3)$$

H_e conserves the total spin projection: $\Sigma_z = \sigma_z^Q + \sigma_z^C + \sigma_z^{Q'}$. Therefore the evolution can be partitioned into subspaces of different Σ_z :

$$H_e = H_0 \oplus H_1 \oplus H_2 \oplus H_3 \quad (8.4)$$

where H_i is the Hamiltonian of the subspace with $\Sigma_z = 2i - 3$ (see Table 8.2). The central (control) qubit is set to $|0\rangle$ initially and so we need not consider the H_3 space further. Let us now analyse the dynamics of the other subspaces.

Subspace	Σ_z	Component States
H_3	3	$ 111\rangle$
H_2	1	$\{ 110\rangle, 101\rangle, 011\rangle\}$
H_1	-1	$\{ 100\rangle, 010\rangle, 001\rangle\}$
H_0	-3	$ 000\rangle$

Table 8.2: Table showing the four uncoupled subspaces and the notation used for each.

8.3.1 H_1 subspace ($\Sigma_z = -1$)

In the basis of states $\{|010\rangle, |100\rangle, |001\rangle\}$, and for qubits with equal g -factors ($E_Q = E_{Q'}$) we have

$$H_1 = E_C \begin{pmatrix} R & J'_1 & J'_2 \\ J'_1 & 1 & 0 \\ J'_2 & 0 & 1 \end{pmatrix}, \quad (8.5)$$

with $R \equiv (2E_Q/E_C) - 1$, $J'_1 \equiv 2J_1/E_C$ and $J'_2 \equiv 2J_2/E_C$. There is always one eigenvector that is orthogonal to $|A\rangle \equiv |010\rangle$:

$$|E\rangle = \frac{J'_1|100\rangle - J'_2|001\rangle}{\sqrt{J_1'^2 + J_2'^2}}. \quad (8.6)$$

Let us define a state that is orthogonal to both $|A\rangle$ and $|E\rangle$:

$$|T\rangle = \frac{J'_2|100\rangle + J'_1|001\rangle}{\sqrt{J_1'^2 + J_2'^2}}, \quad (8.7)$$

and rewrite the Hamiltonian in the basis $\{|A\rangle, |T\rangle, |E\rangle\}$

$$H_1 = E_C \left(\begin{array}{cc|c} R & \sqrt{J_1'^2 + J_2'^2} & 0 \\ \sqrt{J_1'^2 + J_2'^2} & 1 & 0 \\ \hline 0 & 0 & 1 \end{array} \right). \quad (8.8)$$

Prior to laser excitation, the system contains no component of $|A\rangle$; it can therefore be written as a superposition of $|E\rangle$ and $|T\rangle$. $|E\rangle$ is an eigenstate and so it only accumulates phase. $|T\rangle$ undergoes Rabi cycling to $|A\rangle$; after each cycle all population returns to $|T\rangle$, and the system ‘revives’ such that the central spin returns to state $|0\rangle$. This ‘revival time’ t_{rev} is given by:

$$t_{\text{rev}} = \frac{2n\pi}{E_C \sqrt{(R-1)^2 + 4J_1'^2 + 4J_2'^2}}, \quad (8.9)$$

where n is some integer, the number of oscillations that have occurred. At revival, we have

$$\begin{aligned} |T\rangle &\rightarrow e^{i\theta_T} |T\rangle, \\ |E\rangle &\rightarrow e^{i\theta_E} |E\rangle \end{aligned} \quad (8.10)$$

where

$$\begin{aligned} \theta_T &= \pi n - \frac{E_C(1+R)t_{\text{rev}}}{2} = \pi n \left(1 - \frac{(R+1)}{\sqrt{(R-1)^2 + 4J_1'^2 + 4J_2'^2}} \right), \\ \theta_E &= -E_C t_{\text{rev}} = -\frac{2n\pi}{\sqrt{(R-1)^2 + 4J_1'^2 + 4J_2'^2}}. \end{aligned} \quad (8.11)$$

8.3.2 H_2 subspace ($\Sigma_z = 1$)

In the basis $\{|101\rangle, |011\rangle, |110\rangle\}$ we can write

$$H_2 = E_C \begin{pmatrix} -R & J_1' & J_2' \\ J_1' & -1 & 0 \\ J_2' & 0 & -1 \end{pmatrix}. \quad (8.12)$$

After the same revival time t_{rev} , C again returns to the $|0\rangle$ state, such that the state $|A'\rangle \equiv |101\rangle$ undergoes the following transformation:

$$|A'\rangle \rightarrow e^{i\theta_{A'}} |A'\rangle, \quad (8.13)$$

where

$$\theta_{A'} = \pi n + \frac{E_C(1+R)t_{\text{rev}}}{2} = \pi n \left(1 + \frac{(R+1)}{\sqrt{(R-1)^2 + 4J_1'^2 + 4J_2'^2}} \right).$$

8.3.3 H_0 subspace ($\Sigma_z = -3$)

Finally, we have

$$|000\rangle \rightarrow e^{i\theta_Z} |000\rangle, \quad (8.14)$$

where

$$\theta_Z = -E_C(R+2)t_{\text{rev}} = -\frac{2(R+2)n\pi}{\sqrt{(R-1)^2 + 4J_1'^2 + 4J_2'^2}}. \quad (8.15)$$

8.3.4 Evolution of the logical qubits & entangling power

Combining the dynamics for the different subspaces, the overall unitary evolution of the system in the logical basis of qubits Q and Q' ($\{|00\rangle, |01\rangle, |10\rangle, |11\rangle\}$) is

$$U' = \begin{pmatrix} e^{i\theta_Z} & 0 & 0 & 0 \\ 0 & \Delta_1 & \Delta_2 & 0 \\ 0 & \Delta_2 & \Delta_3 & 0 \\ 0 & 0 & 0 & e^{i\theta_{A'}} \end{pmatrix}, \quad (8.16)$$

where

$$\begin{aligned}
\Delta_1 &= \frac{e^{i\theta_E} J_1'^2 + e^{i\theta_T} J_2'^2}{J_1'^2 + J_2'^2}, \\
\Delta_2 &= \frac{J_1' J_2' (e^{i\theta_T} - e^{i\theta_E})}{J_1'^2 + J_2'^2}, \\
\Delta_3 &= \frac{e^{i\theta_T} J_1'^2 + e^{i\theta_E} J_2'^2}{J_1'^2 + J_2'^2}.
\end{aligned} \tag{8.17}$$

To determine the extent to which this evolution creates entanglement, we use the measure of average gate entangling power developed by Zanardi *et al.* [193], who consider a bipartite pure state $|\Psi\rangle$, which lives in a Hilbert space $\mathcal{H}_1 \otimes \mathcal{H}_2$. The entangling power of U is found by taking the average of the linear entropy of the reduced density matrix, $\rho_1 = \text{tr}_2(|\Psi\rangle\langle\Psi|)$, over a uniform distribution of input product states $|\psi_1\rangle \otimes |\psi_2\rangle$:

$$e(U) = \overline{E(U|\psi_1\rangle \otimes |\psi_2\rangle)}^{\psi_1, \psi_2}, \tag{8.18}$$

where $E(|\Psi\rangle) = 1 - \text{tr}(\rho_1^2)$ is the linear entropy of ρ_1 . The theoretical maximum value of the entangling power, which is achieved by the CPHASE or the CNOT gate, is $2/9$ (i.e. about 0.22), falling to zero for a gate that produces no entanglement. Using Eq. (5) from Ref. [193] we determine that

$$\begin{aligned}
e(U) &= \frac{1}{18} (8 - 2|\Delta_1|^2 - |\Delta_1|^4 - 4|\Delta_2|^2 - 2|\Delta_2|^4 \\
&\quad - 2|\Delta_3|^2 - |\Delta_3|^4 - 2\text{Re}[e^{i(\theta_Z + \theta_{A'})} \Delta_2^2] \\
&\quad - 2\text{Re}[e^{i(\theta_Z + \theta_{A'})} \Delta_1 \Delta_3]).
\end{aligned} \tag{8.19}$$

Fig. 8.2 shows the average entangling power of the gate, $e(U)$, after the first revival $n = 1$, for $R = 1$.¹ The entangling power drops to zero when either $J_1 = 0$

¹For even n the average entangling power vanishes, $e(U) = 0$, whereas for odd n the entanglement revives to the same level as for $n = 1$.

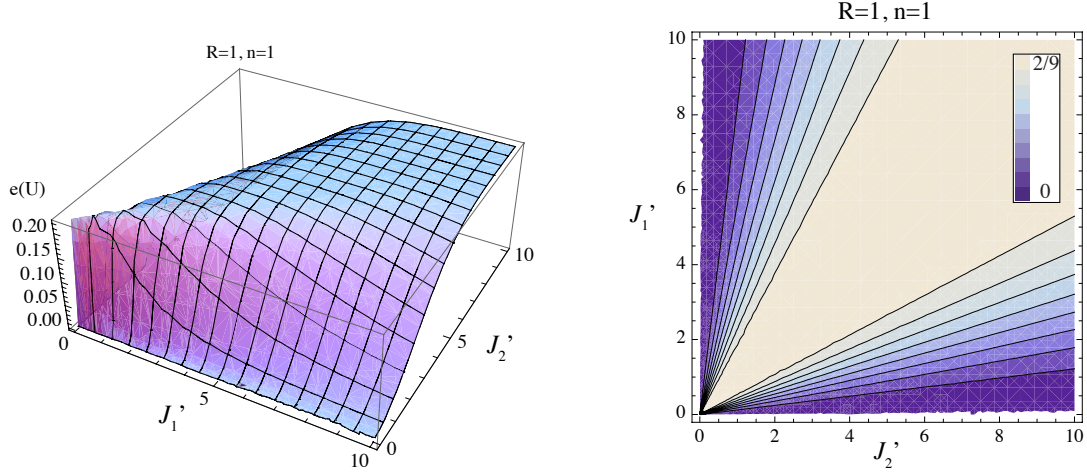


Figure 8.2: Average entangling power $e(U)$ of the dynamic two qubit gate after the first revival $n = 1$.

or $J_2 = 0$: If either of the qubits is not interacting with the central control, then no entanglement is possible. By contrast $e(U)$ is maximised (and reaches its theoretical maximum) when $J'_1 = J'_2$, but good entanglement may still be reached even when $J'_1 \neq J'_2$. Fig. 8.3 shows this equal coupling case $e(U)$ for different values of the ratio R . As R get closer to unity, the entangling power is larger for smaller $J'_1 = J'_2$. For larger $J'_1 = J'_2$ all plots approach maximal entangling power. Overall, we conclude that the dynamic gate has a reasonable entangling power over a wide range of parameter space.

We can now also see why it is essential that $\alpha = 1$ for the dynamic approach to work; if this were not the case then the revival times in the H_1 and H_2 subspaces would not coincide, invalidating the analysis presented here. In order to overcome this restriction we must change our strategy and turn to a different way of implementing the gate.

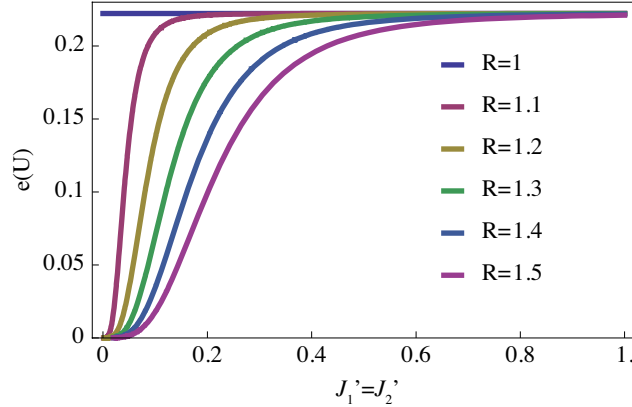


Figure 8.3: Average entangling power of the dynamic gate when $J'_1 = J'_2$, for different values of $R \equiv (2E_Q/E_C) - 1$.

8.4 Adiabatic excitation

An alternative method for creating entanglement in our system relies on adiabatic following of eigenstates. It can be implemented by slowly modulating the intensity of a laser that is close to resonance with the optical transition of the central qubit. Prior to excitation, the system is prepared in a superposition of the computational basis states, $|QQ'\rangle \in \{|00\rangle, |01\rangle, |10\rangle, |11\rangle\}$. The laser intensity is then varied such that adiabatic following of eigenstates occurs, so if the intensity is decreased again all population returns to the computational basis.

With the laser on, each of the eigenstates consists of some superposition of $|g\rangle$ and interacting $|e\rangle$ levels, such that the eigenenergies are determined not only by the optical coupling but also by the Heisenberg interaction between the spins. Fig. 8.4 shows such an eigenspectrum of Hamiltonian (8.2) as a function of Δ/Ω . For $\Delta/\Omega \rightarrow \infty$, the eigenenergies tend to the Zeeman split levels comprising the logical basis. The relative spacing between the eigenstates changes with laser intensity when Δ/Ω approaches zero. Each eigenstate therefore acquires a different dynamical phase as a consequence of its time evolution, which results in different final phases of the logical states – and thus enables the implementation of a CPHASE gate.

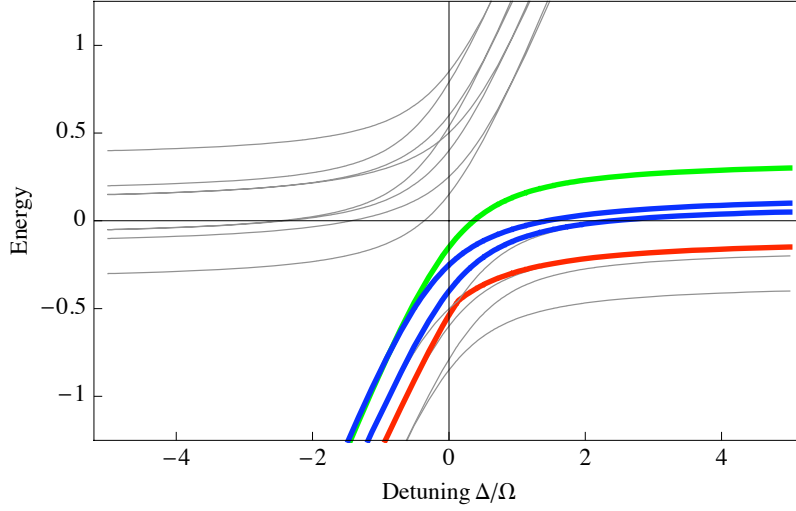


Figure 8.4: Example of eigenspectrum as a function of the detuning Δ for typical parameters. The eigenstates tending to the computational basis states for $\Omega/\Delta \rightarrow 0$ (i.e. to the far right in this figure) are colour-coded as follows: $|00\rangle$: green, $|10\rangle$ and $|01\rangle$: blue, $|11\rangle$: red.

In order to determine which pulse shape and temporal profile is suitable for achieving adiabatic following, we shall for the moment neglect the coupling between the spins in the excited levels. This gives us eight uncoupled 2LS, each of which is driven independently by the laser. This is exactly the situation for which we have already derived an ‘adiabaticity condition’ ensuring eigenstate following in Eq. (5.29) of Chapter 5:

$$\left| \frac{\dot{\Omega}(t)\Delta(t) - \Omega(t)\dot{\Delta}(t)}{2(\Delta(t)^2 + \Omega(t)^2)^{3/2}} \right| \ll 1. \quad (8.20)$$

Furthermore, for a Gaussian profile of the laser intensity,

$$\Omega(t) = \Omega_0 \exp(-t^2/\tau^2), \quad (8.21)$$

and constant detuning Δ , the inequality (8.20) can be satisfied by demanding $\Omega_0/\Delta^2 \ll \tau$. Adiabatic following is therefore always achieved in the limit $\Omega_0 \ll \Delta$

together with a sufficiently large pulse duration τ .²

The spin-spin interactions mean that the system cannot be regarded as eight separate 2LS. Rather, the eigenstates are coupled states which ultimately generate the desired entanglement. Nonetheless, the inequality (8.20) is still a requirement for achieving adiabatic following, but it is not always sufficient; it is also essential that the eigenstates belonging to the computational subspace without laser irradiation must be energetically distinguishable from those outside this subspace. This avoids population leakage from the computational basis associated with a mixing of eigenstates - and is reasonable in our scheme which presupposes two different species for Q and C .

8.4.1 Action of the adiabatic operation

As in Section 8.3, it suffices to analyse different Σ_z subspaces separately. Unlike for the dynamic excitation, however, the restriction $\alpha = 1$ is not a requirement for the adiabatic scheme - and neither is $E_Q = E_{Q'}$. Once more, the control qubit should be initialised to $\sigma_z = -1$.³ The zero excitation subspace then only contains the logical $|00\rangle$ state. Similarly, the two excitation subspace contains only the logical $|11\rangle$ state. Therefore, no population transfer between these and other logical states is possible and each merely accumulates a phase after the adiabatic pulse has been applied. The situation is different for the single excitation subspace, which is populated by the two interacting states $|01\rangle$ and $|10\rangle$. For this subspace, a more complex unitary operation between the logical states results from the adiabatic operation.

The most general action of the adiabatic operation can be therefore described by

²For small Ω_0/Δ , τ must be automatically large since the interacting levels are only weakly excited.

³It is important to have the central qubit in a well-defined initial state because of subspace dependent energy shifts of the eigenstates. In general, this leads to different pulse durations for a successful entangling operation depending on whether C is initially in the $\sigma_z = -1$ or the $\sigma_z = +1$ state.

the following unitary matrix in the basis of the logical states $\{|00\rangle, |01\rangle, |10\rangle, |11\rangle\}$:

$$U_{ad} = \begin{pmatrix} e^{i\phi_{00}} & 0 & 0 & 0 \\ 0 & \psi & \chi' & 0 \\ 0 & \chi & \psi' & 0 \\ 0 & 0 & 0 & e^{i\phi_{11}} \end{pmatrix}, \quad (8.22)$$

where ϕ_{ij} is the phase acquired by the state $|ij\rangle$. The coefficients ψ , ψ' , χ and χ' form a unitary 2×2 submatrix accounting for population transfer between $|10\rangle$ and $|01\rangle$ as well as the phase acquired by each of these two states. The complexity of the Hamiltonian (8.2) makes it difficult to find analytical expressions for the elements of Eq. (8.22). Nevertheless, for a given set of system and laser control parameters U_{ad} is straightforward to obtain numerically.

The structure of U_{ad} takes the same form as Eq. (8.16) obtained in Sec. 8.3 for the dynamic operation. This enables a direct comparison of the entangling power of the dynamic and adiabatic approach using the measure defined in Eq. (8.19).

8.4.2 Adiabatic CPHASE gate

Under certain conditions the off-diagonal terms in Eq. (8.22) vanish (recall that this is not required for a general entangling gate, but it enables an operation resembling a CPHASE gate). Using the numerical techniques discussed earlier, we find that this is the case when either of the following two sets of conditions is true:

1. the logical qubits are degenerate $E_Q = E_{Q'}$ with equal coupling $J_1 = J_2$ to the control qubit, or
2. the logical qubits are not degenerate $E_Q \neq E_{Q'}$ and the pulse duration τ is longer than $(E_Q - E_{Q'})^{-1}$.

In this case the adiabatic operation is simply

$$U_{phase} = \begin{pmatrix} e^{i\phi_{00}} & 0 & 0 & 0 \\ 0 & e^{i\phi_{01}} & 0 & 0 \\ 0 & 0 & e^{i\phi_{10}} & 0 \\ 0 & 0 & 0 & e^{i\phi_{11}} \end{pmatrix}, \quad (8.23)$$

which is locally equivalent to the CPHASE gate when [132, 145]:

$$\varphi = \phi_{00} - \phi_{01} - \phi_{10} + \phi_{11} = \pi. \quad (8.24)$$

This last condition can always be satisfied simply by choosing an appropriate pulse duration.

We now explain why a system satisfying the less restrictive second set of conditions from above gives a unitary operation of CPHASE form. Our explanation need only consider the $i = 1$ subspace since the others are in CPHASE form under any conditions. We write a general initial state in this subspace characterised by the amplitudes α and β as

$$|\psi(0)\rangle = \alpha|100\rangle + \beta|001\rangle. \quad (8.25)$$

Adiabatic following of eigenstates means that this state evolves under the influence of the laser into

$$|\psi(t)\rangle = \alpha|\mu(t)\rangle + \beta|\nu(t)\rangle, \quad (8.26)$$

where $|\mu(t)\rangle$ tends to $|100\rangle$ and $|\nu(t)\rangle$ to $|001\rangle$ as the Rabi frequency $\Omega(t)$ goes to zero. The time evolution of the slowly changing constituent eigenstates of Eq. (8.26)

follows

$$|\mu(t)\rangle = e^{iE_\mu t} |\mu\rangle, \quad (8.27)$$

$$|\nu(t)\rangle = e^{iE_\nu t} |\nu\rangle, \quad (8.28)$$

where μ and ν denote the instantaneous eigenstates and E_μ and E_ν are their associated eigenvalues. The time evolution of $|\psi(t)\rangle$ may thus be written as

$$|\psi(t)\rangle = e^{iE_\mu t} (\alpha|\mu\rangle + e^{i(E_\nu - E_\mu)t} \beta|\nu\rangle). \quad (8.29)$$

The $i = 1$ subspace consists of three spin states in each of the ground and excited optical states, so that $|\mu\rangle$ and $|\nu\rangle$ will be composed of up to six different states. Focussing on the physical states which correspond to the two logical states, $|\mu\rangle$ and $|\nu\rangle$ can be written as follows:

$$|\mu\rangle = (p|100\rangle + q|001\rangle) \otimes |g\rangle + |m\rangle, \quad (8.30)$$

$$|\nu\rangle = (r|100\rangle + s|001\rangle) \otimes |g\rangle + |n\rangle, \quad (8.31)$$

where p, r, q and s are appropriate amplitudes of this decomposition and $|m\rangle$ and $|n\rangle$ contain the contributions of the four remaining states $|010\rangle|g\rangle, |100\rangle|e\rangle, |001\rangle|e\rangle$ and $|010\rangle|e\rangle$. Inserting Eqs. (8.30, 8.31) into Eq. (8.29) yields

$$\begin{aligned} |\psi(t)\rangle &= e^{iE_\mu t} (\alpha|m\rangle + e^{i(E_\nu - E_\mu)t} \beta|n\rangle \\ &+ (\alpha p + e^{i(E_\nu - E_\mu)t} \beta r) |100\rangle \\ &+ (\alpha q + e^{i(E_\nu - E_\mu)t} \beta s) |001\rangle). \end{aligned} \quad (8.32)$$

At the end of the pulse, adiabaticity ensures that the first two terms of Eq. (8.32) disappear. The third and fourth terms show cycles of constructive and destructive

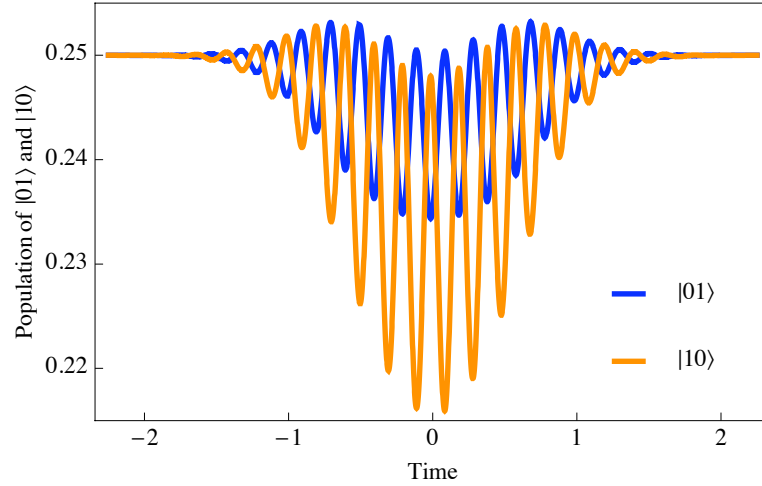


Figure 8.5: Interference of population between $|10\rangle_L$ and $|01\rangle_L$ in the $i = 1$ subspace. This effect is a consequence of the time evolution of a superposition of eigenstates as explained in the main text. As shown, a sufficiently long pulse duration damps the oscillations out and restores all population back into the original levels at the end of the pulse. The Gaussian pulse is centred around $t = 0$, where time is given in units of the pulse width τ , which in this case is set to 150 ps.

interference of the states $|100\rangle|g\rangle$ and $|001\rangle|g\rangle$. The interference oscillations range between $\alpha p \pm \beta r$ for $|100\rangle|g\rangle$ and $\alpha q \pm \beta s$ for $|001\rangle|g\rangle$ with a period of $(E_\nu - E_\mu)/2\pi = \Delta E/2\pi$. Our simulations show that if the interference period $\Delta E/2\pi$ is considerably faster than the pulse duration τ , all population is restored to the original levels at the end of the pulse, as illustrated in the scenario of Fig. 8.5 – which of course allows the realisation of a CPHASE gate with suitable laser control parameters. Conversely, comparatively fast laser pulses generally transfer population between the two logical states, leading once more to the gate described by Eq. (8.22).

8.4.3 Entangling power

We simulate the adiabatic operation by integrating Hamiltonian Eq. (8.1) with a Gaussian profile of the Rabi frequency as in Eq. (8.21). In order to prevent Landau-Zener transitions between the eigenstates, τ needs to be suitably large. For obtaining

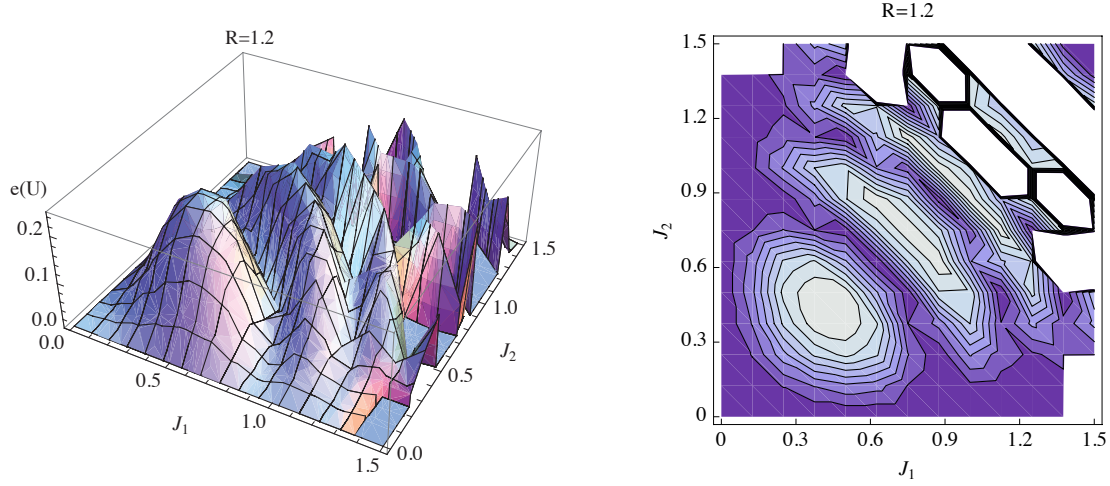


Figure 8.6: Average entangling power $e(U)$ of the adiabatic two qubit gate. The white areas of the contour plot (light blue patches without grid lines in the 3D plot) correspond to parameter combinations for which the adiabatic gate leads to population leakage out of the computational basis, making $e(U)$ ill-defined. J_1 and J_2 are given in units of 0.1 ps^{-1} . The pulse duration $\tau = 0.5 \text{ ns}$, the detuning $\Delta = 0.5 \text{ ps}^{-1}$ and coupling strength $\Omega_0 = 0.3 \text{ ps}^{-1}$. N.B. that for particular J_1, J_2 , it is possible to optimise the speed of the gate by varying τ , Δ and Ω .

the entangling power $e(U)$, we simply extract the unitary matrix Eq. (8.22) after the operation has finished.⁴ The average entangling power of both these quantities can then be determined using Eq. (8.19).

We find that a more pronounced difference between the Zeeman splittings of all three spins makes the adiabatic following more robust and permits the application of a pulse with a shorter duration. This might be achieved by using species with varying g values, or through an inhomogeneous magnetic field.

Fig. 8.6 shows a typical plot of the average adiabatic entangling power (as in Eq. (8.18)) as a function of J_1 and J_2 , and Fig. 8.7 presents a cross section along the diagonal $J_1 = J_2$.⁵ As for the dynamic gate, the most entangling region occurs

⁴If a pure phase gate is desired, one could also directly extract the non-trivial phase as in Eq. (8.24), but this case is implicitly included in the more general treatment performed here.

⁵In order to compare these simulations to those for the dynamic gate we choose more restrictive conditions than are necessary: Matching onsite energies $E_Q = E_{Q'} = 0.1 \text{ ps}^{-1}$, XY type coupling ($\alpha = 1$) and $E_C = 0.1 \text{ ps}^{-1}$.

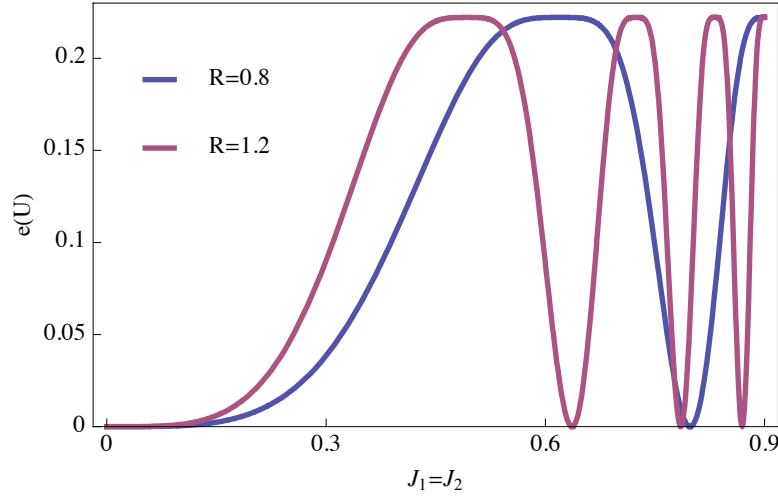


Figure 8.7: Average entangling power of the adiabatic gate when $J_1 = J_2$ (in units of 0.1 ps^{-1}), for different values of R . Since all other parameters are kept fixed rather than optimised for maximum entangling power, we obtain the clearly visible oscillatory behaviour. The first dip after the maximum then corresponds to the situation for which the entangling operation has effectively been applied ‘twice’ and consequently ‘untangled’ the state again.

along the diagonal where J_1 and J_2 are equal; in contrast to the dynamic gate, the graph shows a rather complicated oscillatory structure. This is connected to how phase accumulates in the adiabatic gate – which is in turn related to the values of J_1 and J_2 . The plots contain some areas where adiabatic following does not occur, causing significant population leakage away from the computational basis states, which establishes itself in the fact that the extracted matrix U_{ad} defined in Eq. (8.22) is not unitary.⁶ In this case $e(U)$ is ill-defined and we refrain from plotting its value. However, a well defined adiabatic operation can always be recovered by making adjustments to the choice of laser control parameters.

⁶For numerical purposes, we examine $U_{ad}^\dagger U_{ad} - \mathbf{1}_4$ and if any element of the resulting matrix is larger than 0.001, we declare the operation to be non-unitary.

8.5 Decoherence

We shall now discuss the effect of decoherence on our predictions. The dominant decoherence source or sources will be different for each physical implementation of Hamiltonian (8.1), and a full discussion of each possible process is beyond the scope of this study. However, we shall discuss decoherence that arises from spontaneous decay of the optically excited state. This could either be radiative or non-radiative [194, 195], and would be the dominant source for deep donors in silicon [50, 189].

We use a standard quantum optical master equation [80] to model the decay affecting the control qubit Q

$$\dot{\rho} = -i[H, \rho] + \Gamma_0 \left(\sigma_- \rho \sigma_+ - \frac{1}{2}(\sigma_+ \sigma_- \rho + \rho \sigma_+ \sigma_-) \right), \quad (8.33)$$

where ρ is the system's density matrix, Γ_0 is the decay rate (the inverse of the natural lifetime) and σ_+ and σ_- are raising and lowering operators with respect to $|e\rangle$ and $|g\rangle$.

We will explore two figures of merit: the amount of population returned to the desired computational basis states, and the purity of the density matrix, after the application of the gate. For both gate types we have performed a full numerical simulation that for the dynamic gate includes the two (rectangular) laser π pulses. Fig. 8.8 shows that the fast dynamic gate retains a higher purity even for fast decay rates. However, the adiabatic gate is more robust to loss of population from the computational basis.

In Fig. 8.9, we analyse the dependence of the two figures of merit on $J_1 = J_2$. For the dynamic gate, we keep $\Omega_0 = 5 \text{ ps}^{-1} \approx 3 \text{ meV}$ constant; this introduces an increasing intrinsic error as t_{rev} decreases and the transient regimes of excitation and de-excitation become more important. On the other hand, the purity improves as t_{rev}

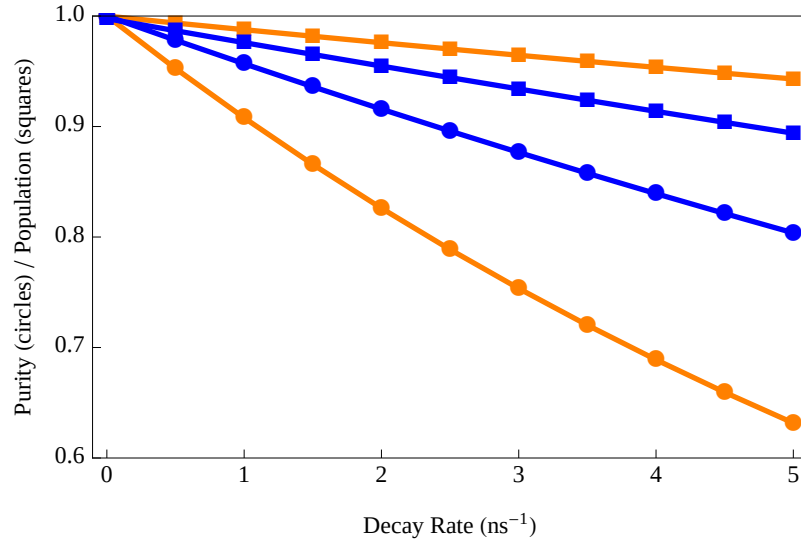


Figure 8.8: Comparison of final population in the computational basis (squares) and final purity of the system's density matrix (circles) for the adiabatic gate (orange) and the dynamic gate (blue). The decay rate is given in units of ns^{-1} and the system parameters are $J_1 = J_2 = 0.05 \text{ ps}^{-1}$ and $R = 1.2$. Typical laser control parameters have been used, such that both gates achieve an entangling power very close to the maximal value of $e(U) \approx 0.22$.

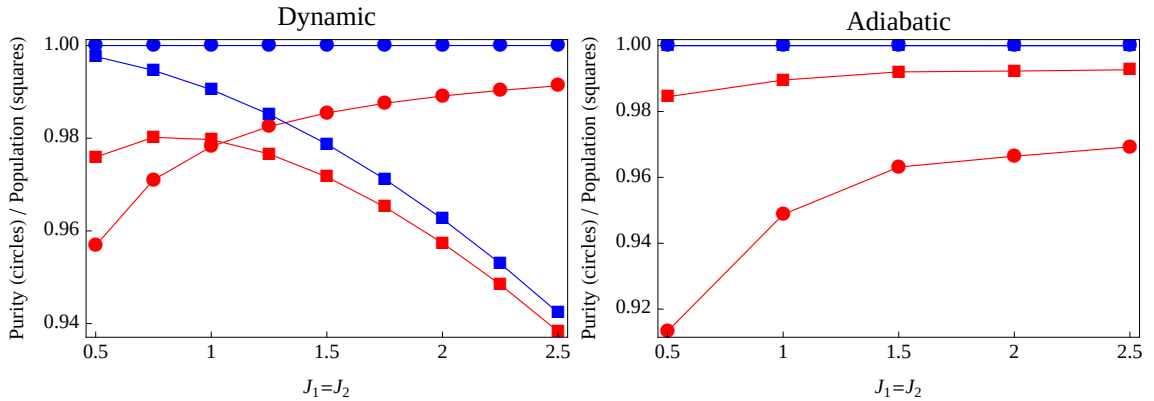


Figure 8.9: Intrinsic gate errors (blue curves) and additional effects of decoherence (red curves): the final purity and the final population of the computational basis states are shown as a function of the coupling strength $J_1 = J_2$ (in units of 0.1 ps^{-1}). A decay rate of 1 ns^{-1} , $R = 1.2$ and typical laser pulse parameters (as defined in the main text) have been used.

becomes shorter when radiative decay is included, as shown by the red curves. For the adiabatic gate, a pulse duration $\tau = 0.5 \text{ ns}$ and coupling strength $\Omega = 0.066 \text{ meV}$

are used, with the detuning adjusted in the range $\Delta = 0.16 - 0.6 \text{ meV}$ to give a maximum entangling power for each of the $J_1 = J_2$ points shown. As can be seen in Fig. 8.9, the adiabaticity condition is very well satisfied and there is no population leakage in the absence of radiative decay. However, decay events inevitably lead to some population leakage when included in the model. Unsurprisingly, a stronger XY interaction improves performance for both purity and loss of population; this is in contrast to the dynamic scheme, for which the loss of population gets worse when the interaction strengths are larger.

8.6 Conclusion and further work

In summary, we have shown that a central control can be a mediator of entanglement between two qubits, even if the coupling strengths between the mediator and the qubits are different. When a dynamic approach is taken, the coupling must be of XY form – but if an alternative adiabatic gate is performed *any* coupling form is permissible. In both cases, the gate is close to maximally entangling over a wide range of parameter space.

The proposed protocol is applicable to a range of experimental systems, including molecules with coupled electron and nuclear spins, and donors in silicon. A possible proof-of-principle experiment would endeavour to demonstrate remote entanglement between two centres that are not directly coupled by means of a simple algorithm or quantum state tomography. Quantum state tomography [3] on the qubits requires individual addressability of each spin, which translates into the requirement of different Zeeman splittings of Q and Q' . Fortunately, this is compatible with the adiabatic approach.

A natural extension to the scheme presented in this chapter consists of molecular systems with an electronic degree of freedom as the mediator and two attached

nuclear spins qubits, each coupled to the excited electronic state via a hyperfine interaction. An example of a system with a spin one electronic excitation would be a functionalised C_{60} molecule with two attached ^{13}C nuclear qubits. Encouragingly, the principle of generating entanglement through selective coupling to the mediating electronic excitation still works for this molecular structure [196, 197].

However, in this kind of system a somewhat different strategy needs to be employed for two reasons. First, the huge separation in energy between the mediator and the qubits entails different spin dynamics, but also allows certain simplifications in the analysis. Second, a spin one mediator generally necessitates a different approach compared to a spin half mediator. For the particular example of the functionalised buckyball, the normal unitary evolution of the system generates fast entanglement between the satellite qubits whenever the state of the spin one mediator has spin projection $|S_z| = 1$ rather than $S_z = 0$ [196]. Finally, we note that in this scenario the decay of the electron from the $S_z = 0$ state can result in the removal of the principal decoherence mechanism of the nuclei, leaving a pair of long-lived entangled nuclear spin qubits behind [196].

Chapter 9

Quantum coherence in the avian compass

In the previous chapters, we have been concerned with harnessing the deep quantum phenomena of superposition and entanglement mainly with the aim to build new quantum technologies. We have seen that these fundamental quantum properties are fragile, often decaying rapidly unless cryogenic temperatures are used.

Could life have evolved to exploit such phenomena [198]? Certain migratory birds have the ability to sense very subtle variations in the Earth's magnetic field [199]. In this chapter, we will consider the well-developed 'radical pair' model of the avian compass [200] together with recent experimental observations [201] in order to examine the system's vulnerability to environmental noise. Remarkably, we shall find that the room temperature noise tolerance in this natural system appears greater than that of the best man-made molecular radical, N@C₆₀ [202]. Moreover, we shall see that entanglement, though probably not an essential feature of this process, appears to persist to tens of microseconds, or more.

9.1 Introduction

The intriguing possibility that living systems already use non-trivial quantum effects to optimise some tasks has led to a number of recent suggestions, ranging from the role of quantum physics in natural selection itself [203], through to the observation that ‘warm and wet’ living systems can embody entanglement given a suitable cyclic driving [204, 205]. In particular, the physics of photosynthesis has received considerable attention; a remarkable idea here is that environmental noise is not only tolerated, but even beneficial [206, 207, 208, 209]. In this final chapter of the thesis, we shall examine a particularly important form of natural information processing known as magnetoreception. Unlike the other senses, the organ of magnetoreception is difficult to find, because organic tissue is mostly transparent to magnetic fields.

There are three main mechanisms explaining biological compasses: electromagnetic induction, ferromagnetism, and radical pairs (RP) [199]. Although the first two are essentially classical effects, there is a growing body of evidence in certain species (including birds [200], fruit flies [210] and even plants [211]) for the RP mechanism — which relies on the quantum evolution of a spatially-separated interacting pair of electron spins. Such systems have been studied in the field of spin chemistry [212, 213, 214, 215, 216] and recent experiments were able to demonstrate a chemical compass [217].

In the following sections, we will introduce the simplest form of the RP model and develop a master equation approach to describe its dynamics. By applying recent experimental data to our model, we will be able to infer a timescale for the compass mechanism and then use it to estimate a lower bound for the noise tolerance of the constituent quantum spins. Finally, we will examine the extent of entanglement and its evolution throughout the process, and comment on its likely significance in magnetoreception by the RP mechanism.

9.2 The radical pair model

There have been a series of experiments in which migratory birds are captured and exposed to artificial magnetic fields [218, 219, 220, 201]. By manipulating a bird's environment and recording its response, one can make inferences about the mechanism of the magnetic sensor. For European Robins, it is found that the birds are only sensitive to the inclination and not the polarisation of the magnetic field [218]. Secondly, if the ambient photons are of low energy (i.e. yellow, red), then the birds' preferred direction becomes random. If the ambient light is of higher energy (i.e. blue, green, white), then birds show a strong preference for a specific direction [219]. Finally, a very small oscillating field can disrupt the magnetic orientation behaviour [220, 201]. All these experiments can be explained with the RP model.

The intriguing idea behind the RP model is that birds 'see' the geomagnetic field. Oriented molecules, embedded in the bird eye's retina, form a signal pattern dependent on the inclination of Earth's magnetic field (see Fig. 9.1). The simplest RP model describes the spin of two electrons [200, 221] and one nucleus of the molecule. Absorption of a photon and subsequent transfer of one electron to an acceptor part of the molecule, gives rise to the radical pair. Due to the spatial separation it now becomes meaningful to talk about electron spin entanglement. Without the hyperfine interaction both electrons would precess around the same magnetic field, leaving the singlet state invariant. With the nuclear interaction present, the singlet state is no longer an eigenstate of this Hamiltonian, leading to a singlet-triplet oscillation dependent on the angle with the Earth's magnetic field. In other words, both electrons are subject to different local operations. Recombination occurs either from the singlet or triplet state, leading to different chemical end-products. The concentration of those products constitutes a macroscopic chemical signal, which, due to the hyperfine interaction, is sensitive to the orientation of the

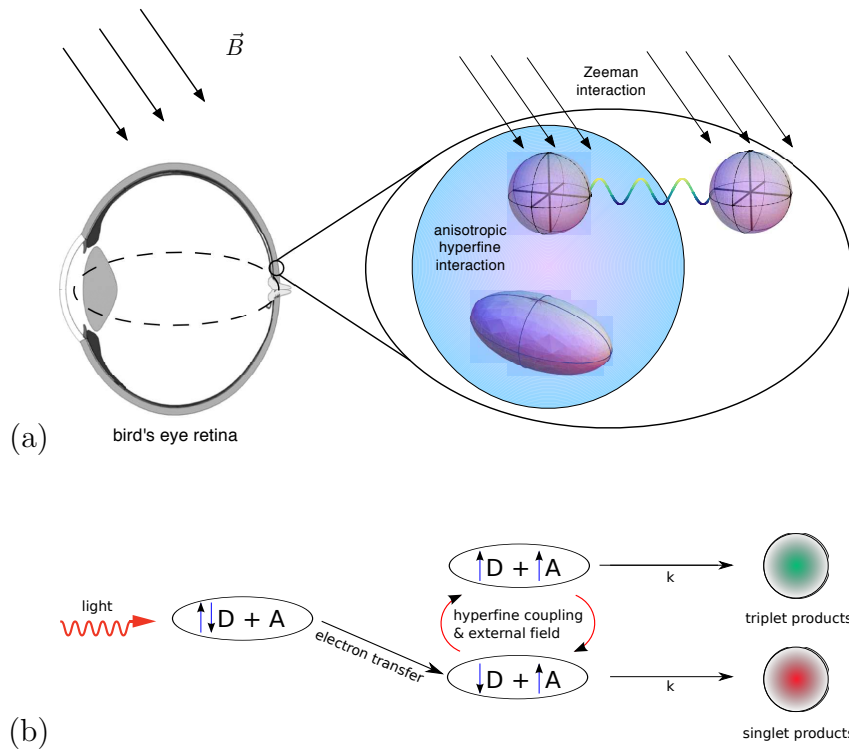


Figure 9.1: **(a) Schematic of the bird's eye.** The back of the eye contains numerous molecules, fixed with specific orientations. In the simplest RP model, each such molecule involves three crucial components (see inset): there are two electrons, and a nuclear spin that couples to *one* of the electrons. This coupling is anisotropic, so that the molecule has a directionality to it. **(b) Schematic of the radical pair mechanism.** Following photoexcitation, one of the electrons moves from the donor to the acceptor part of the RP, initially in the singlet configuration. From there, the system evolves and eventually decays with rate k to one of two possible end-products.

molecule with the magnetic field.

9.3 Hamiltonian

We employ the Hamiltonian corresponding to the system once the two electrons have become separated. The anisotropic hyperfine (HF) tensor, coupling the nucleus and

electron 1, is conveniently written in its diagonal basis

$$\mathbf{A} = \begin{pmatrix} A_x & 0 & 0 \\ 0 & A_y & 0 \\ 0 & 0 & A_z \end{pmatrix} \quad (9.1)$$

and we assume an axially symmetric (or cigar-shaped) molecule with $A_z = 10^{-5}$ meV and $A_x = A_y = A_z/2$, consistent with the general shape and magnitude of Ref. [222]. This is the simplest assumption that can provide us with directionality. It is also possible to choose a more general tensor with lower symmetry, though such a choice complicates the analysis (necessitating the use of two angles to describe the orientation). Nevertheless, we have compared the two and found that the results are qualitatively similar. The Hamiltonian is

$$H = \boldsymbol{\sigma}^n \cdot \mathbf{A} \cdot \boldsymbol{\sigma}^{e1} + \gamma \mathbf{B} \cdot (\boldsymbol{\sigma}^{e1} + \boldsymbol{\sigma}^{e2}), \quad (9.2)$$

where $\boldsymbol{\sigma}^n = (\sigma_x, \sigma_y, \sigma_z)$ is the nuclear spin operator, $\boldsymbol{\sigma}^{ei} = (\sigma_x, \sigma_y, \sigma_z)_i$ are the electron spin operators ($i = 1, 2$), $\mathbf{B}$ is the magnetic field vector and $\gamma = \mu_0 g/2$ the gyromagnetic ratio with μ_0 being Bohr's magneton and $g = 2$ the g-factor. The factor 1/2 in the gyromagnetic ratio accounts for the fact that we have a spin half system, but we will use Pauli matrices with eigenvalues ± 1 , e.g. $\sigma_z = \text{diag}\{1, -1\}$.¹ Generally, the magnetic field we employ is

$$\begin{aligned} \mathbf{B} &= B_0(\cos \varphi \sin \vartheta, \sin \varphi \sin \vartheta, \cos \vartheta) \\ &+ B_{\text{rf}} \cos \omega t (\cos \phi \sin \theta, \sin \phi \sin \theta, \cos \theta), \end{aligned} \quad (9.3)$$

¹ $\text{diag}\{a, b\}$ denotes a matrix with a and b on its diagonal and 0 elsewhere.

where $B_0 = 47 \mu\text{T}$ is the Earth's magnetic field in Frankfurt [201], and the angles describe the orientation of the magnetic field to the basis of the HF tensor. $B_{\text{rf}} = 150 \text{ nT}$ is an additional oscillatory field only applied in our simulations where explicitly mentioned. For resonant excitation with the uncoupled electron spin, $\hbar\omega = 2\gamma B_0$, so that $\nu = \omega/(2\pi) = 1.316 \text{ MHz}$.

Because of the axial symmetry of the HF tensor we can set $\varphi = 0$ and focus on the angle ϑ in the range $[0, \pi/2]$ without loss of generality. Furthermore, for the oscillatory field, we set $\phi = 0$ and introduce $\kappa = \theta - \vartheta$ as the angle enclosed by the oscillatory and static field.

We model the dynamics of the system with a quantum ME approach. We employ operators representing the reaction processes; specifically, we include two ‘shelving states’ which represent the system having decayed either from an electron singlet state, or from one of the triplet states. Ultimately one of these two forms of relaxation will occur. The three spins span an eight-dimensional Hilbert space to which we therefore add two further levels $|S\rangle$ and $|T\rangle$ for the singlet and triplet decay outcomes, respectively. The populations of these levels then correspond to the singlet and triplet yield.

With the usual definition of singlet $|s\rangle = (|0, 1\rangle - |1, 0\rangle)/\sqrt{2}$ and triplet states $|t_0\rangle = (|0, 1\rangle + |1, 0\rangle)/\sqrt{2}$, $|t_+\rangle = |1, 1\rangle$ and $|t_-\rangle = |0, 0\rangle$ in the electronic subspace (where $|0\rangle$ is spin ‘down’ and $|1\rangle$ spin ‘up’), while $|\uparrow\rangle$ and $|\downarrow\rangle$ describe the nuclear spin state, we define the following decay operators:

$$P_{S,\uparrow} = |S\rangle\langle s, \uparrow|,$$

$$P_{T_0,\uparrow} = |T\rangle\langle t_0, \uparrow|,$$

$$P_{T_+,\uparrow} = |T\rangle\langle t_+, \uparrow|,$$

$$P_{T_-,\uparrow} = |T\rangle\langle t_-, \uparrow|,$$

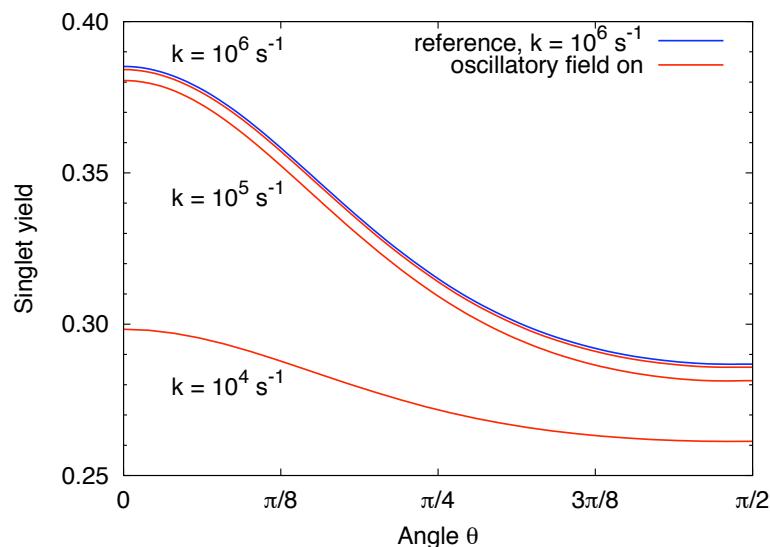


Figure 9.2: Angular dependence of the singlet yield in the presence of an oscillatory field. The blue reference curve shows the singlet yield obtained in the Earth's magnetic field with $B_0 = 47 \mu\text{T}$, which is independent of the decay rate k for $k \leq 10^7 \text{ s}^{-1}$. For better visibility, this blue curve has been shifted upwards by 0.001. The red curves show the singlet field when a 150 nT magnetic field oscillating with a frequency resonant with the Zeeman splitting of the uncoupled electron (1.316 MHz) magnetic field is superimposed perpendicular to the direction of the static field. There is no appreciable effect on the singlet yield unless k is of order 10^4 s^{-1} .

and similarly for the 'down' nuclear states. This gives a total of two singlet projectors and six triplet projectors. For simplicity, and because this choice corresponds exactly to the expression for singlet yield used in previous literature, all eight decay projectors have the same decay rate k .

9.4 Establishing a timescale

Upon photoexcitation the RP undergoes an electronic relaxation during which electron 2 moves to the acceptor part of the RP. This process is widely believed to be fast and to preserve the spin state of the two electrons. Consequently, we start our model with an initial density matrix $\rho(0)$ corresponding to the electrons in a pure

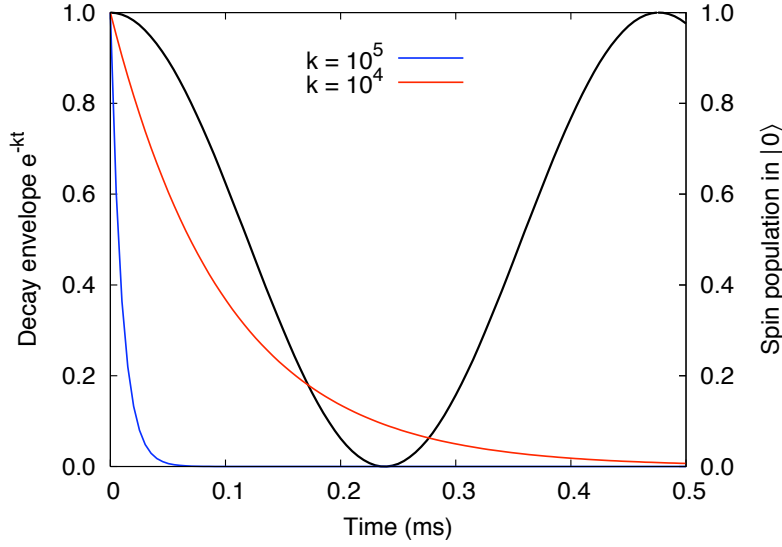


Figure 9.3: Decay envelopes for different values of k compared to the Rabi oscillations of an isolated electron spin. The black curve shows the evolution of population in the initial state $|0\rangle$ when to a resonant 150 nT oscillatory field is applied perpendicular to the static field B_0 . The effective Rabi frequency is $\Omega = \gamma B_{\text{rf}}/\hbar = 13.19$ kHz, and the Rabi period is $T = 2\pi/\Omega = 0.47$ ms.

singlet state, and a completely mixed nuclear state, i.e.,

$$\rho(0) = \frac{1}{2} \otimes |s\rangle\langle s| = \frac{1}{2} (|s, \downarrow\rangle\langle s, \downarrow| + |s, \uparrow\rangle\langle s, \uparrow|). \quad (9.4)$$

The decay from here to the two shelving levels is then described using a standard quantum ME with the above decay operators which effectively discriminate singlet and triplet decay events

$$\dot{\rho} = -\frac{i}{\hbar}[H, \rho] + k \left(\sum_{i=1}^8 P_i \rho P_i^\dagger - \frac{1}{2} (P_i^\dagger P_i \rho + \rho P_i^\dagger P_i) \right). \quad (9.5)$$

For comparison, we have also performed a more general approach with coherent decay from each basis state into its own corresponding bin level, with subsequent projection onto singlet and triplet components. That approach gives rise to identical system dynamics and singlet yield and so is unnecessarily complicated for the

problem at hand.

In the absence of noise operators (which we shall introduce below), it is possible to compare the predictions of our ME approach with the singlet yield integral that is commonly used in the prior literature. Specifically, we compare the ultimate population of our singlet shelf $|S\rangle$ with the quantity

$$\Phi = \int_0^\infty dt \, k e^{-kt} \langle \psi^- | \text{tr}_n(\rho(t)) | \psi^- \rangle. \quad (9.6)$$

We find that the two quantities agree whenever the triplet and singlet reaction times are equal (as we assume throughout this study). However, in principle the ME equation approach provides a more complete description. For instance, when k is large, i.e. the decay process occurs on a timescale that is comparable to the one of the coherent dynamics, the RWA that has implicitly been made in Eq. (9.5) is no longer justified, and including the cross-terms between different Lindblad operators would renormalise the system dynamics (see Chapter 3). Beyond that regime, for unequal singlet and triplet decay rates much faster than the singlet-triplet interconversion, it becomes necessary to consider the reaction process as a continuous quantum measurement [223].²

Next, we wish to determine an appropriate choice for our parameter k in Eq. (9.5). In Ref. [201], the authors report that a perturbing magnetic field of frequency of 1.316 MHz (i.e. the resonance frequency of the ‘remote’ electron 2) can disrupt the avian compass. They note that this immediately implies a bound on the decay rate, since the field would appear static for sufficiently rapid decay. Here we aim to refine this bound on k by considering the oscillating magnetic field *strength* which suffices to completely disorient the bird’s compass, i.e. 150 nT. (Indeed, even a 15 nT field was reported as being disruptive, but to be conservative in our conclusions we

²Resolving the precise dynamics requires a deeper understanding of the physics of the chemical reaction. Fortunately, this is not necessary in the limit of fairly small k which we are interested in.

take the larger value here.) To model this effect, we activate the oscillatory field component defined in Eq. (9.3) and examine the eventual population of shelving state $|S\rangle$, i.e. singlet yield, as a function of the angle between the Earth's field and the molecular axis. Consistent with the experimental work, we find that there is no effect at such weak fields when the oscillatory field is parallel to the Earth's field. Therefore for our analysis we set the oscillatory field to be perpendicular, i.e. $\kappa = \pi/2$. The results are shown in Fig. 9.2. We conclude that if the oscillating field is to disorient the bird, as experiments showed, then the decay rate k should be approximately 10^4 s^{-1} or less. For higher values of k (shorter timescales for the overall process) there is no time for the weak oscillatory field to significantly perturb the system; it relaxes before it has suffered any effect. This is clearly elucidated by considering the evolution of an isolated electron spin subject to the oscillatory field as a function of time compared to decay envelopes for different k as shown in Fig. 9.3. Finally, we note that such a small value for the decay rate is consistent with the long RP lifetimes in certain candidate cryptochrome molecules found in migratory birds [224].

9.5 Noise model

Settling for the value $k = 10^4 \text{ s}^{-1}$, we are able to move to the primary question of interest: how robust this mechanism is against environmental noise. The particularly long lifetime of the process gives the environment ample opportunity to decohere the system during its evolution. There are several reasons for decoherence other than the primary decay into the shelving states. For example, the dipole-dipole interaction, electron-electron distance fluctuations and other particles' spin interaction with the electrons will cause decoherence.

We shall describe environmental noise with a standard Lindblad ME technique [3,

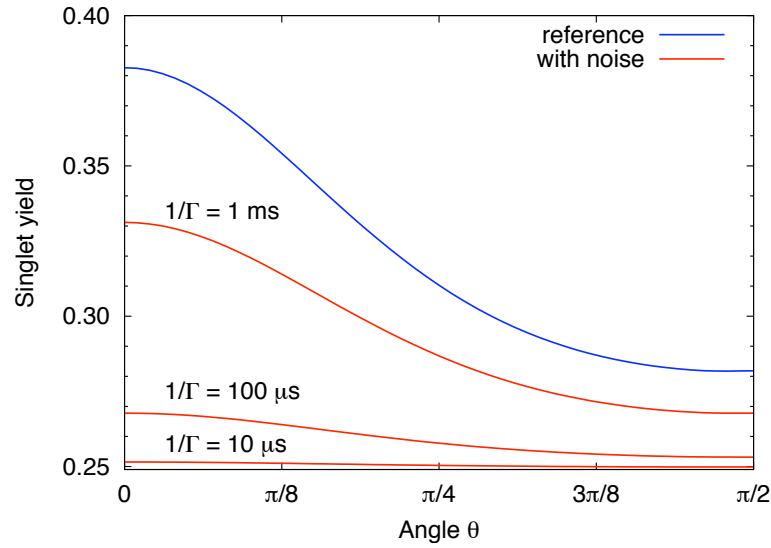


Figure 9.4: Angular dependence of the singlet yield in the presence of noise. All curves were obtained using $k = 10^4 \text{ s}^{-1}$. The blue curve provides a reference in the absence of noise and the red curves show the singlet yield for different noise rates given. It is apparent from the plot that a noise rate $\Gamma > 0.1k$ has a dramatic effect on the magnitude and contrast of the singlet yield.

76], where the following dissipator is additional to the unitary evolution and decay operators from above:

$$\dot{\rho} = \text{RHS of Eq. (9.5)} + \sum_i \Gamma_i \left(L_i \rho L_i^\dagger - \frac{1}{2} \left(L_i^\dagger L_i \rho + \rho L_i^\dagger L_i \right) \right). \quad (9.7)$$

9.5.1 Isotropic noise model

First, we look at generic environmental noise, which can flip and dephase spins with equal likelihood; considering the hot and wet environment of organic tissue, this is the kind of noise one would expect [225].

For this scenario, the noise operators L_i are σ_x , σ_y , σ_z for each electron spin individually (i.e. tensored with identity matrices for the nuclear spin and the other electron spin). This gives a total of six different and independent noise operators L_i and we use the same decoherence rate Γ for all of them. We are now in a position

to determine the approximate level of noise which the compass may tolerate, by finding the magnitude of Γ for which the angular sensitivity fails. This is shown in Fig. 9.4. Conservatively, we can say that when $\Gamma \geq k$, the angular sensitivity is highly degraded. This is remarkable, since it implies the decoherence time of the two-electron compass system is of order $100 \mu\text{s}$ or more! For comparison, the best laboratory experiment involving preservation of a molecular quantum state has accomplished a decoherence time of $80 \mu\text{s}$ at room temperature [202].

9.5.2 Pure dephasing noise

Although a physical mechanism for pure dephasing is somewhat questionable, the question of whether all types of noise are equally disruptive to the avian compass is worthwhile pursuing.

Interestingly, if we begin the simulation with a completely dephased state, $(|s\rangle\langle s| + |t_0\rangle\langle t_0|)/2$, then the classical correlations are still sufficient for achieving adequate angular visibility. Indeed, for this particular initial state we observe an angular contrast of the singlet yield that is comparable to that of the pure initial state (shown as the reference curve in Fig. 9.4). For some specific choices of hyperfine tensor the overall contrast may even be slightly larger, although the curve is typically also subject to a slight downwards shift. Thus, neither quantum phase coherence nor entanglement seems to be a prerequisite for the efficiency of the avian compass.

We now turn to the more realistic case of pure dephasing occurring *during* the unitary evolution which drives the singlet-triplet interconversion. Again, we shall use Eq. (9.7), and we need to derive suitable pure dephasing operators L_i , which we label Z to distinguish them from the generic noise operators. In the following, we discuss two different methods of implementing pure dephasing: an intuitive but

approximate method and a second, arguably more rigorous method. In both cases, we partition the entire system into two subsystems that can be treated separately: that of the HF-coupled electron and nuclear spin, and that of the remote electron 2.

Let us begin with the simpler case of the remote electron. In general, pure dephasing refers to an elastic process which does not change the system's energy. For an isolated spin with external field along the z -direction, this type of process is described by a simple σ_z -Lindblad operator [3]. However, since the basis of our system is determined by the HF tensor and the magnetic field is only parallel to the z -axis for $\theta = 0$, we need to tilt the pure dephasing operator as a function of θ :

$$Z_2 = \sigma_z \cos \theta + \sigma_x \sin \theta. \quad (9.8)$$

For the coupled electron-nuclear-spin subsystem, we write the general Hamiltonian of this subsystem's Hilbert space, $\mathcal{H}_4 = \mathcal{H}_n \otimes \mathcal{H}_e$, in matrix form in the basis $\{|0, \uparrow\rangle, |1, \uparrow\rangle, |0, \downarrow\rangle, |1, \downarrow\rangle\}$ of the diagonal HF tensor (where $|0\rangle$ and $|1\rangle$ denote the electronic and $|\downarrow\rangle$ and $|\uparrow\rangle$ the nuclear spin state), obtaining

$$H_4 = \begin{pmatrix} \mu_0 B_z + A_z & \mu_0(B_x - iB_y) & 0 & A_x - A_y \\ \mu_0(B_x + iB_y) & -(\mu_0 B_z + A_z) & A_x + A_y & 0 \\ 0 & A_x + A_y & \mu_0 B_z - A_z & \mu_0(B_x - iB_y) \\ A_x - A_y & 0 & \mu_0(B_x + iB_y) & -(\mu_0 B_z - A_z) \end{pmatrix}. \quad (9.9)$$

For our specific choice of parameters, we have $B_y = 0$, $A_x - A_y = 0$, $A_x + A_y = A_z$, while $B_z = B_0 \cos \theta$ and $B_x = B_0 \sin \theta$. By neglecting the $A_x + A_y$ terms,³ which describe flips of the nuclear spin, we realise that the electron spin experiences either an increased or a reduced effective magnetic field along the z -direction depending on the state of the nuclear spin. Hence, we proceed by defining two effective field

³This is strictly valid in the limit where $A_x + A_y \ll 2\mu_0 B_z$.

angles

$$\theta_{eff}^{\uparrow} = \arctan\left(\frac{\mu_0 B_x}{\mu_0 B_z + A_z}\right) = \arctan\left(\frac{\sin \theta}{\cos \theta + A_z/(\mu_0 B_0)}\right), \quad (9.10)$$

$$\theta_{eff}^{\downarrow} = \arctan\left(\frac{\mu_0 B_x}{\mu_0 B_z - A_z}\right) = \arctan\left(\frac{\sin \theta}{\cos \theta - A_z/(\mu_0 B_0)}\right). \quad (9.11)$$

Using these, we define the dephasing operator for this subspace as follows

$$\begin{aligned} Z_4 &= \text{diag}\{1, 0\} \otimes (\sigma_z \cos \theta_{eff}^{\uparrow} + \sigma_x \sin \theta_{eff}^{\uparrow}) \\ &+ \text{diag}\{0, 1\} \otimes (\sigma_z \cos \theta_{eff}^{\downarrow} + \sigma_x \sin \theta_{eff}^{\downarrow}). \end{aligned} \quad (9.12)$$

As we expect, this operator coincides with Eq. (9.8) tensored by the nuclear Hilbert space in the limit of no HF coupling. Of course, Eq. (9.12) is only an approximate dephasing operator owing to the fact that we have neglected the off-diagonal nuclear spin coupling. Nevertheless, it provides a useful and intuitive first attempt at describing pure dephasing in such a complex system.

Before we look at results, let us continue with introducing the second and more precise dephasing model, where we go back to the notion of fluctuating energy levels as the cause of pure dephasing. An appropriate dephasing operator would then have the eigenvalue -1 for the particular level $|\lambda_i\rangle$ undergoing fluctuations and eigenvalues $+1$ for all other levels. Let $\{|\lambda_i\rangle\}$, ($i = 1, \dots, 4$) be the normalised eigenvectors of Hamiltonian (9.9). It is easy to construct an operator with the required properties:

$$Z_{4,i} = -|\lambda_i\rangle\langle\lambda_i| + \sum_{j \neq i} |\lambda_j\rangle\langle\lambda_j| = \mathbf{1}_4 - 2|\lambda_i\rangle\langle\lambda_i|, \quad (9.13)$$

where the second equality holds since the sum over all eigenlevels must evaluate to the identity. For a general angle θ between the field and HF tensor, Hamiltonian

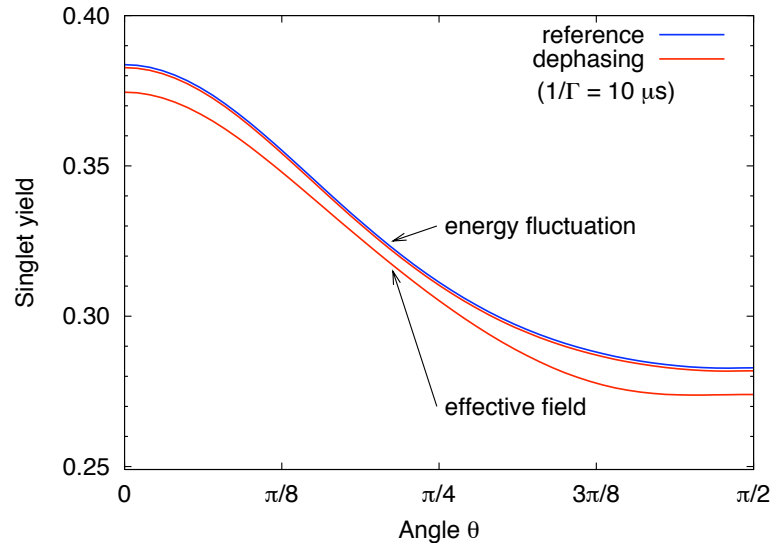


Figure 9.5: Angular dependence of the singlet yield in the presence of pure dephasing with a dephasing rate $\Gamma = 10k$ and decay rate $k = 10^4 \text{ s}^{-1}$. For better visibility, the blue reference curve has been shifted upwards by 0.001. Remarkably, even aggressive dephasing does not degrade the singlet yield contrast for either dephasing model – and it does not affect the singlet yield at all in the case of ‘energy fluctuation’ model.

(9.9) is too complicated to obtain the four resulting dephasing operators analytically, but a numerical calculation is straightforward.

The same procedure can be applied to the subspace of the remote electron spin analytically, and we simply regain the expression Eq. (9.8), once for each of the two levels.⁴ The reason for obtaining two operators is of course that we now assume that both its levels fluctuate, whereas previously only one of them did. For a fair comparison to the previous dephasing model, we multiply all of the ‘energy fluctuation’ dephasing operators by the factor $1/\sqrt{2}$, hence achieving identical dephasing of the remote spin.

Fig. 9.5 compares the results of the two dephasing models for an aggressive dephasing rate of $\Gamma = 10k$, which is a *hundred* times faster than the fastest generic noise rate that still features pronounced angular sensitivity. Strikingly, the singlet

⁴One of those operators features an irrelevant overall minus sign.

yield is essentially unaffected by this particular kind of noise. Moreover, we find that this is qualitatively true independent of the dephasing rate for both models. While the singlet yield contrast does not become degraded in the ‘effective field’ model, there is a slight dependence of the ‘downward shift’ towards the noise level of a complete mixture at 0.25, and this shift depends on Γ . On the other hand, within the ‘energy fluctuation’ dephasing model, the singlet yield coincides exactly with the ideal reference curve for any dephasing rate!

Remarkably, we have shown that the RP model of the avian compass is inherently robust against this (artificially defined) type of noise, while at the same time requiring extraordinary protection from more general noise. However, applying our noise model *and* the oscillatory 150 nT field at the same time (perpendicular to the Earth’s static field), we find that aggressive dephasing removes all sensitivity to the oscillatory field. For a dephasing rate of $\Gamma \geq 10k$, there is no longer any appreciable reduction of singlet yield contrast in the presence of the oscillatory field. Therefore, we conclude that the surprising noise protection of the RP mechanism not only prevents spin relaxation but also encompasses the preservation of phase coherence; and that the dephasing rate can be at most ten times faster than the generic noise rate that also includes spin relaxation.

9.6 The role of entanglement

It is interesting to ask, what is the significance of entanglement between the spins in the avian compass? Having inferred approximate values for the key parameters, we can plot an appropriate entanglement measure over the course of the process, from the initial singlet generation to the eventual decay. The metric we use is the negativity [226]:

$$N(\rho) = \frac{||\rho^{TA}||}{2} \quad (9.14)$$

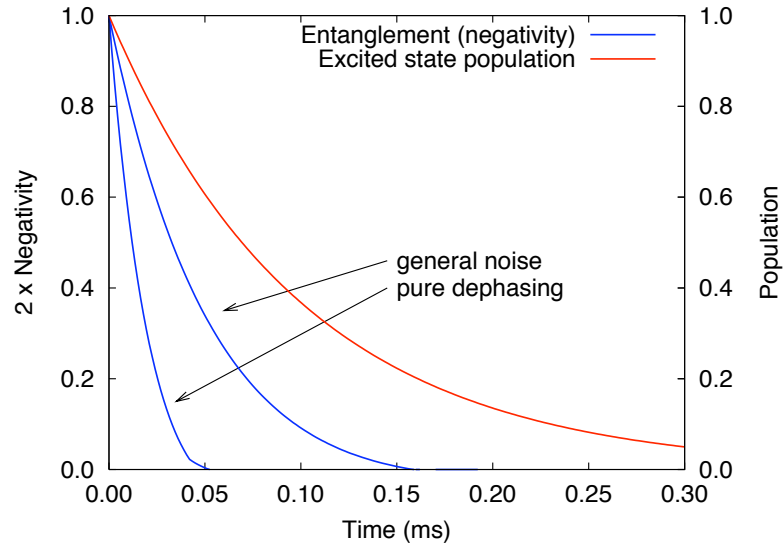


Figure 9.6: A plot indicating the decline and disappearance of entanglement in the compass system, given the parameter $k = 10^4 \text{ s}^{-1}$, and the noise severity $\Gamma = 0.1k$ for general noise and $\Gamma = k$ for pure dephasing. Here the angle between the Earth's field and the molecular axis is $\pi/4$, although the behaviour at other angles is similar.

where $||\rho^{TA}||$ is the trace norm of the partial transpose of the system's density matrix. The transpose is applied to the uncoupled electron, thus performing the natural partitioning between the electron, on one side, and the coupled electron plus its nucleus, on the other. Fig. 9.6 shows how this negativity evolves under our noise models. Clearly, the initial singlet state is maximally entangled. Under noise, entanglement falls off at a faster rate than the decay of population from the excited state. Furthermore, pure dephasing is sufficient to kill entanglement, with all entanglement having vanished after about $50 \mu\text{s}$ under the maximal tolerable dephasing rate $\Gamma = k$.

As we have seen in the previous section, the compass would be equally efficient with only classical correlations, so entanglement does not seem to play an important part in the RP process - contrary to the speculation of a related recent study by Cai *et al.* in Ref. [227]. Nonetheless, the sensitivity to the oscillatory field implies that entanglement may well be present for an extended period of time, yet it is likely a

mere by-product.

9.7 Conclusion and further work

In summary, we have used a master equation approach to examine the implications of recent experimental data on European Robins upon spin coherence in biological magnetoreception. We have been conservative in our line of inference (indeed, we could reasonably have started with an oscillatory field of 15 nT rather than 150 nT) and yet found remarkably long spin coherence times under both a general and a pure dephasing noise model. We further conclude that while entanglement does not appear necessary to the process of avian magnetoreception under the RP mechanism, it is most likely not only present, but persists out to long times.

Our observations raise a host of intriguing questions connected to the puzzle of how it is possible that organic matter seems to protect spin coherence better than N@C₆₀ in a carefully chosen solvent. Furthermore, it would be interesting to gain a better understanding of the entire RP process, beginning with the photoexcitation and subsequent relaxation to the singlet state, all the way to the physical and chemical details of the decay to the singlet and triplet end-products. While it is hard to imagine how this would change our principal conclusions, it could lead to a refinement of the model and possibly make testable predictions to help to further unravel the mystery surrounding the workings of the avian compass.

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