# Quantum spins in semiconductor nanostructures: Hyperfine interactions and optical control 

Arian Vezvaee<br>Dissertation submitted to the Faculty of the Virginia Polytechnic Institute and State University in partial fulfillment of the requirements for the degree of

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(ABSTRACT)

Quantum information technologies offer significantly more computational power for certain tasks and secure communication lines compared to the available classical machines. In recent years there have been numerous proposals for the implementation of quantum computers in several different systems that each come with their own advantages and challenges. This dissertation primarily focuses on challenges, specifically interactions with the environment, and applications of two of such systems: Semiconductor quantum dots and topological insulators. The first part of the dissertation is devoted to the study of semiconductor quantum dots as candidates for quantum information storage and sources of single-photon emission. The spin of the electron trapped in a self-assembled quantum dot can be used as a quantum bit of information for quantum technology applications. This system possesses desirable photon emission properties, including efficiency and tunability, which make it one of the most advanced single-photon emitters. This interface is also actively explored for the generation of complex entangled photonic states with applications in quantum computing, networks and sensing. First, an overview of the relevant developments in the field will be discussed and our recent contributions, including protocols for the control of the spin and a scheme for the generation of entangled photon states from coupled quantum dots, will be presented. We then look at the interaction between the electron and the surrounding nuclear spins and describe how its interplay with optical driving can lead to dynamic nuclear polarization. The second part of the dissertation follows a similar study in topological insulators: The role of time-reversal breaking magnetic impurities in topological materials and how spinful impurities enable backscattering mechanisms by lifting the topological protection of edge modes. I
will present a model that allows for an analytical study of the effects of magnetic impurities within an experimental framework. It will be discussed how the same platform also enables a novel approach for applications of spintronics and quantum information, such as studying the entanglement entropy between the impurities and chiral modes of the system.

# Quantum spins in semiconductor nanostructures: Hyperfine interactions and optical control 

Arian Vezvaee
(GENERAL AUDIENCE ABSTRACT)

Quantum information science has received special attention in recent years due to its promising advantages compared to classical machines. Building a functional quantum processor is an ongoing effort that has enjoyed enormous advancements over the past few years. Several different condensed matter platforms have been considered as potential candidates for this purpose. This dissertation addresses some of the major challenges in two of the candidate platforms: Quantum dots and topological insulators. We look at methods for achieving high-performance optical control of quantum dots. We further utilize quantum dots special ability to emit photons for specific quantum technology applications. We also address the nuclear spin problem in these systems which is the main source of destruction of quantum information and one of the main obstacles in building a quantum computer. This is followed by the study of a similar problem in topological insulators: Addressing the interaction with magnetic impurities of topological insulators. Included with each of these topics is a description of relevant experimental setups. As such, the studies presented in this dissertation pave the way for a better understanding of the two major obstacles of hyperfine interactions and the optical controllability of these platforms.

Dedication

To my family and friends

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7.1 Two QDMs in cavity connected to the same cavity mode $\omega_{c}$. A future outlook is to design a two-qubit gate between the two qubits.

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## List of Abbreviations

CPT Coherent Population Trapping

DNP Dynamic Nuclear Polarization

DRAG Derivative Removal by Adiabatic Gates

HF Hyperfine

HH Heavy Hole

LH Light Hole

LR Lindner-Rudolph

QD Quantum Dot

QDM Quantum Dot Molecule

TI Topological Insulator

TR Time-Reversal

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## Chapter 1

## Introduction and overview

Quantum information processing and the physical realization of quantum computers in solid state systems have gained considerable attention in recent years. In principle, a standard quantum computer with a large enough of number of quantum bits (qubits) is capable of tackling several tasks which are not possible using classical machines - the so-called quantum advantage [11]. Simple quantum mechanical effects such as quantum superposition, quantum measurement collapse, and quantum entanglement, enable several interesting applications which are expected to outperform classical systems for certain tasks. One famous example of such applications is the Shor algorithm for the factoring problem [12]. This dissertation is mainly devoted to improving the physical realization of systems capable of such tasks. The requirements for such physical implementations of quantum computers were set in 2000 by DiVincenzo [13]:

1. Well characterized and scalable qubits.
2. Sufficently long decoherence time.
3. Initialization capability.
4. Universal set of quantum gates.
5. Measurement capability.

A well-characterized qubit, in its ideal form, is a two-level quantum system. Several different quantum systems are considered for such realizations. A few examples include trapped ions [14], superconducting circuits [15, 16], electrons and holes in semiconductor quantum dots (QDs), nitrogen-vacancy (NV) centers in diamond [17], and non-Abelian anyon excitations in quantum topological materials. [18]. Although in this dissertation the focus is particularly on properties and challenges faced in III-V semiconductors and topological insulators, some universal concepts are shared across all platforms.

The two-level system that constitutes a qubit in Hilbert space $\mathcal{H}_{q}$ is described by the wavefunction $|\psi\rangle=\alpha|0\rangle+\beta|1\rangle$, where $|\alpha|^{2}+|\beta|^{2}=1$. In most physical platforms, the qubit can be initialized to one or two particular states. A quantum gate can be applied to change the state of the qubit. This quantum gate corresponds to a unitary evolution operator $U$ that evolves the wavefunction of the qubit $|\psi\rangle$. For instance, an $X$-rotation, refers to the implementation of the $\hat{X}$ Pauli operator on the state of qubit: $\hat{X}|\psi\rangle=\alpha|1\rangle+\beta|0\rangle$. Furthermore, multi-qubit gates can also be devised; e.g., a Controlled- $Z(\mathrm{CZ})$ gate is a two-qubit gate that applies the $\hat{Z}$ operator on the second qubit, depending on the state of the first qubit. Of course, matrix representations can be associated with gate operations. For example,:

$$
C Z=\left[\begin{array}{cccc}
1 & 0 & 0 & 0  \tag{1.1}\\
0 & 1 & 0 & 0 \\
0 & 0 & 1 & 0 \\
0 & 0 & 0 & -1
\end{array}\right]
$$

which acts on the full Hilbert space $\mathcal{H}_{q 1} \otimes \mathcal{H}_{q 2}$. The performance of these gates is quantified by the gate fidelity, which measures how close the implemented evolution operator is to the intended quantum gate. Several factors may lower the fidelity of the operations, such as interaction with the environment (e.g. a nuclear spin bath), leakage out of the Hilbert
space of the system, etc. In this dissertation, we will study the challenges associated with implementing qubit operations in condensed matter systems.

The thesis is structured as follows. In Chapter 2 a brief review of necessary concepts for understanding the basics of semiconductor physics is presented. Once the basic tools become available, we discuss the formation of self-assembled quantum dots and a specific type of vertically stacked QDs known as coupled quantum dots or quantum dot molecules (QDMs). In Chapter 3 we discuss some of the relevant recent advancements in using these systems for quantum information processing and especially their role as sources of photon emission. We discuss protocols of quantum control in these systems in Chapter 4. Chapter 5 is dedicated to studying the main source of decoherence of quantum information in these systems: Hyperfine interaction of the electron with its surrounding nuclear spin bath. We finally look at the topological band theory and formation of topological insulators in Chapter 6 and study the importance of hyperfine interactions in these systems with magnetic impurities.

## Chapter 2

## Basics of quantum spins in semiconductor systems

Semiconductor QDs are zero-dimensional nanostructures capable of confining electrons and holes with the precision of exactly a single charge. The confinement potential of these structures leads to discrete atomic-like energy levels in these systems. While several systems are available in forming these structures, such as gated dots [19], colloidal QDs [20], etc., in this thesis we solely focus on self-assembled quantum dots ${ }^{1}$, that are formed through epitaxial growth of two layers of semiconducting materials (III-V semiconductors for their optical properties) with different lattice parameters, where the lower band gap semiconductor is embedded in a higher band gap semiconductor. As a result of this band gap difference, electronic confinement due to the band offsets appears. In much of the remaining work, we consider InAs-based QDs embedded in GaAs.

### 2.1 Self-assembled quantum dots: Growth and charges

A growth mechanism of self-assembled quantum dots was developed as early as 1938 by Ivan Nikolov Stranski and Lubomir Krastanov [21]. In this method, lattice-mismatched materials

[^0]are grown layer by layer, until at some critical thickness of the QD layer, self-assembled islands appear spontaneously since the energy for island formation is lower than the strain energy to keep the QD layer. InAs and GaAs have band gap energies of 0.43 eV and 1.52 eV , respectively (at 4 K ). Once the InAs layer is grown on top of the GaAs during the growth process, due to $7.2 \%$ lattice mismatch of the two layers, a wetting layer is formed. The strain between the two layers increases and the total energy is minimized by formation of InAs islands (Fig. 2.1 (a)). An additional epitaxial layer of GaAs leads to three-dimensional quantum confinement of the QD islands. This formation process occurs spontaneously, and therefore the QD sizes and spatial positions are random. This strain-driven process leads to breaking of the cubic symmetry of the system and the appearance of a non-zero electric field gradient in the dot. This non-zero electric field may couple to the quadrupolar moment of gallium and arsenic nuclei, which has important consequences that will be discussed in detail in Chapter 5.

At this point, all the discretized valence energy states are filled and the conduction states are empty (see Fig. 2.1 (b)). As in normal semiconductors, one can excite one of the electrons from the valence band to an empty conduction band and create an electron-hole bound state. This bound state is known as exciton (Fig. 2.3). These states are denoted by $X^{ \pm}$. We will discuss the structure of hole states in detail in Section. 2.2.1.

In principle, one could use the ground state of the QD as a $|0\rangle$ state and the excited exciton as a $|1\rangle$ state. This encoding is known as the exciton qubit [22, 23, 24]. This encoding is particularly interesting since initialization of the qubit comes at a low cost: The ground state of the QD is the $|0\rangle$ state. Furthermore, single qubit operations of these qubits are demonstrated in Ref. [22], and two-qubit gates are also demonstrated in Ref. [24]. However, the gate fidelities are quite low due to fast recombination rates of the excitons [24]. Therefore, it is preferable to instead use the long dephasing times of excess electrons in QDs. In the


Figure 2.1: (a) Transmission electron microscopy of self-assembled quantum dots (taken from Ref. [1]), and (b) schematic energy level diagram of self-assembled quantum dots (taken from Ref. [2]).


Figure 2.2: (a) A neutral QD with all-filled valence states and all-empty conduction states. (b) Exciation of a single electron from the valence state to an empty conduction state by creating a hole in valence state. The electron-hole bound state is called the exciton. Notice that this is a single-particle schematic and does not reflect the many body interactions of the system.
following section, we will charge these structures with one single electron.

The remarkable control over the exact number of charged particles inside a single QD is due to the doping capability of semiconductor materials with $n$-type or $p$-type impurities which allows controlling the chemical potential of the QD through an applied electric voltage. By embedding the QD in a charge-tunable device, i.e., a heavily doped layer that performs as a reservoir, energy levels of the dot can be adjusted with respect to the Fermi level of this reservoir, and by lowering the energy level of the dot below the doped layer, a single electron can tunnel into the dot and occupy the empty conduction state. In this scheme, the two spin degrees of freedom of the electron can be encoded as qubit carriers: $|0\rangle \equiv|\uparrow\rangle$ and $|1\rangle \equiv|\downarrow\rangle$. Just as in the case of a neutral QD, valence band electrons can be excited to other unoccupied conduction states. These states are known as trion states and are composites of the excess electron and the bound state of an excited electron and a hole.


Figure 2.3: A neutral QD (left) vs a QD charged with a single electron (right). The two spin degrees of freedom of the excess electron can be used as our qubit.

### 2.2 Electronic band structure and selection rules

In this section, we look at the structure of holes of valence states and derive the optical selection rules of the system. Generally speaking, since the size of the QD is large enough such that the system contains a large amount of lattice sites $\left(\sim 10^{5}\right)$, much of our knowledge from bulk semiconductor physics applies to the QD case as well. There are several wellknown methods for deriving semiconductor band structures such as single-band effective mass, the $\vec{k} . \vec{p}$ method, and pseudopotential methods [25]. Here we will use the single-band effective mass as it also helps to understand the level structure of the QDs as well. As such, we will use the Luttinger Hamiltonian [26] to study the hole structure. It is helpful to review some of the general concepts from zincblende semiconductor physics as QDs hold the underlying symmetry of a semiconductor. The study of properties of semiconductors is greatly simplified by using symmetries of the system and group theoretical methods. The most basic symmetry of a crystal is its invariance under translations which is often accompanied by further rotational and reflection symmetries.

The eigenstates of a particle in a periodic potential $V(\vec{r}+\vec{R})=V(\vec{r})$ are given by the Bloch plane waves that are modulated by the periodicity of the crystal: $\Phi_{\vec{k}}(\vec{r})=u_{\vec{k}}(\vec{r}) e^{i \vec{k} \cdot \vec{r}}$ where
$u_{\vec{k}}\left(\vec{r}+\vec{R}_{\ell}\right)=u_{\vec{k}}(\vec{r})$, with $\vec{R}$ being the lattice vector of the crystal. $\vec{k}$ is called the wave vector and corresponds to the crystal momentum, in the units $\hbar \equiv c \equiv 1$, which we will take for the remainder of this thesis. The eigenvalues $E_{n, \vec{k}}$ of Bloch wavefunctions from the Schrödinger equation $H \Phi_{n, \vec{k}}=E_{n, \vec{k}} \Phi_{n, \vec{k}}$ are known as the dispersion relation of the system, where $n$ is an integer number, denoting the band index. Due to periodicity of the system, we may define the reciprocal lattice vector $\vec{G}$ with the relation $\vec{R} \cdot \vec{G}=2 \pi m$ with $m$ being an integer. Therefore, the periodicity also dictates that $\Phi_{n, \vec{G}+\vec{k}}=\Phi_{n, \vec{k}}$. However, we only focus on the range of $\vec{k}$ that resides within the first Brillouin zone: The set of $\vec{k}$ that are closer to the center $\vec{k}=0$ compared to all other reciprocal lattice points $\vec{G} \neq 0$.

### 2.2.1 Valence state structure: Light and Heavy holes

III-V semiconductors have zincblende structure and are direct band gap semiconductors; the top of the valence band and the bottom of the conduction band occur at the same $\vec{k}$ value. In this case, this point is at $\vec{k}=0$ which is also known as the $\Gamma$ point. Additionally, the crystal structure of zincblende materials have face-centered cubic (often abbreviated fcc) lattices. The knowledge of lattice structure of a crystal is crucial since the symmetry of the Brillouin zone results from the symmetry of the crystal. While the translational symmetry of the lattice allowed us to use the Bloch theorem, rotational and reflection symmetries of the crystal will simplify the calculations of energy band structures. This is utilized by noting that wavefunctions will transform according to the symmetry operations of the crystal. The center of the Brillouin zone ( $\Gamma$ point) possesses the highest possible symmetry and its group is isomorphic to the point group of the lattice. Therefore we can classify the wavefunctions based on the symmetry operations of the crystal. As such, we will use group theory.

From a group theoretical point of view, the space group (i.e., the group that contains both

|  | $\{E\}$ | $\left\{3 C_{2} / 3 \hat{E} C_{2}\right\}$ | $\left\{6 S_{4}\right\}$ | $\{6 \sigma / 6 \hat{E} \sigma\}$ | $\left\{8 C_{3}\right\}$ | $\{\hat{E}\}$ | $\left\{6 \hat{E} S_{4}\right\}$ | $\left\{8 \hat{E} C_{3}\right\}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
| $\Gamma_{1}$ | 1 | 1 | 1 | 1 | 1 | 1 | 1 | 1 |
| $\Gamma_{2}$ | 1 | 1 | -1 | -1 | 1 | 1 | -1 | 1 |
| $\Gamma_{3}$ | 2 | 2 | 0 | 0 | -1 | 2 | 0 | -1 |
| $\Gamma_{4}$ | 3 | -1 | -1 | 1 | 0 | 3 | -1 | 0 |
| $\Gamma_{5}$ | 3 | -1 | 1 | -1 | 0 | 3 | 1 | 0 |
| $\Gamma_{6}$ | 2 | 0 | $\sqrt{2}$ | 0 | 1 | -2 | $-\sqrt{2}$ | -1 |
| $\Gamma_{7}$ | 2 | 0 | $-\sqrt{2}$ | 0 | 1 | -2 | $\sqrt{2}$ | -1 |
| $\Gamma_{8}$ | 4 | 0 | 0 | 0 | -1 | -4 | 0 | 1 |

Table 2.1: Character table of the double group of $T_{d}$ for the $\Gamma$ point in zincblende structures. The wavefunctions at the $\Gamma$ point will transform according to the irreducible representations of this group.
translational and rotational symmetries) of zincblende is $T_{d}^{2}$. Its point group is identical to the point group of the tetrahedral point group $T_{d}$ with 24 elements: Identity, eight $C_{3}$ operations, three $C_{2}$ operations, six $S_{4}$ operations, and six $\sigma$ (reflection) operations. This is valid when we ignore the spin of charge carriers. However, upon inclusion of spin and taking the spin-orbit interaction into account, we will be dealing with the double cover group of the crystal. The double cover group of zincblende structures has $24 \times 2=48$ elements and can be shown to fall into eight conjugacy classes. As such it will have eight irreducible representations. We will use the notation $\Gamma_{i}$ with $i=1,2,3,4,5,6,7,8$ to denote these irreducible representations and the corresponding character table is shown in Table 2.1. The wavefunctions at the $\Gamma$ point always transform according to the irreducible representations of the point group of the crystal which means that the Bloch functions of the $\Gamma$ point can be labeled according to the irreducible representations of Table 2.1.

Upon inspection of the double group characters, it can be verified that the valence bands are in the $\Gamma_{8}$ representation with four states and $\Gamma_{7}$ with two states, while the lowest conduction bands are in the $\Gamma_{6}$ representation (see Fig. 2.4(a)). To see this we note that the conduction bands correspond to $s$-type orbitals, with atomic orbital angular momentum $\ell=0$, and
the corresponding valence bands are $p$-type with $\ell=1$ with three different values of $m_{\ell}=$ $0, \pm 1$. Taking the effects of spin-orbit interaction into account, the total angular momentum $J=\ell+s$ should be conserved. This does not affect the conduction band since $\ell=0$. However, this gives us the different possibilities of the valence band: We have two states with total $J=1 / 2$ with $J_{z}=\{+1 / 2,-1 / 2\}$ (two-dimensional representations, so $\Gamma_{7}$ or $\Gamma_{6}$ symmetry, but it can be shown that it belongs to $\Gamma_{7}[25]$ ), and four states with $J=3 / 2$ with $J_{z}=\{+3 / 2,+1 / 2,-1 / 2,-3 / 2\}$ (four-dimensional representations, therefore it is $\Gamma_{8}$ symmetry since this is the only four-dimensional irreducible representation). From here on, we will always take the quantization axis to be along the growth in the $z$ direction. As seen from Fig. 2.4(a), $\Gamma_{7}$ has much lower energy due to the spin-orbit effect (as such, it is referred to as the split-off band) and therefore can be neglected. We only focus on the four states from the $\Gamma_{8}$ states. The Luttinger Hamiltonian for $\Gamma_{8}$ states reads [25]:

$$
\begin{equation*}
H_{L}=\frac{1}{2 m}\left[\left(\gamma_{1}+\frac{5}{2} \gamma_{2}\right) k^{2}-2 \gamma_{3}(\vec{k} \cdot \vec{J})^{2}+2\left(\gamma_{3}-\gamma_{2}\right) \sum_{i}\left(k_{i} J_{i}\right)^{2}\right] \tag{2.1}
\end{equation*}
$$

where parameters $\gamma_{i}, i=1,2,3$ are dimensionless and are known as the Kohn-Luttinger parameters. In this form, the first two terms of the Hamiltonian above have spherical symmetry and the cubic symmetry is represented by the last two terms. Writing this Hamiltonian in a matrix form in the basis of $\left\{\left|J_{z}=3 / 2\right\rangle,\left|J_{z}=1 / 2\right\rangle,\left|J_{z}=-1 / 2\right\rangle,\left|J_{z}=-3 / 2\right\rangle\right\}$, we find,

$$
H_{L}=\frac{1}{2 m_{0}}\left(\begin{array}{cccc}
P_{1} & Q & R & 0  \tag{2.2}\\
Q^{\dagger} & P_{2} & 0 & R \\
R^{\dagger} & 0 & P_{2} & -Q \\
0 & R^{\dagger} & -Q^{\dagger} & P_{1}
\end{array}\right)
$$

where,

$$
\begin{align*}
P_{1} & =\left(\gamma_{1}-2 \gamma_{2}\right) k_{z}^{2}+\left(\gamma_{1}+\gamma_{2}\right)\left(k_{x}^{2}+k_{y}^{2}\right)  \tag{2.3a}\\
P_{2} & =\left(\gamma_{1}+2 \gamma_{2}\right) k_{z}^{2}+\left(\gamma_{1}-\gamma_{2}\right)\left(k_{x}^{2}+k_{y}^{2}\right)  \tag{2.3b}\\
Q & =-2 \sqrt{3} \gamma_{3} k_{z}\left(k_{x}-i k_{y}\right)  \tag{2.3c}\\
R & =-\sqrt{3}\left[\gamma_{2}\left(k_{x}^{2}-k_{y}^{2}\right)-2 i \gamma_{3} k_{x} k_{y}\right] \tag{2.3d}
\end{align*}
$$

We can now find the effective masses. By assuming that the $\vec{k}$ vector points along the $z$ axis,

$$
\begin{aligned}
& H_{L}^{H H}=\frac{k_{z}^{2}}{2 m}\left(\gamma_{1}-2 \gamma_{2}\right) \text { for } J_{z}= \pm 3 / 2 \\
& H_{L}^{L H}=\frac{k_{z}^{2}}{2 m}\left(\gamma_{1}+2 \gamma_{2}\right) \text { for } J_{z}= \pm 1 / 2
\end{aligned}
$$

We call these different types of valence band holes, the heavy holes (HHs), with $J_{z}= \pm 3 / 2$ and effective mass of $m_{H H}=m\left(\gamma_{1}-2 \gamma_{2}\right)^{-1}$, and light holes (LHs) with $J_{z}= \pm 1 / 2$ and effective mass of $m_{L H}=m\left(\gamma_{1}+2 \gamma_{2}\right)^{-1}$.

Our calculations thus far have assumed the translational symmetry of a lattice with a periodic potential. This symmetry in a QD is lowered by the confinement potential $V_{c}(x, y, z)$ in all three spatial directions. Since the characteristic length of this confinement potential is large compared to the atomic scale, one can take the envelope function approximation $\Psi(\vec{r})=\Phi_{\vec{k}=0}(\vec{r}) F(\vec{r})$, in which the wavefunction of each particle is the product of the Bloch function $\Phi_{\vec{k}=0}(\vec{r})$ (originating from the underlying semiconductor structure), and a slowly varying envelope function $F(\vec{r})$, originating from the of the quantum-dot Hamiltonian. This quantum confinement leads to the splitting of the hole states in QDs. Since in QDs the
confinement size is much smaller compared to the lateral size, we consider the case of strong quantum confinement $V_{c}(z)$ along the $z$ axis (growth direction), however, the conclusion does not depend on this particular choice [25, 27]. In this case, the holes are described by the Luttinger Hamiltonian $H_{L}$ from Eq. (2.2), subject to the confinement potential:

$$
\begin{equation*}
H_{h, Q D}=V_{c}(z)+H_{L} \tag{2.4}
\end{equation*}
$$

However, it is important to note that the spherical symmetry of the original problem from Eq. (2.2) is now broken by the confinement of the QD. For the systematic treatment of the problem with the envelop function approximation, one needs to replace the $\vec{k}$ of the Luttinger Hamiltonian with operators [28], however, for the simplest case of achieving a qualitative understanding of the valence states of a QD, we assume that the off-diagonal terms, i.e., $Q$ and $R$ of the Luttinger Hamiltonian are negligible. This means that the Hamiltonian above becomes:

$$
\begin{equation*}
H_{h, Q D}=V_{c}(z)+\frac{1}{2 m}\left[\left(\gamma_{1}+\frac{5}{2} \gamma_{2}\right) k^{2}-2 \gamma_{2} \sum_{i}\left(k_{i} J_{i}\right)^{2}\right] . \tag{2.5}
\end{equation*}
$$

We can now separate this problem into finding the effective masses along two separate directions: One along the confinement $(z)$ and one perpendicular to the confinement $(\perp)$. We find the effective masses $m_{H H}^{z}=m\left(\gamma_{1}-2 \gamma_{2}\right)^{-1}$ and $m_{L H}^{z}=m\left(\gamma_{1}+2 \gamma_{2}\right)^{-1}$ along the confinement, and $m_{H H}^{\perp}=m\left(\gamma_{1}+\gamma_{2}\right)^{-1}$ and $m_{L H}^{\perp}=m\left(\gamma_{1}-\gamma_{2}\right)^{-1}$ for the in-plane motion. We can also find the separation along the confinement between the HHs and LHs to be $\Delta_{H H, L H}=2 \gamma_{1} k_{z}^{2} / m[25,27]$. By including the off-diagonal terms into the Luttinger Hamiltonian, we find the effects of hole mixing; the $Q$ term couples $| \pm 3 / 2\rangle$ states to $| \pm 1 / 2\rangle$ states, and the $R$ term couples $| \pm 3 / 2\rangle$ to $|\mp 1 / 2\rangle$ states. This essentially means that hole
(a)

(b)


Figure 2.4: (a) Band structure of bulk III-V semiconductors. (b) Effective masses of hole states in a QD subject to strong confinement along the $z$ (growth) axis.
spins are spinors that contains some contribution from all four of hole projections. Although, it is often the case that one HH component is the dominant term so we denote them by the notations $|\Uparrow\rangle$ and $|\Downarrow\rangle$. The overall compositions are

$$
\begin{align*}
|\Uparrow\rangle & =|+3 / 2\rangle+c_{L H}|+1 / 2\rangle+c_{L H}^{\prime}|-1 / 2\rangle+\bar{c}|-3 / 2\rangle,  \tag{2.6}\\
|\Downarrow\rangle & =|-3 / 2\rangle+c_{L H}|-1 / 2\rangle+c_{L H}^{\prime}|+1 / 2\rangle+\bar{c}|+3 / 2\rangle, \tag{2.7}
\end{align*}
$$

where $\bar{c} \sim c_{L H} \times c_{L H}^{\prime}$, and coefficients $c_{L H}$ and $c_{L H}$ are very small numbers (less that $10 \%$ ) for InAs as are experimentally measured from the absorption spectroscopy [29, 30].

### 2.2.2 Optical selection rules

Using our knowledge about the structure of the holes we can now find the optical selection rules of exciting QDs to higher energy states. The exciton and trion states that we discussed


Figure 2.5: Optical selection rules in QDs. In (a) a polarized light excites a single excess electron into a new configuration with higher energy. The colors indicate how these different states are denoted in the selection rules shown in (b) for the case of a spin up and $\sigma^{+}$light. A similar process takes place for a spin down and $\sigma^{-}$light.
in the previous section are achieved by shining a polarized light at the frequency of the transition frequency of the QD. Since left-, and right-circularly polarized light carries angular momentum $\ell= \pm 1$ (denoted by $\sigma^{+}$and $\sigma^{-}$respectively), the conservation of angular momentum from the ground states to exciton and trion states will dictate what configuration of charges are possible.

As we discussed in the previous section, LH bands lie deeper compared to HH bands (Fig. 2.4(b)). Therefore we only consider configurations with HHs, i.e., we assume the hole states carry angular momentum of $J_{z}= \pm 3 / 2$. As such, by shining $\sigma^{+}\left(\sigma^{-}\right)$on a neutral QD with no initial angular momentum, only the bright excitons $X^{+}=|\downarrow \uparrow\rangle\left(X^{-}=|\uparrow \downarrow\rangle\right)$ are allowed. The two states with parallel spins, $|\uparrow \Uparrow\rangle$ and $|\downarrow \downarrow\rangle$ (with total $J_{z}$ being +2 and -2, respectively), are known as dark excitons and are mainly optically inactive. They can be either accessed through hole spin mixing (discussed below), or through radiative decay of biexcitons; a bound state of two excitons [31].

The same principle applies to charged QDs. If the QD is charged with a spin up $|\uparrow\rangle($ down $|\downarrow\rangle)$ electron then using $\sigma^{+}\left(\sigma^{-}\right)$light we will have access to the bright trion states $|T+\rangle=|\uparrow \downarrow \uparrow\rangle$
$(|T-\rangle=|\downarrow \uparrow \downarrow\rangle)$. This process is depicted in Fig. 2.5. In Fig. 2.5(a) the process of shining light on different initial configurations of a charged QD is shown. The secondary configurations in (a) are the excited state charge configurations. We show these processes in a QD in a short-hand notation used in (b), in which the corresponding configurations from (a) are color-coded for the case of $\sigma^{+}$. In terms of the hole mixing description of the previous section, we can find the dipole matrix elements between the electron spin states and the trion states as

$$
\begin{align*}
\langle T \pm| \mathbf{d} \cdot \boldsymbol{\sigma}^{ \pm}| \pm 1 / 2\rangle & =1  \tag{2.8a}\\
\langle T \pm| \mathbf{d} \cdot \boldsymbol{\sigma}^{\mp}| \pm 1 / 2\rangle & \sim c_{L H}  \tag{2.8b}\\
\langle T \mp| \mathbf{d} \cdot \boldsymbol{\sigma}^{\mp}| \pm 1 / 2\rangle & \sim c_{L H}^{\prime}  \tag{2.8c}\\
\langle T \mp| \mathbf{d} \cdot \boldsymbol{\sigma}^{ \pm}| \pm 1 / 2\rangle & \sim \bar{c} \tag{2.8d}
\end{align*}
$$

These selection rules have direct consequences on applications of QDs for quantum information processing. Applying certain quantum gates corresponds to transferring populations from one state of the qubit to another; that is, an $X$ gate corresponds to flipping the spin of the QD. It is clear from the selection rules in the equations above that the matrix elements from the opposite trions to the spin manifold are not strong enough due to the smallness of the coefficients $c_{L H}, c_{L H}$, and $\bar{c}$. Therefore the dominant selection rules are those shown in Fig. 2.5(b). The required selection rules for implementation of quantum operations could be achieved by application of an in-plane external magnetic fields perpendicular to the growth axis (z), the so-called Voigt geometry. Let us pick the magnetic field to be along the $x$ axis. In that case the spin states will have to be the eigenstates of the $X$ Pauli operator. As such the two electron ground states will be


Figure 2.6: Optical selection rules in QDs with an in-plane external magnetic field, known as the Voigt geometry. The lights are linearly polarized and using circularly polarized light will lead to formation of $\Lambda$-systems.
$|x\rangle=\sqrt{1 / 2}(|\uparrow\rangle+|\downarrow\rangle)$ and $|\bar{x}\rangle=\sqrt{1 / 2}(|\uparrow\rangle-|\downarrow\rangle)$. Correspondingly, the trion levels will be $\left|T_{x}\right\rangle=\sqrt{1 / 2}(|\downarrow \uparrow\rangle-|\uparrow \downarrow\rangle)(|\Downarrow\rangle+|\Uparrow\rangle)$ and $\left|T_{\bar{x}}\right\rangle=\sqrt{1 / 2}(|\downarrow \uparrow\rangle-|\uparrow \downarrow\rangle)(|\downarrow\rangle-|\uparrow\rangle)$. Now, upon using linearly polarized light, that is $\pi_{x}=\sqrt{1 / 2}\left(\sigma^{+}+\sigma^{-}\right)$and $\pi_{y}=\sqrt{1 / 2}\left(\sigma^{+}-\sigma^{-}\right)$, we will have the transitions shown in Fig. 2.6 [32]. This means that if we pick one polarization of the light, we will end up with a $\Lambda$-system. This $\Lambda$-system, upon using let us say, $\sigma^{+}$, will be the three-level system of $\left\{\left|T_{\bar{x}}\right\rangle,|x\rangle,|\bar{x}\rangle\right\}$. Thus, we can imagine that by transferring the population through the auxiliary trion level in this $\Lambda$-system we can implement desired quantum gates. Some potential schemes will be discussed in Chapter 4.

### 2.3 Quantum dot molecules

As we discussed in Section 2.1, the spatial position of QDs during epitaxial growth is random. Moreover, each dot will have a slightly different band gap energy which means exciting more than one dot at a time requires specific lasers focused on a particular dot. This clearly makes the scalability argument of the DiVincenzo criterion impossible. In Ref. [33], Economou, Doty et al. have proposed using a coupled pair of vertically stacked InAs QDs, known as QDMs [34], to overcome this challenge. In the following, we look at the basic concept of


Figure 2.7: Six vertically stacked quantum dots. Taken from Ref. [3]
growth in these systems and their optical selection rules.

### 2.3.1 Growth procedure

QDMs are two vertically stacked pairs of InAs QDs, grown on top of each other. As discussed in Section 2.1, the appearance of islands of QDs is random, therefore by simply growing two layers of QDs, there is no guarantee that the dots will appear on top of each other. For this purpose, the cap and flush method [3] is used; first, at low temperatures, InAs QDs are grown, and then a partial cap of GaAs is grown. By raising the temperature the top of the QD is desorbed and the dot is capped. This is followed by completing the growth of the GaAs layer. In order to control the coupling of multiple layers of QDs, the strain of the first layer, propagates through the capping layer to the surface as a result of the deformation of the GaAs from the larger lattice constant InAs of the QD. The increase in lattice constant at the surface creates the required nucleation site for a secondary QD in any following growth. This process can be repeated multiple times without loss of vertical alignment with QDs. Fig. 2.7, shows this process for six quantum dots grown on top of one another.

### 2.3.2 Molecular states in QDMs

Single-particle bound states with electrons or holes can be isolated in QDMs with several charge configurations. We label these states by QD occupancy with respect to the top and bottom dots: $\binom{e_{B} e_{T}}{h_{B} h_{T}}$. The occupancies are determined by the Pauli exclusion principle such that each orbital can be populated by a maximum of two charges. Excitonic bound states can occur with charges within the same dot as direct excitons, or two opposite dots as indirect excitons. We denote these two states as $\binom{0, \uparrow}{0, \Downarrow}$ being an example of a direct exciton and $\binom{\uparrow, 0}{0, \Downarrow}$ being an example of an indirect exciton.

Electrons and holes in QDMs are coupled through the pairwise exchange interaction. Additionally, each pair of particles is coupled through the direct Coulomb interaction as well. The multi-particle Hamiltonian of a QDM, by taking into account the single particle Hamiltonians and the interaction among the charges in different dots, can be written as

$$
\begin{equation*}
H=H_{\mathrm{e}}+H_{\mathrm{h}}+H_{\mathrm{Coulomb}}+H_{\text {exchange }} \tag{2.9}
\end{equation*}
$$

The single-particle terms of the Hamiltonian read,

$$
\begin{equation*}
H_{\mathrm{e}}+H_{\mathrm{h}}=\sum_{\alpha} E_{i}^{\alpha} n_{\alpha i}-\sum_{\alpha c} t_{\alpha}\left(c_{\alpha B \sigma}^{\dagger} c_{\alpha T \sigma}+c_{\alpha B \sigma}^{\dagger} c_{\alpha T \sigma}\right) \tag{2.10}
\end{equation*}
$$

with $\alpha=(e, h)$, in each dot, $i=(B, T)$ and $c_{\alpha i \sigma}\left(c_{\alpha i \sigma}^{\dagger}\right)$ being the annihilation (creation) operators for localized single-particle states. The wavefunction overlap between QDs leads to intrinsic interdot tunnel coupling $-t_{\alpha}=\langle B, \sigma| H_{\alpha}|T, \sigma\rangle$. Hybridized wavefunctions as symmetric and antisymmetric superpositions $\left(|B, \sigma\rangle_{\alpha} \pm|T, \sigma\rangle_{\alpha}\right) / \sqrt{2}$, are formed by bringing the tunnel coupled energy levels into resonance. In analogy with diatomic molecules, an anticrossing pattern with the energy level splitting of $\Delta E_{\alpha}=\left|2 t_{\alpha}\right|$ occurs; hence the name

QDMs. We model this anticrossing of the system as follows. We first need to understand the tunneling resonances of QDMs in a external electric field applied in the growth direction. Application of such electric fields allow to control the energy levels of QDs via quantumconfined Stark effect [34]. For an electric field $F$, the resulting Stark shift is $\Delta E_{i}^{\alpha}(F)=$ $-p_{i \alpha} F$, where $p_{i \alpha}=q_{\alpha}\left\langle z_{i}\right\rangle$ is the static dipole moment, $\alpha=\{e, h\}$ with $q_{e, h}=\mp e$, and $i=(B, T)$. Notice that due to the presence of a barrier between the two dots (with thickness $d)$, energy levels in separate QDs are shifted the most by the built-in interdot dipole $p_{0}=e d$. As such indirect excitons have a strong energy dependence on applied electric fields since they have electron and hole that are located in two separate QDs, while direct excitons have a small electron-hole dipole moment since charges are located in the same dot, therefore they have a weak energy dependence on applied external electric fields. The neutral exciton Hamiltonian in the basis of direct and indirect excitons is

$$
\widehat{H}^{X}=\left(\begin{array}{cc}
E_{0} & -t_{X}  \tag{2.11}\\
-t_{X} & E_{0}-e d F
\end{array}\right)
$$

with $E_{0}$ being the exciton energy and $t_{X}$ the tunneling matrix element, determined by the overlap of the hole wavefunctions in the two dots. It is important to note that the states of QDMs continuously transform between atomic-like and molecular orbitals as a function of the electric field $F$ : At zero electric field the eigenvalues are $E_{0}-t_{X}$ and $E_{0}+t_{X}$ and the corresponding eigenstates are the symmetric (bonding) and antisymmetric (antibonding) combination of the direct and indirect excitons. On the contrary, for large electric fields, the tunnel coupling is much smaller than the Stark shift ( $e d F$ ) and the eigenvalues of the Hamiltonian are the energies of the two basis states, direct and indirect exciton. The anticrossing of these states is demonstrated in Fig. 2.8.


Figure 2.8: Energies of observed photoluminescence lines for the neutral exciton show an anticrossing as a function of applied electric field. Taken from Ref. [4].

### 2.3.3 Hole spin mixing in quantum dot molecules and scalable structures

We now turn to the experimental observation of hole spin mixing in QDMs as reported in Ref. [5]. As we discussed in Section 2.2.2, hole spin mixing between the HHs and LHs is expected at transverse magnetic fields which results in the appearance of dark excitons in optically measured spectra. However, in QDMs this phenomenon occurs in the absence of transverse magnetic fields. In Fig. 2.9(b) both bright and dark (circled in green) excitons are observed with no applied transverse magnetic fields. The appearance of the dark state signals the coherent coupling of the direct dark and the indirect bright states which is equivalent to having a superposition of the heavy and light hole spins. This hole spin mixing is due to the spin-orbit interactions of the valence band that we discussed, and the symmetry breaking of QDMs due to the misalignment of the two dots. As pointed out in Section 2.2.2, hole states contain contributions of varying degree from all four heavy and light holes. In the case of a QDM, the coupling of the LH between two QDs is very strong due to spin-orbit interactions


Figure 2.9: (a) Theoretical model of bright and dark exciton energies of QDMs as a function of external electric field. (b) The corresponding experimental observation in the absence of in-plane magnetic fields. Taken from Ref. [5].
and misalignment of the two dots.

This spin hole mixing has been at the heart of the proposal by Economou et al. in Ref. [33] as it enables arbitrary rotations between the qubit states defined by the two HHs $\binom{0,0}{0, \Downarrow}$ and $\binom{0,0}{0, \Uparrow}$ ) without the need for external magnetic fields. The spin mixing along with the indirect optical transitions, provide a pathway to scalable QD-based systems: Although each QD in the QDM has its own unique direct transition, the indirect transition can be tuned into resonance with a cavity using local electric fields. In that sense, multiple QDMs can be tuned with the same frequency of a single laser rather than having an individual laser tuned to each QDM. The level structure and the selection rules of these hole spin qubit systems, all based on indirect transitions of QDMs, are shown in Fig. 2.10(b).

The hole spin mixing enables an optical transition between the basis states and a superpos-


Figure 2.10: (a) Schematic depiction of two coupled QDs grown vertically on top of one another. (b) Optical selection rules from hole spin qubits in these systems.
ition state that contains both $\binom{0, \uparrow}{0, \uparrow \downarrow}$ and $\binom{0, \uparrow}{\Downarrow, \Downarrow}$ components. This $\Lambda$-system can be used to perform rotations on the hole spin basis. Two transitions to $\binom{0, \downarrow}{\uparrow, \Downarrow}$ and $\binom{0, \uparrow}{\downarrow, \uparrow}$ can be used for selective readout since the optically excited states couple optically to only one of the qubit states. We call these the cycling transitions since photon emission does not lead to relaxation to the other hole basis state. However, the hole spin mixing also proves to be problematic as two molecular states with close energy values are formed: The target $|t\rangle$ and the unwanted level $|u\rangle$. These states have opposite molecular symmetries but both have the same optical coupling to the qubit basis states. This will lead to off-resonant coupling of the excitations and leakage out of the intended Hilbert space which is detrimental to the fidelity of the quantum gates. We will develop quantum control techniques to improve the fidelities of these gates in Chapter 4.

## Chapter 3

## Quantum information processing with quantum dots

In this chapter, we first review the applications of self-assembled quantum dots for quantum information processing, and in particular, their role as sources of single photon emission. We review the emission properties of QDs and why they are ideal sources of single-photon emission and we go over some of their applications for the generation of photonic graph states. In Section 3.3.4, we end the chapter by presenting our results in the form of proposals for the generation of photonic graph states from QDMs.

### 3.1 Quantum dots as sources of single-photon emission

An ideal single-photon source is defined as a source of photon emission that in response to an external trigger emits a single-photon. This can be modeled as a two-level atomic-like system with a ground state $|g\rangle$ and an excited state $|e\rangle$, shown in Fig. 3.1. By initially putting the population in the ground state level, an optical or electrical excitation transfers the population to the excited level. Through the decay of the system from the unstable excited level a photon is spontaneously emitted. This process can be repeated for a string of photons. In a sense, this process can be thought of as if the two-level system is used to convert the coherent state of the light from the trigger pulse into a single-photon stream.

Step 1: Excitation


Figure 3.1: An ideal source of single photons. A two-level system that can be excited through an external trigger. Upon relaxation, a photon is emitted spontaneously.

The light trigger pulse used in this process is a combination of plane wave modes where each mode is a quantized harmonic oscillator with polarization $\varepsilon$ and frequency $\omega=|\mathbf{k}|$ $(\hbar \equiv c \equiv 1)$. Defining $n$ as the occupation number of a mode, we have $\sum_{\mathbf{k}, \varepsilon, n} c_{\mathbf{k}, \varepsilon}|n\rangle_{\mathbf{k}, \varepsilon}$. The outcoming photon from the system, on the other hand, is a light field composed of single-photon Fock states $|1\rangle_{\mathrm{k}, \varepsilon}$, with $n=1$.

Some important features of a good source of single-photon emission are:

- Indistinguishability: Two simultaneous incident photons on a two-input beam splitter interfere such that both exit from a single output due to their Bose-Einstein statistics [35]. Two purely indistinguishable photons will have a completely destructive interference. An ideal source of single-photon emission will emit photons that are completely indistinguishable. However, the transition frequency of the system (and consequently the photons) can be affected by the dephasing and spectral diffusions of the system [36]. The spectral fluctuations of the optical frequency of the emitter will lead to distinguishability of successively emitted photons from the system.
- Wavelength: An ideal single-photon source should be a narrow linewidth emitter, meaning that once the two-level transition frequency is driven on-resonance, it leads


Figure 3.2: Observed spectrum of resonant excitations of InAs/GaAs QD and the energy of the emitted photon (taken from Ref. [2]).
to emission of a photon with a well-defined frequency.

- Emission rate: The emission rate of photons is determined by the inverse of their spontaneous emission lifetime which depends on the refractive index of the medium, transition frequency, and the transition dipole moment of the ground and excited states.

All these features are consistent with the optical properties of self-assembled quantum dots. They have a high emission rate (order of ns [36]), and a very narrow bandwidth of emitted photons (see Fig. 3.2). The most advantageous aspect of these systems is the ease of integrability with optical structures that can be grown around the same materials.

The emitted photons from QDs can be utilized as flying qubits over long distances due to their lack of interaction with the environment. There are various ways to use different degrees of freedom of photons for encoding information. The two methods that we will discuss in the following sections are polarization (left- or right-handed), and time-bin (presence or absence) encoding.

### 3.2 Measurement-based quantum computation and quantum networks

Due to the remarkable photon emission properties of QDs, they are one of the mostinvestigated systems for photonic technologies [1, 36]. In this section, we look at some of these technologies, namely the graph states and their generation protocols from QD-based systems. Graph states can be used for implementation of both measurement-based quantum computation (MBQC) [37] and also quantum networks [38, 39]. The idea behind MBQC is to prepare a highly entangled system of qubits, and then perform single qubit adaptive measurements (classical feed forwarding) to mimic the behavior of a quantum circuit (where quantum computations are implemented by applying unitary operations rather than measurements). Quantum networks, on the other hand, are a collection of several quantum processors that are connected through quantum channels and can exchange quantum information for the purpose of quantum communication or distributed quantum computation. Since these applications all require exchanging quantum information across at long distances it is natural to do so using photons. In this section, we give a brief introduction to graph state structures and then we follow up with protocols for the generation of photonic graph states from QDs for applications in MBQC and quantum networks.

### 3.2.1 Graph states and cluster states

A graph state is a multi-qubit state represented by a graph where each vertex represents a qubit and the edges between the qubits denote the entanglement between each pair. Graph states can have either complex forms or much simpler structures. A cluster state is a specific type of graph state where the graph is a d-dimensional lattice or array (Fig. 3.3(a) shows


Figure 3.3: Examples of photonic graph states. (a) Cluster states used for MBQC and (b) repeater graph states uesd in quantum networks.
shows a two-dimensional cluster state).

Rigorously speaking, the graph states correspond to the mathematical graph $G=(V, E)$ with the set of vertices $V$ and the set of edges $E$. A constructive mathematical definition of graph states can be given in terms of $C Z$ gates. In this definition for the graph state represented by graph $G=(V, E)$, we first start by preparing all qubits in the $|+\rangle \equiv(1 / \sqrt{2})(|0\rangle+|1\rangle)$ state, and then we apply a two-qubit $C Z$ gate among each pair of qubits connected by an edge:

$$
\begin{equation*}
|G\rangle=\left(\prod_{(a, b) \in E} C Z_{a b}\right) \bigotimes_{i \in V}|+\rangle_{i} \tag{3.1}
\end{equation*}
$$

As an example, we construct the two-qubit and three-qubit graph states. We start with the initial setup in which $|\psi\rangle=|++\rangle$. Upon applying the $C Z$ between the two qubits, we end up with

$$
\begin{align*}
|G\rangle & =C Z|\psi\rangle=C Z|++\rangle \\
& =C Z\left[\frac{1}{2}(|00\rangle+|01\rangle+|10\rangle+|11\rangle)\right] \\
& =\frac{1}{2}(|00\rangle+|01\rangle+|10\rangle-|11\rangle) \tag{3.2}
\end{align*}
$$

For the three-qubit linear cluster state we start with $|\psi\rangle=|+++\rangle$. Upon applying the $C Z$ between each pair of connected qubits (i.e., vertices that are connected by an edge), we get

$$
\begin{align*}
|G\rangle & =C Z_{12} C Z_{23}|\psi\rangle \\
& =C Z_{12} C Z_{23}\left[\frac{1}{\sqrt{8}}(|000\rangle+|001\rangle+|010\rangle+|011\rangle+|100\rangle+|101\rangle+|110\rangle+|111\rangle)\right] \\
& =\frac{1}{\sqrt{8}}(|000\rangle+|001\rangle+|010\rangle-|011\rangle+|100\rangle+|101\rangle-|110\rangle+|111\rangle) . \tag{3.3}
\end{align*}
$$

Therefore, it can be summarized that a graph state is uniquely determined by a graph via the following rules:

1. Each vertex corresponds to a qubit initialized in a $|+\rangle$ state.
2. Each edge corresponds to a $C Z$ gate between the two corresponding vertices.

### 3.3 Generation of cluster states from quantum dots

In this section, we look at the available protocols for the generation of cluster states from an optically active system. We first look at the Lindner-Rudolph protocol [40] and the challenges with that setup. We also look at an experimental implementation of this protocol. We then look at some of the available alternatives and finally, we present a proposal in QDMs.

### 3.3.1 Lindner-Rudolph protocol

The Lindner-Rudolph (LR) protocol [40] is a method for the generation of cluster states from optically active quantum emitters. The protocol can be implemented in QDs in the


Figure 3.4: Left: The required level structure and selection rules, necessary for the pumping process of the LR protocol for generation of linear cluster states from a quantum emitter such as the self-assembled QDs. Right: The modified selection rules of a self-assembled QD in transverse magnetic fields, necessary requirement for the control process of the LR protocol.
absence of a perpendicular magnetic field that leaves us with the selection rules depicted in Fig. 3.4(a) (Faraday geometry). In this setup, the qubit states are the spin states of the electron in the $Z$ basis (with $J_{z}= \pm 1 / 2$ ) and the excited states are the corresponding trions, $|\Uparrow\rangle$ and $|\Downarrow\rangle$, which have a total $J_{z}= \pm 3 / 2$. In this setup, there is no Zeeman splitting between the qubit states and therefore the transitions have similar energies and are only distinguishable via different polarizations of light; using $\sigma^{ \pm}$with $J_{z}= \pm 1$ excites one or the other transition based on conservation of $J_{z}$. Only photons along the $z$ axis are considered. If the initial state of the source is $|\uparrow\rangle(|\downarrow\rangle)$, an excitation to the state $|T+\rangle(|T-\rangle)$ followed by a spontaneous emission, results in the emission of a single right (left)-circularly polarized photon $\sigma^{+}\left(\sigma^{-}\right)$and leaves the source in the state $|\uparrow\rangle(|\downarrow\rangle)$.

The protocol is based on using a pulse that couples equally to both transitions. This could happen if we send in a pulse in a superposition state, i.e., $(1 / \sqrt{2})\left(\sigma^{+}+\sigma^{-}\right)$which causes the processes described above to happen in superposition:

- Step 0 (initialization): The system is in the state $|\uparrow\rangle+|\downarrow\rangle$.
- Step 1 (pump): Upon excitation with linearly polarized light, now the system is in the
state $|T+\rangle+|T-\rangle$.
- Step 2 (emission): The trion superposition will spontaneously decay to the electron state almost instantaneously, by emitting a single photon. The possible transitions for this singe photon are $|T+\rangle \rightarrow|\uparrow\rangle\left|\sigma^{+}\right\rangle$and $|T-\rangle \rightarrow|\downarrow\rangle\left|\sigma^{-}\right\rangle$. However, since both paths take place simultaneously, the state of the emitted photon and spin will be $|\uparrow\rangle\left|\sigma^{+}\right\rangle+|\downarrow\rangle\left|\sigma^{-}\right\rangle$.

As it can be imagined, repeating this protocol multiple times will lead to several photons entangled with the emitter in the same manner:

$$
|\uparrow\rangle\left|\sigma_{1}^{+}\right\rangle\left|\sigma_{2}^{+}\right\rangle\left|\sigma_{3}^{+}\right\rangle \ldots+|\downarrow\rangle\left|\sigma_{1}^{-}\right\rangle\left|\sigma_{2}^{-}\right\rangle\left|\sigma_{3}^{-}\right\rangle \ldots,
$$

which upon spin encoding $|\uparrow\rangle \equiv|0\rangle,|\downarrow\rangle \equiv|1\rangle$, and polarization encoding $\left|\sigma^{+}\right\rangle \equiv|0\rangle$, $\left|\sigma^{-}\right\rangle \equiv|1\rangle$, corresponds to what is known as a GHZ state [41]:

$$
\begin{equation*}
|\mathrm{GHZ}\rangle \sim|000 \ldots\rangle+|111 \ldots\rangle . \tag{3.4}
\end{equation*}
$$

These states are represented by so-called star graphs in which one vertex is coupled to all others. Here, the central vertex is the emitter, while the others are photons. To produce cluster states, we modify step 2 above with a Hadamard-like gate which rotates the spin within the qubit subspace between each pumping. The original idea by Lindner and Rudolph was to use a $\pi / 2$ rotation about the $Y$-axis, which corresponds to the unitary $\exp (-i Y \pi / 4)$. To see how this works, let us imagine we already have the first photon emitted and the system in step 2 is in the state,

$$
|\uparrow\rangle\left|\sigma_{1}^{+}\right\rangle+|\downarrow\rangle\left|\sigma_{1}^{-}\right\rangle .
$$

Applying the rotation makes the spin state evolve to

$$
(|\uparrow\rangle+|\downarrow\rangle)\left|\sigma_{1}^{+}\right\rangle+(-|\uparrow\rangle+|\downarrow\rangle)\left|\sigma_{1}^{-}\right\rangle .
$$

Now repeating the pump process leads to a second photon emission and the two photons and the electron spin will end up in the state

$$
\begin{equation*}
\left.\left(|\uparrow\rangle\left|\sigma_{2}^{+}\right\rangle+|\downarrow\rangle\left|\sigma_{2}^{-}\right\rangle\right)\left|\sigma_{1}^{+}\right\rangle+\left(-|\uparrow\rangle\left|\sigma_{2}^{+}\right\rangle+|\downarrow\rangle\left|\sigma_{2}^{-}\right\rangle\right)\left|\sigma_{1}^{-}\right\rangle\right) \tag{3.5}
\end{equation*}
$$

This, followed by another $\pi / 2$ rotation of the spin, leads to

$$
\begin{align*}
|G\rangle \sim & (|\uparrow\rangle+|\downarrow\rangle)\left|\sigma_{2}^{+} \sigma_{1}^{+}\right\rangle+(-|\uparrow\rangle+|\downarrow\rangle)\left|\sigma_{2}^{-} \sigma_{1}^{+}\right\rangle \\
& -(|\uparrow\rangle+|\downarrow\rangle)\left|\sigma_{2}^{+} \sigma_{1}^{-}\right\rangle+(-|\uparrow\rangle+|\downarrow\rangle)\left|\sigma_{2}^{-} \sigma_{1}^{-}\right\rangle \\
= & \left|\uparrow \sigma_{2}^{+} \sigma_{1}^{+}\right\rangle+\left|\downarrow \sigma_{2}^{+} \sigma_{1}^{+}\right\rangle-\left|\uparrow \sigma_{2}^{-} \sigma_{1}^{+}\right\rangle+\left|\downarrow \sigma_{2}^{-} \sigma_{1}^{+}\right\rangle \\
& -\left|\uparrow \sigma_{2}^{+} \sigma_{1}^{-}\right\rangle-\left|\downarrow \sigma_{2}^{+} \sigma_{1}^{-}\right\rangle-\left|\uparrow \sigma_{2}^{-} \sigma_{1}^{-}\right\rangle+\left|\downarrow \sigma_{2}^{-} \sigma_{1}^{-}\right\rangle . \tag{3.6}
\end{align*}
$$

Now if we use the encodings $\left|\sigma^{+}\right\rangle \equiv|0\rangle$ and $\left|\sigma^{-}\right\rangle \equiv-|1\rangle$, we can see that the state above corresponds to

$$
\begin{equation*}
|G\rangle \sim|000\rangle+|001\rangle+|010\rangle-|011\rangle+|100\rangle+|101\rangle-|110\rangle+|111\rangle \tag{3.7}
\end{equation*}
$$

which is exactly the three-qubit linear cluster state from Eq. 3.3. Repeating this protocol will produce a continuous chain of photons in an entangled linear cluster state.

The $\exp (-i Y \pi / 4)$ rotation of the procedure is a Hadamard-like gate; in fact a more systematic way to think about the LR proposal is that it is an alternation between optical pumping and applications of this Hadamard-like gate. A generalization of this Hadamard-like gate is [42]

$$
H^{\prime}=\frac{1}{\sqrt{2}}\left(\begin{array}{cc}
e^{i \theta_{1}} & e^{i\left(\theta_{1}+\phi\right)}  \tag{3.8}\\
e^{i\left(\theta_{2}-\phi\right)} & -e^{i \theta_{2}}
\end{array}\right)
$$

For the maximally entangled state we will require $\theta_{2}=\theta_{1}=\phi=0$. In the original LR protocol, the implementation of the $Y$-rotation is suggested through application of an external magnetic field in the $Y$ direction: The idea is to allow the spin to precess in the so-called Voigt geometry (i.e., in the presence of a transverse magnetic field, Fig. 3.4(b)) in order to perform the spin rotation; for a Larmor frequency of $\omega_{B}$, the desired rotation is achieved with the cycle $T_{\text {cycle }}=\pi / 2 \omega_{B}$. This could be problematic in the case of single QDs because the Voigt field will remove the Faraday selection rules since in a Voigt geometry both vertical and diagonal transitions are present.

### 3.3.2 Experimental demonstration of a one-dimensional cluster state

Schwartz et al., demonstrated an experimental implementation of the LR protocol in selfassembled quantum dots in 2016 [6]. In this experiment they used the dark excitons (see Section 2.2.2) as their qubit states (in terms of Fig. 3.4, $|q+\rangle \equiv|\uparrow \Uparrow\rangle=|+2\rangle$, and $|q-\rangle \equiv|\downarrow \Downarrow\rangle=|-2\rangle$ ) and their transitions to the biexciton (in terms of Fig. 3.4, $|T+\rangle \equiv|+3\rangle$, and $|T-\rangle \equiv|-3\rangle)$ levels to create the required level structure of the LR protocol. They successfully showed the entanglement between the dark exciton qubit and a string of photons

## A Real $\hat{\rho}_{+Z}^{\text {meas }}$



Figure 3.5: Deterministic generation of entangled cluster states by Schwartz et al. [6]. The left figure shows the selection rules between the dark exciton states $(| \pm 2\rangle)$ and their corresponding excited states, the biexcitons $(| \pm 3\rangle)$. The right figure shows the experimental data of the resulting density matrix of the emitted photon and the qubit (dark exciton) which corresponds to an entangled state. Both figures are taken from Ref. [6].
that were generated from this system (see Fig. 3.5). The control part of the LR protocol was implemented here by the precession of the dark excitons (about 3 ns [31]). The number of entangled of photons in this experiment was limited due to the fact that the qubit states are excited states and therefore have relatively short lifetimes, and furthermore, the nuclear environment may lead to dephasing effects. The protocol that we will introduce in Section 3.3.4 will overcome both of these issues.

### 3.3.3 Time-bin protocols

To overcome the control issues of the LR protocol, in Ref. [43] Lee et al. proposed to start in a Voigt geometry and selectively enhance one of the transitions to the trion states (highlighted blue transition in Fig. 3.4(b)) by placing the QD in a cavity and encoding the photons in time-bins of emission from this transition. Time-bin encoding, as opposed to the polarization
encoding that we used in the previous section (i.e., $\left|\sigma^{+}\right\rangle \equiv|0\rangle$, $\left|\sigma^{-}\right\rangle \equiv|1\rangle$ ), encodes the quantum bits in terms of presence and absence of photon, where $|0\rangle(|1\rangle)$ represents the absence (presence) of a photon in a time bin. This proposal significantly improves the controllability process of the LR protocol but it is not fully deterministic as there is a nonzero probability of emission from the unenhanced transition. Comparing this strategy to the LR protocol we find a competition between emission and control: Spin control works best in a Voigt geometry with a three-level $\Lambda$-system to enable the coherent control, but it is not optimal for the emission of photons since a trion would spontaneously emit a photon through the spin up or down transitions. On the other hand, the Faraday geometry is suitable for good photon emission but the spin control implementation is complicated.

### 3.3.4 Generation of cluster states from quantum dot molecules

In this section, we present protocols for generating photonic cluster states using the features and selection rules of QDMs introduced in Section 2.3. An important feature is the fact that the hole spins states are intrinsically split without the use of an external magnetic field due to hole mixing. Furthermore, using hole spin qubits naturally removes the difficulty of nuclear spin interactions due to their $p$-like orbitals. The full control scheme of QDMs is depicted in Fig. 2.10, where the $\Lambda$-type system in the middle is used to manipulate the spin state, and the two external arms (cycling transitions) are used for readout. We will make use of these selection rules for different strategies given in the following.

For the implementation of a LR-like protocol, we use the readout arms. A fundamental difference between the actual LR proposal and the QDMs is that in the LR scheme, the two transitions have the same energy, that is, the only way to distinguish the photons is through their polarization. In the QDMs (and similarly, QDs with the Voigt geometry) the
two available transitions do not have the same energy.
Alternatively, we may consider the protocol that corresponds to the time-bin protocol discussed in Section 3.3.3 for QDs. In this case let us consider the cycling transition with $\sigma^{-}$ to be the corresponding enhanced transition for the time-bin protocol. Here $\binom{0,0}{0, \uparrow} \equiv|0\rangle$ and $\binom{0,0}{0, \Downarrow}=|1\rangle$. In the following steps of the protocol we will use $|h\rangle \equiv\binom{0,0}{0, \Uparrow}$ and $|\bar{h}\rangle \equiv\binom{0,0}{0, \Downarrow}$, to denote the two hole states. The time-bin protocol will be as follows:

- Step 0: We initialize the system by putting the population in $\binom{0,0}{0, \uparrow}$ state and then create a superposition state $|+\rangle=(1 / \sqrt{2})(|h\rangle+|\bar{h}\rangle)$ by performing a $\pi / 2$ rotation through the $\Lambda$-system.
- Step 1: Then we drive the $\sigma^{-}$cycling transition. This creates the first time bin in the form of

$$
\begin{equation*}
\frac{1}{\sqrt{2}}\left(|h\rangle\left|0_{\tau=1}\right\rangle+|\bar{h}\rangle\left|1_{\tau=1}\right\rangle\right) \tag{3.9}
\end{equation*}
$$

where the $\tau$ subscript denote the first time bin.

- Step 2: In order to create a time-bin photon in the opposite state, now we need to flip the spin. We perform $R_{Y}(i \pi)=\exp (i Y \pi / 2)$. This results in the state

$$
\begin{equation*}
\frac{1}{\sqrt{2}}\left(|\bar{h}\rangle\left|0_{\tau=1}\right\rangle-|h\rangle\left|1_{\tau=1}\right\rangle\right) \tag{3.10}
\end{equation*}
$$

- Step 3: Now again we drive the same $\sigma^{+}$cycling transition, to get

$$
\begin{equation*}
\frac{1}{\sqrt{2}}\left(|\bar{h}\rangle\left|0_{\tau=1} 1_{\tau=2}\right\rangle-|h\rangle\left|1_{\tau=1} 0_{\tau=2}\right\rangle\right) \tag{3.11}
\end{equation*}
$$

- Step 4: Another spin flip, similar to the previous step, leads to

$$
\frac{1}{\sqrt{2}}\left(-|h\rangle\left|0_{\tau=1} 1_{\tau=2}\right\rangle+|\bar{h}\rangle\left|1_{\tau=1} 0_{\tau=2}\right\rangle\right) .
$$

- Step 5: Repeating these steps three more times leaves us with

$$
\frac{1}{\sqrt{2}}\left(|\bar{h}\rangle\left|0_{\tau=1} 1_{\tau=2} 0_{\tau=3} 1_{\tau=4} 0_{\tau=5} 1_{\tau=6}\right\rangle-|h\rangle\left|1_{\tau=1} 0_{\tau=2} 1_{\tau=3} 0_{\tau=4} 1_{\tau=5} 0_{\tau=6}\right\rangle\right) .
$$

- Step 6: The final step is to measure out the spin part. If the spin is in $|-\rangle$, and we take photon in an odd numbered time-bin to be a logical 1 and a photon in an even numbered time-bin as a logical 0 we get the GHZ state

$$
\frac{1}{\sqrt{2}}(|000\rangle+|111\rangle)
$$

Up to this point we have only used the cycling transitions to produce GHZ states. It is straight forward to expand this protocol and apply extra rotations in between the steps above to get linear cluster states [43].

As seen so far, the level structure of the QDMs provides us with several advantages in implementing the time-bin protocol, both compared to the LR protocol, and also to the time-bin protocol with individual QDs:

1. External magnetic field and Voigt geometry: In the original LR proposal a weak perpendicular magnetic field is introduced to implement the Hadamard gates ( $Y$-rotations). But this could be problematic since the LR protocol requires Faradaylike selection rules and it is not clear how much the weak magnetic field will affect the selection rules. In QDMs, however, none of this would be a problem due to the intrinsic splittings of the hole states in the absence of an external magnetic field.
2. Implementation of rotations: In the original time-bin protocol it is proposed to use the same $\Lambda$-system used for the time-bin photons to implement the necessary rotations for the protocol. It is not clear how the transfer of populations and spontaneous emission from the system would affect the procedure (the leakage to the other spin state). In QDMs these rotations are implemented through the independent $\Lambda$-system that does not participate in the rest of the time-bin protocol and is solely used for rotations. Put another way, QDMs have the advantage of using cycling transitions specifically for the time-bin protocol qubit generation and the $\Lambda$-system for rotations, while the original proposal needs to use one single $\Lambda$-system for both purposes.
3. Cavity enhancement of a single transition: In the original time-bin protocol the enhancement of a particular transition of the $\Lambda$-system (highlighted transition Fig. 3.4(b)) is required (while both transitions of the $\Lambda$-system have the same selection rules). This enhancement requirement is eased in QDMs since we instead use the cycling transitions, which have opposite polarizations. Therefore it is less costly to produce the time-bins in QDMs.

In other words, in QDMs, we have the best of both worlds: A $\Lambda$-system that implements the rotations, and cycling transitions even at zero magnetic field that emit photons. In a way, it is as if we have Faraday transitions for optical pumping and Voigt transitions for spin control.

As one would expect, though, no advantages come for free: As we discussed in Section 2.3, the $\Lambda$-system in QDMs has two closely spaced excited levels $|t\rangle$ and $|u\rangle$ that will be problematic for implementation of quantum gates. The off-resonant coupling to the excited level will cause phase errors which hurt the fidelity of our gates. In the following chapter we develop a method for implementing the desired gates while battling the off-resonant coupling to
unwanted levels.

## Chapter 4

## High-fidelity control in $\Lambda$-systems with leakage

In this chapter we provide a quantum control method for $\Lambda$-systems with leakage. While the treatment presented here is generic and applicable to any optically active system, we bear in mind the application to the $\Lambda$-systems of the QDMs. The work presented in this section is partially based on preprint the 'Ultrafast high-fidelity control of $\Lambda$-systems in the presence of unwanted transitions' by Arian Vezvaee, Eva Takou, Paul Hilaire, Matthew Doty, and Sophia E. Economou.

### 4.1 Motivation

Quantum information processing requires the manipulation of qubits via fast gates with high fidelities. Qubits are formed when a particular two-level subspace is chosen from a larger Hilbert space of a physical system. In specific cases, energy levels outside of the qubit subspace are used for auxiliary transfer of population within the qubit subspace. An important class of such setups are $\Lambda$-type systems that appear at the heart of several optically active systems such as QDs [44, 45, 46, 47, 48], nitrogen-vacancy (NV) centers [49, 50, 51], and trapped ions [52, 53, 54, 55]. Successful manipulation of $\Lambda$-systems in these optical devices is the key step to performing either quantum information processing, or enabling sources


Figure 4.1: Schematic depiction of a general $\Lambda$-type system with leakage. The qubit is defined in the subspace of the two lower levels $\{|0\rangle,|1\rangle\}$. The coherent control is done through driving the transitions to the auxiliary target level $|t\rangle$ (with the detuning $\delta$ ) which is separated from the unwanted level $|u\rangle$ by the splitting $\varepsilon$. The off-resonant coupling to the unwanted level will cause low fidelities. We present resolutions for this problem by introducing a modification to the pulse and the detuning of the system.
of single-photon emission for both linear optical quantum computing [56] and quantum communications [57, 58]. Consequently, various methods for manipulation of $\Lambda$-systems and their variations have been studied extensively as an important tool in quantum optics $[59,60,61,62,63,64]$. However, in most systems a bare three-level $\Lambda$-system is merely an idealization; perfect isolation and manipulation of a dissociated part of the Hilbert space is almost infeasible [65]. Unintended interactions with other levels cause unwanted leakage out of the three-level subspace that is detrimental to the performance of quantum gates [33, 66]. The effect of these off-resonant unwanted couplings is intensified when the extra levels are closely spaced in energy with the auxiliary state.

The issue of interactions with unwanted levels can be dealt with in several forms. For instance, in superconducting qubits it is resolved by intentionally driving the harmful transition [67]. The same approach has been shown to be applicable to NV centers as well [68]. Machine learning has also been used recently to optimize the performance of quantum gates [69]. Furthermore, there also exist frameworks that involve adiabatic removal of the leakage from the system $[70,71,72,73,74]$. A recent example of such adiabatic methods is the Magnus
expansion [75]. Another well-known adiabatic method is Derivative Removal by Adiabatic Gates (DRAG) which has been widely established as a powerful tool in dealing with unwanted dynamics and leakage cancellations [76, 77, 78]. The underlying mechanism behind DRAG allows one to infer additional control elements that oppose the leakage. For instance, such extra elements could be in the form of a second control pulse that drives the same target transition, leading to cancellation of the leakage to unwanted levels. The DRAG approach was originally proposed in superconducting devices where the qubits are encoded in the form of a two-level system in the energy spectrum of an anharmonic oscillator. Since its proposal, DRAG has been extensively studied over the past decade for implementations in superconducting devices [79, 80] and several DRAG-inspired methods have been developed [81].

In this chapter we develop a novel DRAG-based leakage cancellation framework relevant for systems with $\Lambda$-type selection rules, where the auxiliary (target) excited state is separated from an unwanted excited level (Fig. 4.1) by a small energy splitting. We present an ultrafast solution in the form of additional control elements that oppose the leakage and remove the need for impractical narrow-band pulses that have gate times that far exceed the quantum emitter lifetime. The chapter is structured as follows. In Section 4.2 we present the system under consideration and give an overview of Coherent Population Trapping (CPT) [63], which is a quantum optical tool that enables implementation of arbitrary rotations in an idealized version of $\Lambda$-systems. In Section 4.3 we discuss the DRAG methodology and develop a new formalism that resolves the leakage issue of $\Lambda$-systems and leads to high-fidelity rotations. We conclude in Section 4.4, and the subsequent sections contain details of the DRAG analysis for an arbitrary axis of rotation under the CPT framework.


Figure 4.2: Fidelity of an $X$-, and equivalently a $Y$-rotation by $\pi, R_{X / Y}(\pi)$, in terms of the dimensionless parameter $\sigma / \varepsilon$. Here, $\sigma$ is the bandwidth of the sech pulse and $\varepsilon$ is the splitting between the unwanted and target levels. The fidelities are shown for different values of $\eta$ that determines the composition of the target and unwanted levels. Without any corrective measures, reasonable fidelities required for quantum information processing are only achievable by using extremely narrow bandwidth pulses.

### 4.2 Overview of the system

In an ideal CPT scheme the transitions of a perfectly isolated $\Lambda$-system are driven using two drive fields. When the drive fields interfere destructively the population is trapped into a dark state. This phenomenon depends on the drive parameters, which also define the transformation of the qubit states to the CPT (dark-bright) frame; in this frame, the dark state is completely decoupled from the dynamics of the system. Essentially, the three-level problem reduces to a two-level system where transitions are driven between the auxiliary excited state and the bright state. Combining CPT together with sech pulse envelopes we can design arbitrary single qubit rotations about any axis, as explained in Ref. [46]. We leave the technical details of the CPT framework and the sech-pulse control to Section 4.5.

The system under consideration is depicted in Fig. 4.1; a collection of four states where the qubit is encoded in the two lower levels $\{|0\rangle,|1\rangle\}$ and either of the two excited levels may be used as an auxiliary level for the qubit control. In contrast to the ideal CPT scheme, the extra excited level introduces competing transitions that cause leakage out of the threelevel subsystem. Our goal is to perform the control of the qubit states using the auxiliary (target) level while avoiding the detrimental effect of the unwanted transition. In this work we use the lower excited level to be our target, and denote its energy splitting from the upper unwanted level with $\varepsilon$. We further choose the frequency of the control pulses to be smaller than the transition frequency of the target transitions (i.e. negative detuning, $\delta$ ); this choice minimizes the leakage to the unwanted level. However, the analysis we present in this work will be exactly the same if we swap the definition of the unwanted and target levels and change the sign of the detuning. We also assume that the two excited states, $|t\rangle$ and $|u\rangle$, are superpositions of two basis states $\left|b_{0}\right\rangle$ and $\left|b_{1}\right\rangle:|t\rangle=\sin (\eta)\left|b_{1}\right\rangle-\cos (\eta)\left|b_{0}\right\rangle$ and $|u\rangle=\cos (\eta)\left|b_{1}\right\rangle+\sin (\eta)\left|b_{0}\right\rangle$.

We consider the case where the two $\Lambda$-transitions are distinct and thus, each transition is driven by a single drive field $\left[E_{0}(t)\right.$ or $\left.E_{1}(t)\right]$, as shown in Fig. 4.1. This can be satisfied by either polarization selection rules or energy separation of the ground states. In the former case, the orthogonality of the two transition dipoles ensures that each transition couples to a single drive. In the latter case, sufficient energy separation of the ground-state levels implies that the off-resonant couplings of the drive fields to the opposite $\Lambda$-transitions average out. In the following we first consider the case of distinct couplings to the drive fields, where $|0\rangle$ couples only to $\left|b_{0}\right\rangle$ with Rabi frequency $\Omega_{0}(t) \equiv-\sin (\eta) d_{0, b_{0}} E_{0}$, and $|1\rangle$ couples only to $\left|b_{1}\right\rangle$ with Rabi frequency $\Omega_{1}(t) \equiv \cos (\eta) d_{1, b_{1}} E_{1}$. This choice translates into couplings to the unwanted level given by $\lambda_{0}$ (for the $|0\rangle \leftrightarrow|u\rangle$ transition driven by $E_{0}$ ) and $\lambda_{1}$ (for the $|1\rangle \leftrightarrow|u\rangle$ transition driven by $\left.E_{1}\right)$ that are inversely related: $\lambda_{0} \equiv \tan (\eta)=-1 / \lambda_{1}$. However, the formalism we develop is general and not restricted by the composition of the excited states. We keep this relation among the dipole elements implicit and discuss how our framework can be applied to the case of bare coupling to target and unwanted (i.e., in the absence of any basis states) in Section 4.4.

As shown in Fig. 4.2, the presence of an unwanted transition leads to low gate fidelities when the bandwidth of the pulses is not extremely narrow. One solution to preserve selectivity and ensure high-fidelity gates is to use extremely long pulses. Nevertheless, this is an impractical approach as the operations need to performed well within the coherence times. In all relevant solid state emitters, long gates suffer from spontaneous emission. Spontaneous emission generally occurs on very fast timescales (e.g., $\sim$ one ns in QDs [82] and 10 ns in NV centers [83]), implying that high-fidelity gates require pulses much shorter than these timescales. In this work we develop a formalism that deals with the issue of leakage without trading off the duration of the gates for selectivity, ensuring operations performed within the coherence times. We will show how to implement fast and leakage-protected gates by


Figure 4.3: Selection rules in the dressed basis of CPT in the case of equal couplings, i.e., $\eta=\pi / 4$. In this case the problem reduces to two dissociated two-levels (bright and target, and dark and unwanted), each subject to a transitionless pulse.
modifying the pulse shape and the static detuning of the drive fields.

### 4.3 DRAG formalism with coherent population trapping

The presence of an additional excited state induces leakage outside of the $\Lambda$-subspace, which in the dressed (CPT) frame translates into error transitions that link the bright and dark states to the unwanted $|u\rangle$ level. In an ideal $\Lambda$-system, one would drive the target transitions with the fields $E_{\ell}(t)=\Omega_{\ell, o}(t)(\ell=0,1)$ (and static detuning $\delta$ ) to implement the desired gate operation. To resolve the leakage we modulate the original pulses. We consider an additional corrective drive $\Omega_{l, c}(t)$ (for each of the two fields), phase detuned from the original by $\pi / 2$; we further set the frequency to be the same as that of the original drive, hence reducing the experimental overhead of an additional pulse. From here on we will use the letters $o$ and $c$ to refer to any subsequent parameters of these two drive fields.

The total fields of the system are $E_{\ell}(t)=\Omega_{\ell, o}(t) \cos \left(\omega_{\ell, d} t\right)+\Omega_{\ell, c}(t) \sin \left(\omega_{\ell, d} t\right)$ for $\ell=0,1$,
which in the rotating wave approximation (RWA) are equivalent to $E_{\ell}(t)=\exp \left(-i \omega_{d} t\right)\left(\Omega_{\ell, o}(t)+\right.$ $\left.i \Omega_{\ell, c}(t)\right)+$ c.c.. Under RWA, the dimensionless Hamiltonian in the CPT frame is given by (see Section 4.5)

$$
\begin{align*}
\bar{H}_{\mathrm{CPT}, \omega d}= & \frac{1}{x} \Pi_{u}+(\bar{\delta} / 2)\left(\Pi_{B}+\Pi_{D}+\Pi_{t}\right) \\
& +\frac{1}{2 \sqrt{2}}\left\{-i\left(\bar{\Omega}_{0, c}-\bar{\Omega}_{1, c}\right)|t\rangle\langle D|\right. \\
& +\left(2 \bar{\Omega}_{o}-i\left(\bar{\Omega}_{0, c}+\bar{\Omega}_{1, c}\right)\right)|t\rangle\langle B| \\
& +\left(\bar{\Omega}_{o}\left(\lambda_{0}-\lambda_{1}\right)+i\left(\lambda_{1} \bar{\Omega}_{1, c}-\lambda_{0} \bar{\Omega}_{0, c}\right)\right)|u\rangle\langle D| \\
& +\left(\bar{\Omega}_{o}\left(\lambda_{0}+\lambda_{1}\right)-i\left(\lambda_{1} \bar{\Omega}_{1, c}+\lambda_{0} \bar{\Omega}_{0, c}\right)\right)|u\rangle\langle B| \\
& + \text { h.c. }\} . \tag{4.1}
\end{align*}
$$

where $\Pi_{m} \equiv|m\rangle\langle m|$ and we have introduced the adiabatic parameter $x=1 /\left(\varepsilon t_{g}\right)$. The dimensionless form is obtained by multiplying all quantities by the gate time $t_{g}[78]: \bar{O}(t)=$ $t_{g} O(t)$ for $O \in\left\{\Omega_{o}, \Omega_{j, c}, \delta\right\}$. We have set the two Rabi frequencies of the target transitions (i.e. $|0\rangle \leftrightarrow|t\rangle$ and $|1\rangle \leftrightarrow|t\rangle$ ) to be equal, that is $\Omega_{0}=\Omega_{1}$, such that we satisfy the transformation for implementing X (or equivalently Y, up to a phase between the two drives) gates. The coupling strengths to the unwanted level, $|u\rangle$, are scaled to the target transition strengths by the parameters $\lambda_{0}$ and $\lambda_{1}$ and satisfy the relation $\lambda_{0} \equiv \tan (\eta)=-1 / \lambda_{1}$. For $\eta=\pi / 4$ the bright-unwanted transition vanishes leading to the dressed frame selection rules shown in Fig. 4.3 ${ }^{1}$. On the other hand, if $\eta \neq \pi / 4$ we have an additional leakage transition to consider.

We now outline the development of our new DRAG technique applicable to $\Lambda$-type structures to mitigate all leakage transitions. Previous formulations of the DRAG method have focused on cancelling out leakage errors in a ladder-type system (e.g. transmon) that occur between

[^1]consecutive energy levels. However, the formalism is not directly applicable to $\Lambda$-systems. The complexity in this case increases as the qubit control is performed indirectly via the excited auxiliary level. As we will discuss shortly, we will also need to develop additional tools to accompany our modified DRAG method and ensure improvement of our control scheme.

The DRAG method relies on adiabatic frame transformations from which analytic counterdiabatic corrections to the driving fields can be derived [81]. DRAG introduces a new frame where the solution to the leakage issue translates into a set of constraints imposed on the Hamiltonian of this frame. The DRAG frame Hamiltonian generated by the transformation $A(t)=e^{-i S(t)}$ is:

$$
\begin{equation*}
H_{\mathrm{DRAG}}=A^{\dagger}(t) H A(t)+i \dot{A}^{\dagger}(t) A(t) \tag{4.2}
\end{equation*}
$$

where $H$ is the Hamiltonian in the original frame (in this work, we start from the CPT frame, such that $\left.H \equiv \bar{H}_{C P T, \omega_{d}}\right)$. The operator $S(t)$ can be any arbitrary Hermitian operator, but needs to respect the boundary conditions of the transformation. That is, the frame transformation has to vanish at the beginning and end of the pulse $\left[A(t)=A\left(t_{g}\right)=\mathbf{1}\right]$, such that the ideal gate we wish to design remains the same in both the CPT and DRAG frames. Besides this restriction, $S(t)$ can be an arbitrary Hermitian operator that aims to decouple the desired evolution from the leakage subspace. The generality of $S(t)$ would in principle generate a wide range of counterdiabatic corrections. However, extracting such closed-form expressions is infeasible for our four-level system (as the pulses do not vary slowly in time) and hence, we turn to a perturbative expansion of the transformation. To that end, in this work we utilize the Schrieffer-Wolff (SW) transformation [84] and its perturbative expansion.

Our goal is to constraint the DRAG-frame Hamiltonian such that it implements our target
evolution. To this end, we define a target Hamiltonian, capable of performing arbitrary rotations within the qubit (dark-bright) subspace,

$$
\begin{equation*}
H_{\mathrm{target}}^{\mathrm{CPT}}(t)=\frac{1}{2} \sum_{i=o, c} h_{i}(t) \sigma_{B, t}^{i}+\frac{1}{2} h_{z}(t)\left(\Pi_{B}-\Pi_{t}\right) \tag{4.3}
\end{equation*}
$$

where $h_{i}(t)$ and $h_{z}(t)$ are arbitrary control fields, and to to indicate the matrix elements for the generic transition $|m\rangle \leftrightarrow|n\rangle$, we have defined $\sigma_{m, n}^{\ell} \equiv e^{i \beta}|n\rangle\langle m|+e^{-i \beta}|m\rangle\langle n|$ with $\beta=0$ for $\ell=o$, and $\beta=\pi / 2$ for $\ell=c$. Here we pick the $h_{i}(t)$ to be sech pulses within the bright and target subspace, and $h_{z}(t)$ corresponds to the detuning. Contrary to the target Hamiltonian, our DRAG frame Hamiltonian includes the error transitions to the unwanted level $|u\rangle$. To ensure that the DRAG frame Hamiltonian implements the intended operation as dictated by $H_{\text {target }}^{\mathrm{CPT}}$, we impose the following constraints:

$$
\begin{align*}
& \operatorname{Tr}\left[H_{\mathrm{DRAG}}(t) \sigma_{B, t}^{i}\right]=h_{i}(t),  \tag{4.4}\\
& \operatorname{Tr}\left[H_{\mathrm{DRAG}}(t)\left(\Pi_{B}-\Pi_{t}\right)\right]=h_{z}(t),
\end{align*}
$$

where $i \in\{o, c\}$. Additionally, to enforce decoupling from the unwanted subspace in the new Hamiltonian, we impose the following constraints:

$$
\begin{align*}
\operatorname{Tr}\left[H_{\mathrm{DRAG}}(t) \sigma_{D, u}^{i}\right] & =0, \\
\operatorname{Tr}\left[H_{\mathrm{DRAG}}(t) \sigma_{B, u}^{i}\right] & =0,  \tag{4.5}\\
\operatorname{Tr}\left[H_{\mathrm{DRAG}}(t) \sigma_{t, u}^{i}\right] & =0,
\end{align*}
$$

where $i \in\{o, c\}$. Note that from the Hamiltonian (4.1), for the transition $\sigma_{D, t}$ (i.e., the



Figure 4.4: Gate error comparison in terms of the dimensionless parameter $\sigma / \varepsilon$ for the original pulse (solid) and DRAG solution (dotted) for (a) $\pi / 2$ and (b) $\pi$ rotation about the X or Y axis. Bottom panels show the required modification that accompanies the corrective pulses in each case. The colors correspond to different weight distributions of the basis states in the target and unwanted levels $|t\rangle=\sin (\eta)\left|b_{1}\right\rangle-\cos (\eta)\left|b_{0}\right\rangle$ and $|u\rangle=$ $\cos (\eta)\left|b_{1}\right\rangle+\sin (\eta)\left|b_{0}\right\rangle$.
transition from dark state to target), the decoupling condition is trivially satisfied only if we pick $\Omega_{0, c}=\Omega_{1, c}$. The constraints can be solved consistently by expanding the control fields of the DRAG Hamiltonian and the Hermitian operator $S(t)$ with respect to the adiabatic parameter $x$ to several orders. The technical details of the derivation for our corrective fields can be found in Section 4.6.

To the first order of expansion, the simplest solution which respects the boundary conditions of the transformation that satisfies the constraints is

$$
\begin{align*}
\Omega_{o}(t) & =\left(\sqrt{2}+\delta \frac{\left(\lambda_{0}+\lambda_{1}\right)^{2}}{4 \sqrt{2} \varepsilon}\right) \Omega(t)  \tag{4.6}\\
\Omega_{0, c}(t) & =\Omega_{1, c}(t)=\frac{\sqrt{2}}{8 \varepsilon}\left(\lambda_{0}+\lambda_{1}\right)^{2} \dot{\Omega}(t) \tag{4.7}
\end{align*}
$$

These solutions are capable of reducing the unintended couplings of the bright-excited subspace to the unwanted level. In the original DRAG framework one would use these modified pulses and expect improvement of the fidelities of quantum gates. However, our scheme relies on the indirect control of the qubit states via the auxiliary target state. The diagonal constraint that we impose in Eq. (4.4) does not lead to a global phase among all states: It imposes an identical phase between bright and excited states, but it does not ensure the same phase with the dark state. As such, we need to investigate the form of the first-order DRAG Hamiltonian to infer this phase error between the dark and bright states. Restricting our attention to the $\Lambda$-system subspace, we find that the first-order DRAG frame Hamiltonian in the basis $\{|D\rangle,|B\rangle,|t\rangle\}$ is given by (see Section 4.6):

$$
H_{\mathrm{DRAG}}^{(1)}=\left[\begin{array}{ccc}
-\frac{1}{8}\left(\lambda_{0}-\lambda_{1}\right)^{2} \Omega^{2} & \frac{1}{8}\left(-\lambda_{0}^{2}+\lambda_{1}^{2}\right) \Omega^{2} & 0  \tag{4.8}\\
\frac{1}{8}\left(-\lambda_{0}^{2}+\lambda_{1}^{2}\right) \Omega^{2} & -\frac{1}{16}\left(\lambda_{0}+\lambda_{1}\right)^{2} \Omega^{2} & 0 \\
0 & 0 & -\frac{1}{16}\left(\lambda_{0}+\lambda_{1}\right)^{2} \Omega^{2}
\end{array}\right]
$$

Notice that the Hamiltonian above is in the DRAG frame, and is enforced to have no leakage, therefore there is no dependence on the splitting, and the value of detuning is fixed for this Hamiltonian from the constraints of Eq. (4.4). However, note that the bright and target states follow the same phase evolution, as dictated by the common diagonal element $-1 / 16\left(\lambda_{0}+\lambda_{1}\right)^{2} \Omega^{2}$. Our qubit states, on the other hand, are composed of the dark and
bright states, which up to the first-order (i.e ignoring the off-diagonal elements of $H_{\text {DRAG }}^{(1)}$ ) evolve with a different phase. Effectively, this implies that the DRAG corrections reduce the leakage to the unwanted level at the cost of inducing a relative phase between the qubit states. This is an immediate consequence of the fact that we counteract leakage indirectly; in the DRAG frame we design a target Hamiltonian that involves transitions between the bright and target levels.

For $\lambda_{0}=-\lambda_{1}$ (i.e., $\eta \approx \pi / 4$ ) Eq. (4.8) takes an interesting form. In this case, both diagonal and off-diagonal elements of $H_{\text {DRAG }}^{(1)}$ vanish (except the diagonal entry of the dark state). The vanishing of the latter implies that in this first-order DRAG frame the higher-order single-qubit rotation errors induced in the qubit subspace are zero. Nevertheless, the dark state evolves with a different phase relative to the bright-target subspace. This phase error can be easily compensated for through a static detuning modification, i.e. a change of the frequency of the driving pulses. Surprisingly enough, at the same time, the corrections to the pulses vanish such that no pulse modification is required to correct the errors.

Let us highlight the procedure for finding the required detuning modification when the unwanted couplings satisfy the condition $\lambda_{0}=-\lambda_{1}$. First, we redefine the Hamiltonian by subtracting from all diagonal entries the $\Pi_{D}$ element and thus, transferring a time-dependent phase evolution into the bright-target subspace (while ensuring zero phase-evolution of the dark state). We denote the ideal evolution operator that corresponds to $H_{\text {DRAG }}^{(0)}$ as $U_{0}$; this is the analytically solvable time evolution operator. Our total Hamiltonian (up to the first order $), H_{\mathrm{DRAG}}=H_{\mathrm{DRAG}}^{(0)}+H_{\mathrm{DRAG}}^{(1)}$, evolves with a time evolution given by $U(t)=U_{0}(t) U\left(t^{\prime}\right.$, and satisfies the Schrödinger equation:

$$
\begin{equation*}
i \dot{U}_{0} U^{\prime}+i U_{0} \dot{U}^{\prime}=\left(H_{D}^{(0)}+H_{D}^{(1)}\right) U_{0} U^{\prime} \tag{4.9}
\end{equation*}
$$

which reduces to:

$$
\begin{equation*}
i \dot{U}^{\prime}=\left(U_{0}^{\dagger} H_{\mathrm{DRAG}}^{(1)} U_{0}\right) U^{\prime} \tag{4.10}
\end{equation*}
$$

$U^{\prime}$ is the evolution operator of $H_{\mathrm{DRAG}}^{(1)}$ in the interaction picture of $H_{\mathrm{DRAG}}^{(0)}$. In the brighttarget subspace, $H_{\mathrm{DRAG}}^{(1)} \propto f(t) \mathbf{1}$, where $f(t)$ is the function that defines the relative phase shift between the dark and bright states. Solving the Schrödinger equation we find that the induced phase shift is given by $\theta=-2 \lambda^{2} \sigma / \varepsilon$. Hence, in order to remove the $\theta$-shift from the target evolution we modify the detuning as:

$$
\begin{equation*}
\delta^{\prime}=\sigma / \tan \left(\frac{\phi+\theta}{2}\right) \tag{4.11}
\end{equation*}
$$

where $\phi$ is the (analytically known) rotation angle we wish to perform. For the general case where $\lambda_{0} \neq-\lambda_{1}$, we follow a similar procedure where we ignore the off-diagonal entries of $H_{\text {DRAG }}^{(1)}$. We find that the induced phase is $\theta=-\left(\lambda_{0}^{2}+\lambda_{1}^{2}-6 \lambda_{0} \lambda_{1}\right) \sigma /(4 \varepsilon)$, and can be accounted for, by modifying the detuning according to Eq. (4.11).

The detuning modification together with Eqs. (4.7) and (4.6) complete our full set of solutions to the pulses. The final pulse shapes will depend on the specific details of the system such as the excited states splitting and the weight of the basis states that form the target and unwanted levels. In Fig. 4.5 we depict the sech pulse envelopes for one specific choice of parameters.

We quantify the performance of the gates by averaging over all input states existing in the Hilbert space. Bowdrey et al. showed that this measure of gate fidelity can be computed by averaging over the fidelities of the six cardinal pure states [85]:


Figure 4.5: An example of pulse shapes for a system with $\eta=\pi / 3$ and splitting of $\varepsilon=80 \mu \mathrm{~V}$. The sech pulse (in blue) is $\Omega(t)=\sigma \operatorname{sech}\left(\sigma\left(t-t_{g}\right)\right.$ ), and its derivative corrective solution (in orange) modulated with the splitting and the couplings is $\Omega(t)=$ $\left(\lambda_{0}+\lambda_{1}\right)^{2}(1 / \varepsilon) \frac{d}{d t} \sigma \operatorname{sech}\left(\sigma\left(t-t_{g}\right)\right)$. The bandwidth is taken to be $\sigma=0.02 \mathrm{meV}$.

$$
\begin{equation*}
F_{i}=\frac{1}{6} \sum_{j= \pm x, \pm y, \pm z} \operatorname{Tr}\left[U_{\text {ideal }} \rho_{j} U_{\text {ideal }}^{\dagger} \mathcal{U}_{i}\left(\rho_{j}\right)\right] . \tag{4.12}
\end{equation*}
$$

Here, $\rho_{j}$ 's are the six cardinal states on the Bloch sphere and $\mathcal{U}_{i}\left(\rho_{j}\right)$ is the evolution of the axial vectors under the actual evolution of the system with $i$ being either the original or DRAG solutions. We demonstrate the performance of our solutions in Figure 4.4, which shows the fidelity of $R_{X / Y}(\pi)$ and $R_{X / Y}(\pi / 2)$ gates, in terms of the dimensionless parameter $\sigma / \varepsilon$, where $\sigma$ is the bandwidth of the sech pulse and $\varepsilon$ is the splitting between the target and unwanted levels. We have considered various cases of couplings to the unwanted level: $\eta=\{\pi / 3, \pi / 4, \pi / 5\}$ in the top panel of Fig. 4.4. The bottom panel shows the required modification of the detuning. As illustrated, the strength of the unwanted couplings plays a major role in our framework.

In Fig. 4.6 we show the fidelity of the $R_{X / Y}(\pi)$ gate with respect to the ratio of weights of basis states that determine the coupling strengths. We define the gate improvement as
the ratio of the original gate error to the gate error using the corrective fields. The largest improvement (up to 2 orders of magnitude) occurs for the case of $\eta=\pi / 4$ which corresponds to equal weights of basis states $(\tan (\eta)=1)$. This can be traced back to the formation of dark and bright states; for $\lambda_{0} \neq-\lambda_{1}$ the total Hilbert space of the four levels becomes dissociated. Specifically, according to the CPT Hamiltonian of Eq. (4.1) when $\lambda_{0}=-\lambda_{1}$, we have two decoupled two-level systems; the dark-unwanted and the bright-target one. Each two level system is subject to a sech pulse driving. For a single two-level system, the population can be mapped back to the ground state at the end of the evolution if the Rabi frequency is fixed to be equal to the bandwidth of the pulse (see Section 4.5). At the end of the pulse, the target evolution generates a relative phase (rotation angle) between the two states. For the two dissociated two-level systems, it is clear that the transitionless pulse condition cannot be satisfied for both. However, approximately, each subsystem evolves with a different phase which we take into account with our detuning modification approach. At the same time, the DRAG framework verifies this behavior and suggests that no pulse modulation (at least, to the first order) is required to improve the fidelity of this special case of couplings.

### 4.4 Discussion and Conclusion

In this work we developed a framework for high-fidelity control of $\Lambda$-systems with unwanted transitions by modulating the pulse shape and slight modification of the detuning. We inferred the form these elements by performing a DRAG analysis on the CPT transformed Hamiltonian of the system, and decoupling the unwanted optical elements of the system from this frame. The pulse modulation that we presented was in the form of a corrective modification $\Omega_{\ell, c}(t)$ to the original pulse $\Omega_{\ell, o}(t)$ that drives each transition $\ell$ of the system. However, it should be noted that this solution is not unique; we have made specific choices


Figure 4.6: Gate improvement for an $X$-rotation by $\pi, R_{X}(\pi)$, using a corrective pulse $\Omega_{c}(t)$ in terms of the dimensionless parameter $(1 / 2 \pi) \sigma / \varepsilon$ and the parameter $\eta$ which indicates the ratio between the strengths of couplings to the unwanted level $\left|\lambda_{1} / \lambda_{0}\right|$ in $|t\rangle=\sin (\eta)\left|b_{1}\right\rangle-$ $\cos (\eta)\left|b_{0}\right\rangle$ and $|u\rangle=\cos (\eta)\left|b_{1}\right\rangle+\sin (\eta)\left|b_{0}\right\rangle$. The best improvements come when the value of the two couplings are close to each other, i.e., $\eta=\pi / 4$.
on the generative parameters of the DRAG frame, in deriving our DRAG solutions (see Section 4.6 for details of the derivations). These generative elements are the free parameters of the system and can be set to arbitrary values as long as they satisfy the DRAG transformation condition $S^{(1)}\left(t_{g}\right)=S^{(1)}(0)=0$. Therefore one can choose different generative elements for alternative DRAG solutions based on the system and the experimental needs. Furthermore, in the text we discussed the implementation of $X$-, and $Y$-rotations. To achieve universal control of the qubit system, we also require implementations of $Z$-rotations. This can be done through the same control scheme we discussed in this work by driving only a single transition of the $\Lambda$-system with a sech pulse [45]. Therefore our formalism applies in the same manner and to perform high-fidelity $Z$-rotations, all we need to do is to apply our pulse modulation to the single pulse that drives the single transition of $\Lambda$-system.

This pulse modification can be implemented by using a beam splitter on the original drive, modulating one part of the drive to acquire the appropriate $\pi / 2$ phase difference and shape, and the rejoining both parts to carry the effect of the correction. We have shown the pulse shapes in Fig. 4.5. The corrective pulse is inversely proportional to the splitting $\varepsilon$ and the coupling to the unwanted level for that transition (Eq. (4.7)). For smaller splittings correction pulse becomes more comparable to the original pulse. Furthermore, as opposed to the original DRAG framework that requires time-dependent detunings, our formalism require only a slight modification of a static detuning, lowering the experimental overhead of chirped detunings.

Finally we comment on the choice of basis states that constructed our target and unwanted levels that we discussed in Section 4.2. The exact same analysis presented in this work can be done in the case of a target and unwanted level which have bare couplings to the two qubit states. In that case we can set the couplings to the target state to unity and take the coupling to the unwanted to be proportional to $\lambda_{0}$ and $\lambda_{1}$ as the bare coupling; Rabi frequencies of
the target transitions are $\Omega_{0}(t) \equiv d_{0, t} E_{0}$ and $\Omega_{1}(t) \equiv d_{1, t} E_{1}$, and the Rabi frequencies of the unwanted transitions are $\lambda_{0} \Omega_{0}(t)$ and $\lambda_{1} \Omega_{1}(t)$. It can be seen that such formulation leads to the same Hamiltonian of Eq. (4.1), except that $\lambda_{0}$ and $\lambda_{1}$ are independent of each other. Therefore the presented analysis will still hold with the same set of DRAG solutions that we have found.

The version of DRAG we have tailored to case of a $\Lambda$-system with a fourth unwanted level is general and can be applied to other possible selection rules in other optically driven systems, such as color centers (e.g., the NV center in diamond), trapped ions, etc. For instance, an application of our framework is in scalable QDMs introduced in Ref. [33]. In these systems the indirect transitions of each QMD can be set to a particular frequency using local electric fields and therefore DRAG modified drives can be applied to several QDMs. However, each QDM might have a different splitting compared to other ones and the DRAG solutions we have are specific to a single $\Lambda$-system with a particular splitting. Although applying a modified drive will improve all qubits, but it will not necessarily yield to the best possible fidelity. However, generally speaking, developing a version of the presented framework for different systems with distinct characteristics could be a potential future direction.

### 4.5 Coherent population trapping

In this section we present mathematical details of the CPT framework. Two-level systems subject to a sech pulse with Rabi frequency $\Omega$ and bandwidth $\sigma$, can be solved analytically [86] and the solutions are in the form of hypergeometric functions. For the case of $\Omega / \sigma \in \mathbb{N}$, these pulses are transitionless [45], i.e., after the passage of the pulse the population will always return to the ground state with the ground state acquiring a non-trivial phase $\phi$ through the process. For the specific case of $\Omega=\sigma$, the Hamiltonian of a generic
two-level system driven by a sech pulse in the rotating frame is

$$
H=\left(\begin{array}{cc}
\delta / 2 & \Omega(t)  \tag{4.13}\\
\Omega(t) & -\delta / 2
\end{array}\right)
$$

where $\Omega(t)=\sigma \operatorname{sech}\left(\sigma\left(t-t_{g} / 2\right)\right)$ and the gate time is $t_{g}=n / \sigma$, with $n$ being a positive real number. For this system the unitary evolution operator at the end of the gate is $U=\operatorname{diag}\left(e^{-i \phi}, e^{i \phi}\right)$, where the phase is given by:

$$
\begin{equation*}
\phi=2 \arctan \left(\sigma / \delta_{o}\right)+\delta t_{g} / 2 \tag{4.14}
\end{equation*}
$$

In the CPT scheme the transitions of the system are excited using a drive field like,

$$
E_{0} f_{0}(t) e^{i \omega_{0} t}+e^{i \alpha} E_{1} f_{1}(t) e^{i \omega_{1} t}+h . c .
$$

For identical temporal envelopes and detunings, with Rabi frequencies $\Omega_{0}(t)$ and $\Omega_{1}(t)$, the transformation of the original qubit states to the bright and dark states is:

$$
\binom{|D\rangle}{|B\rangle}=\left(\begin{array}{cc}
\cos \frac{\theta}{2} & -e^{i \alpha} \sin \frac{\theta}{2}  \tag{4.15}\\
e^{-i \alpha} \sin \frac{\theta}{2} & \cos \frac{\theta}{2}
\end{array}\right)\binom{|0\rangle}{|1\rangle},
$$

where $\sin \frac{\theta}{2}=\Omega_{0} / \Omega_{\text {eff }}, \cos \frac{\theta}{2}=\Omega_{1} / \Omega_{\text {eff }}$, and $\Omega_{\text {eff }}^{2}=\Omega_{0}^{2}+\Omega_{1}^{2}$. In the CPT frame, the transition matrix elements between the target and the dark state vanishes and the bright and target state will have the matrix element defined by the effective Rabi frequency: $V_{t, B}=\Omega_{\mathrm{eff}} f(t) e^{-i \delta t}$. For the case of both drives using a sech temporal envelope, i.e., $f_{0}(t)=f_{1}(t)=\operatorname{sech}(\sigma t)$, the transitionless pulse with $\Omega_{\text {eff }}=\sigma$ will induce the relative phase $\phi$ between the bright and dark states which translates to a rotation in the sub-
space of $|D\rangle$ and $|B\rangle$. Therefore, by varying the drive parameters we can set the unitary transformation of Eq. (4.15) to transform our original qubit states to the desired states in the CPT frame, effectively enabling the rotation about an arbitrary axis of rotation $\hat{n}=(\sin \theta \cos \alpha, \sin \theta \sin \alpha, \cos \theta): R_{n}(\phi)=e^{i \phi \hat{n} . \vec{\sigma}}$.

The CPT framework can be applied to the case of $\Lambda$-system with leakage in a similar way. However, there will be additional transitions from the bright and dark state to the unwanted level. We set $\theta=\pi / 2$ and $\alpha=0$, and in the lab frame of non-ideal system with leakage, for the case of equal detunings, $\delta$, the Hamiltonian in the interaction frame after the RWA can be written as

$$
\begin{equation*}
H_{\text {int }}=\frac{1}{2} e^{i(\delta-\varepsilon) t}\left\{\lambda_{0} \Omega_{0}|u\rangle\langle 0|+\lambda_{1} \Omega_{1}|u\rangle\langle 1|\right\}+\frac{1}{2} \sum_{j=0,1} e^{i \delta t} \Omega_{j}|t\rangle\langle j|+h . c . \tag{4.16}
\end{equation*}
$$

The CPT transformation for an $X$-rotation amounts to having both Rabi freqcuencies to be equal: $\Omega_{o}(t)=\Omega_{0}(t)=\Omega_{1}(t)$. Such a CPT transformation turns this Hamiltonian into

$$
\begin{equation*}
H_{\mathrm{CPT}}=\frac{e^{i \delta t}}{\sqrt{2}} \Omega_{o}|t\rangle\langle B|+\frac{e^{i(\delta-\varepsilon) t}}{2 \sqrt{2}}\left\{\Omega_{o}\left(\lambda_{0}-\lambda_{1}\right)|u\rangle\langle D|+\Omega_{o}\left(\lambda_{0}+\lambda_{1}\right)|u\rangle\langle B|\right\}+\text { h.c. } \tag{4.17}
\end{equation*}
$$

We proceed by removing the oscillatory parts of the CPT Hamiltonian by going to a rotating frame. We do this using the frame transformation

$$
\begin{equation*}
\operatorname{diag}\left[e^{i(\delta / 2) t}, e^{i(\delta / 2) t}, e^{-i(\delta / 2) t}, e^{i(-\delta / 2+\varepsilon) t}\right] \tag{4.18}
\end{equation*}
$$

Upon this transformation we arrive at the Hamiltonian

$$
\begin{align*}
H_{\mathrm{CPT}, \omega d}= & (\delta / 2)\left(\Pi_{D}+\Pi_{B}-\Pi_{t}-\Pi_{u}\right)+\varepsilon \Pi_{u} \\
& +\frac{1}{2 \sqrt{2}}\left\{2 \Omega_{o}|t\rangle\langle B|+\Omega_{o}\left(\lambda_{0}-\lambda_{1}\right)|u\rangle\langle D|\right. \\
& \left.+\Omega_{o}\left(\lambda_{0}+\lambda_{1}\right)|u\rangle\langle B|+\text { h.c. }\right\} . \tag{4.19}
\end{align*}
$$

### 4.6 Mathematical derivation of DRAG solutions

In this section we derive the DRAG solutions presented in Eqs. (4.7). To that end we first need to expand the control fields of the DRAG frame Hamiltonian with respect to the adiabatic parameter $x$ :

$$
\begin{equation*}
H_{\mathrm{DRAG}}^{(n)}(t)=H_{\mathrm{extra}}^{(n)}(t)+\bar{H}^{(n)}(t)+i\left[S^{(n+1)}(t), \Pi_{u}\right], \tag{4.20}
\end{equation*}
$$

where $H_{\text {extra }}$ is generated by the lower orders of the transformation and is usually a nontrivial expression, and,

$$
\begin{equation*}
\bar{H}_{\mathrm{CPT}, \omega d}(t)=\frac{1}{x} \Pi_{u}+\sum_{n=0}^{\infty} x^{n} \bar{H}^{(n)}(t) . \tag{4.21}
\end{equation*}
$$

Notice that this expansion essentially means that the constrains in Eqs. (4.4) and (4.5), and consequently the control fields $h_{i}(t)$ and $h_{z}(t)$ should be made perturbative with respect to the order of this expansion as well. Furthermore, for a d-dimensional system the general form of the $S(t)$ can be written as

$$
\begin{equation*}
S(t)=\sum_{i} s_{i, z}(t) \Pi_{i}+\sum_{i=o, c} \sum_{m<n} s_{i, m, n}(t) \sigma_{m, n}^{i} \tag{4.22}
\end{equation*}
$$

The zeroth-, first- and second-order expressions are as follows [78]:

$$
\begin{align*}
H_{\mathrm{extra}}^{(0)}(t)= & 0  \tag{4.23}\\
H_{\mathrm{extra}}^{(1)}(t)= & i\left[S^{(1)}(t), H^{(0)}(t)\right]-\left[S^{(1)}(t),\left[S^{(1)}(t), \Pi_{u}\right]\right] / 2-\dot{S}^{(1)}(t) .  \tag{4.24}\\
H_{\mathrm{extra}}^{(2)}(t)= & i\left[S^{(2)}(t), H^{(0)}(t)\right]+i\left[S^{(1)}(t), H^{(1)}(t)\right]-\left[S^{(1)}(t),\left[S^{(1)}(t), H^{(0)}(t)\right]\right] / 2 \\
& -\left[S^{(1)}(t),\left[S^{(2)}(t), \Pi_{u}\right]\right] / 2-\left[S^{(2)}(t),\left[S^{(1)}(t), \Pi_{u}\right]\right] / 2 \\
& -i\left[S^{(1)}(t),\left[S^{(1)}(t),\left[S^{(1)}(t), \Pi_{u}\right]\right]\right] / 6+i\left[\dot{S}^{(1)}(t), S^{(1)}(t)\right] / 2-\dot{S}^{(2)}(t) . \tag{4.25}
\end{align*}
$$

Using Eqs. (4.20) and (4.23), we can solve for the constrains in Eqs. (4.4) and (4.5) to obtain the appropriate control elements in terms of different orders of the parameter $x$. The control constraints of (4.4) turn into

$$
\begin{align*}
\sqrt{2} \bar{\Omega}_{o}^{(n)} & =h_{o}^{(n)}-\operatorname{Tr}\left[H_{\mathrm{extra}}^{(n)}(t) \sigma_{B, t}^{o}\right] \\
\sqrt{1 / 2}\left(\bar{\Omega}_{0, c}^{(n)}+\bar{\Omega}_{1, c}^{(n)}\right) & =h_{c}^{(n)}-\operatorname{Tr}\left[H_{\mathrm{extra}}^{(n)}(t) \sigma_{B, t}^{c}\right],  \tag{4.26}\\
\frac{3}{2} \bar{\delta}^{(n)}(t) & =h_{z}^{(n)}(t)-\operatorname{Tr}\left[H_{\mathrm{extra}}^{(n)}(t)\left(\Pi_{B}-\Pi_{t}\right)\right] .
\end{align*}
$$

The decoupling constraints of (4.5) turn into

$$
\begin{align*}
s_{c, D, u}^{(n+1)} & =-\frac{1}{2} \operatorname{Tr}\left[H_{\text {extra }}^{(n)}(t) \sigma_{D, u}^{o}\right]+\frac{1}{2 \sqrt{2}}\left(\lambda_{1}-\lambda_{0}\right) \bar{\Omega}_{o}^{(n)}, \\
s_{o, D, u}^{(n+1)} & =+\frac{1}{2} \operatorname{Tr}\left[H_{\text {extra }}^{(n)}(t) \sigma_{D, u}^{c}\right]-\frac{1}{2 \sqrt{2}}\left(\lambda_{1} \bar{\Omega}_{1, c}^{(n)}-\lambda_{0} \bar{\Omega}_{0, c}^{(n)}\right), \\
s_{c, B, u}^{(n+1)} & =-\frac{1}{2} \operatorname{Tr}\left[H_{\text {extra }}^{(n)}(t) \sigma_{B, u}^{o}\right]-\frac{1}{2 \sqrt{2}}\left(\lambda_{1}+\lambda_{0}\right) \bar{\Omega}_{o}^{(n)}, \\
s_{o, B, u}^{(n+1)} & =+\frac{1}{2} \operatorname{Tr}\left[H_{\text {extra }}^{(n)}(t) \sigma_{B, u}^{c}\right]-\frac{1}{2 \sqrt{2}}\left(\lambda_{1} \bar{\Omega}_{1, c}^{(n)}+\lambda_{0} \bar{\Omega}_{0, c}^{(n)}\right), \\
s_{c, t, u}^{(n+1)} & =-\frac{1}{2} \operatorname{Tr}\left[H_{\text {extra }}^{(n)}(t) \sigma_{t, u}^{o}\right], \\
s_{o, t, u}^{(n+1)} & =+\frac{1}{2} \operatorname{Tr}\left[H_{\text {extra }}^{(n)}(t) \sigma_{t, u}^{c}\right] . \tag{4.27}
\end{align*}
$$

From these constraints, first we set the zero-order solutions. In order to make the CPT framework into the desired form within the bright and target subspace, from Eqs. (4.26) with $n=0$, we find that we need to set $h_{i}^{(0)}(t)=2 t_{g} \Omega(t)$ where $\Omega(t)=\sigma \operatorname{sech}(\sigma t), h_{i, c}^{(0)}(t)=0$ for $i=0,1$, and $h_{z}^{(0)}(t)=(3 / 2) t_{g} \delta$, and as before we set the higher orders to zero. Using the fact that $H_{\text {extra }}^{(0)}(t)=0$, we find

$$
\begin{equation*}
\Omega_{o}(t)=\sqrt{2} \Omega(t), \quad \Omega_{0, c}(t)=\Omega_{1, c}(t)=0 \tag{4.28}
\end{equation*}
$$

The only nonzero first-order generator elements from Eqs. (4.27) are $s_{c, D, u}^{(1)}=\frac{1}{2}\left(\lambda_{0}-\right.$ $\left.\lambda_{1}\right) t_{g} \Omega(t)$, and $\quad s_{c, B, u}^{(1)}=\frac{1}{2}\left(\lambda_{0}+\lambda_{1}\right) t_{g} \Omega(t)$. These two constraints assure prevention of leakage from dark and bright states to the unwanted level, respectively. Using these results along with $H_{\text {extra }}^{(1)}=0$ will provide us with the first-order correction equations:

$$
\begin{align*}
\sqrt{2} \bar{\Omega}_{o}^{(1)} & =2 \dot{s}_{o, B, t}^{(1)}+2 \delta s_{c, B, t}^{(1)},  \tag{4.29}\\
\frac{1}{\sqrt{2}}\left(\bar{\Omega}_{0, c}^{(1)}+\bar{\Omega}_{1, c}^{(1)}\right) & =2\left(\dot{s}_{c, B, t}^{(1)}+\Omega t_{g}\left(s_{z, D}^{(1)}-s_{z, B}^{(1)}\right)-\delta s_{o, B, t}^{(1)}\right), \\
s_{c, B, t}^{(1)} & =\frac{\Omega t_{g}}{16 \sqrt{2}}\left(\lambda_{0}+\lambda_{1}\right)^{2}+\dot{s}_{z, D}^{(1)}-\dot{s}_{z, B}^{(1)} .
\end{align*}
$$

In practice one may find the corrective measures by just using the first-order constraint $S^{(1)}(0)=S^{(1)}\left(t_{g}\right)=0$, however, as we show in the following, although the higher orders will satisfy the decoupling constraints, but the dark-to-target and dark-to-bright constraints cannot be satisfied simultaneously. Nevertheless, we find the corrections to the original pulse as follows. To seek the simplest solution which satisfies the $S^{(1)}(0)=S^{(1)}\left(t_{g}\right)=0$, we pick the free parameters $s_{z, i}^{(1)}$, $s$ and $s_{o, B, t}^{(1)}$ all equal to zero. For two identical corrective drive elements, i.e. $\bar{\Omega}_{0, c}^{(1)}=\bar{\Omega}_{1, c}^{(1)}=\bar{\Omega}_{c}^{(1)}$, by using the third equation in the first two, we find the pulses given in Eqs. (4.7). Using these solutions and the choices we made above for the generative elements of $S(t)$, in the expansion (4.20) (notice that we will need to utilize $H_{\text {extra }}^{(2)}(t)$ since we will need to set the values of $\left.S^{(2)}\right)$ will lead to the first-order form of the DRAG Hamiltonian $H_{\text {DRAG }}^{(1)}(t)$ given in Eq. (4.8).

## Chapter 5

## Nuclear spin problem in quantum <br> dots

The trapped electron in a QD interacts with a bath of $\sim 10^{5}$ nuclear spins [2] through the contact hyperfine (HF) interaction. So far for the scalable QDM systems and generation of cluster states from these systems, we have considered hole spin qubits. As we discussed in Chapter 2, hole spins have $p$-like orbitals with dumbbell shapes. The HF contact interaction vanishes due to the node in the $p$-like orbitals of the holes. But for electron spins with $s$-like orbitals, the HF interaction is quite important. Furthermore, as we discussed in Section 2.1, inhomogeneous electric field gradients are present in QDs due to the strain-driven growth process. These electric field gradients couple to the quadrupolar moment of nuclear spins and affect the dynamics of the system.

This chapter is dedicated to the study of these effects in QDs. The work presented in this chapter is based on "Driven dynamics of a quantum dot electron spin coupled to a bath of higher-spin nuclei" by Arian Vezvaee, Girish Sharma, Sophia E. Economou, and Edwin Barnes [87].

### 5.1 The nuclear spin problem

As we discussed in previous chapters, spins trapped in QDs are under intense investigation for a variety of quantum information applications, including quantum information processing, quantum communication, and quantum transduction [9, 88, 89, 90]. The relatively long coherence times, fast controllability [45, 91, 92], and good photon emission properties of these systems $[1,36,93,94]$ make them promising candidates for achieving high-quality spinphoton interfaces and for producing large-scale multi-photon entangled states [68, 95, 96, 97].

While optically controlled quantum dot spins offer a wide range of technological possibilities, HF interactions between the confined spin and its surrounding nuclear spin bath have been a major impediment. This interaction is the main source of decoherence in these systems, and it also causes spectral wandering and inhomogeneities in quantum dot ensembles, aspects that have been researched extensively over the past two decades $[2,98,98,99,100,100,101$, $102,103,104,105,106,107,108,109,110,111,112,113,114,115,116,117,118,119,120$, 121, 122, 123, 124, 125, 126, 127, 128, 129, 130, 131]. However, many works have shown that the state of the bath, and consequently its deleterious effects, can be influenced by driving the electron spin. For example, several experiments have shown that driving can generate dynamic nuclear polarization (DNP), an effect that has been observed in self-assembled QDs $[29,132,133,134,135,136,137,138,139,140,141,142,143]$ and also in other systems such as gated QDs [144, 145, 146, 147], quantum wires [148] and in bulk materials [149], findings that have been supported by a number of theory works [150, 151, 152, 153, 154, $155,156,157,158,159,160,161,162]$. In self-assembled QDs, it has been shown that DNP can survive on the order of minutes due to the suppression of nuclear spin diffusion caused by strain-induced quadrupolar interactions [163, 164, 165]. An important example of DNP in self-assembled QDs is the mode-locking experiments of Refs. [132, 133, 134, 135, 136, 137, 138], where an ensemble of QD electron spins becomes synchronized with a periodic train
of optical pulses as a consequence of DNP. Continuous-wave laser driving of the electron has been shown to create DNP in QDs as well, leading to interesting phenomena such as the line-dragging effect, i.e. the locking of an optical QD transition to the frequency of the laser [29, 141, 142, 154, 166]. Owing to the long coherence times of nuclear spins, DNP has been proposed for applications such as quantum memories [167, 168], which has recently been demonstrated experimentally [169, 170].

Although most of the fully quantum mechanical theoretical treatments of the HF decoherence problem allow for nuclei with spin greater than $1 / 2[102,103,104]$, studies of the driven, HF-induced generation of DNP have mostly focused on spin $1 / 2$ nuclei to reduce the computational complexity of the problem [133, 150, 151, 159, 160, 171]. The latter works typically rely on either stochastic equations or rate equations to solve for the nuclear polarization distribution. While solving the feedback problem for spin $1 / 2$ nuclear baths can yield qualitative insights about DNP experiments, the quantitative accuracy of such models is limited by the fact that the most commonly studied semiconductor QDs are in materials such as InAs or GaAs, which contain nuclei of $\operatorname{spin} I>1 / 2$. In addition to artificially reducing the size of the bath Hilbert space, assuming spin $1 / 2$ nuclei also ignores effects such as quadrupolar interactions, which are only present for $I>1 / 2$. There do exist a few theoretical works that allow for $I>1 / 2[98,117,152]$. Specifically, Huang and Hu [98] studied DNP arising from hyperfine interactions with the spin $3 / 2$ arsenic nuclei in InGaAs by making use of Fermi's golden rule; however, only qualitative agreement with experiment was achieved due to the need to introduce phenomenological parameters. Yang and Sham [152] presented a general framework for nuclei of arbitrary total spin by unifying the stochastic and rate-equation approaches. In this work they focused on a drift feedback loop (which allows for a possible bias in nuclear spin-flip processes) and obtained a Fokker-Planck equation for the polarization of the bath. Although this framework captures line-dragging and other DNP phenomena seen in
experiments, it has only been established for continuous-wave driving, and so it is not immediately applicable to experiments with periodic driving such as the mode-locking experiments of Refs. [132, 133, 134, 135, 136, 137, 138]. Theoretical works that have specifically focused on mode-locking type experiments have either assumed $I=1 / 2$ nuclear baths $[150,151]$ or utilized semiclassical methods $[172,173,174,175,176,177,178]$. While semiclassical approaches have been successful in reproducing qualitative features seen in experiments including dynamic nuclear polarization and mode-locking, it remains an outstanding challenge to develop a more quantitatively accurate description of the driven electron-nuclear spin system. Unlike in the case of nuclear-spin-induced decoherence, where semiclassical treatments have been shown to agree well with quantum mechanical ones [157, 179, 180, 181], similar comparisons in the context of optically driven DNP have revealed significant quantitative differences [173].

In this chapter, we develop a quantum, non-perturbative framework to solve the dynamics of an optically driven electron spin coupled to a bath of $I>1 / 2$ nuclear spins. We focus on DNP feedback mechanisms that arise from driving the electron with a periodic train of optical pulses while it is subject to hyperfine interactions with a nuclear spin bath, as in the mode-locking experiments $[132,133,134,135,136,137,138]$. Here, we also consider the effect of quadrupolar interactions. To compute DNP and its effect on the evolution of the electron spin, we use an approach based on dynamical maps and kinetic equations introduced in Refs. [150, 151], but, importantly, here we generalize the formalism to higher nuclear spin and treat the problem non-perturbatively, unlike in these earlier works. Our framework provides a self-consistent description of the feedback loop between the driven electron and DNP.

We compute the nuclear spin polarization distribution and its influence on the electron spin evolution for spin 1 and spin $3 / 2$ baths and compare the results to the $I=1 / 2$ case. Our
approach is able to treat bath sizes of up to thousands of nuclear spins in the $I=1 / 2$ and $I=1$ cases, and up to several hundred spins in the $I=3 / 2$ case. Although evidence of mode-locking is seen in all three cases, we find that quadrupolar interactions act to suppress mode-locking for $I>1 / 2$, especially when the angle between the principal strain axis and the applied magnetic field is large. We also find that while HF interactions can produce a significant bath polarization that grows linearly with the number of nuclei for $I>1 / 2$, quadrupolar interactions work to counteract this buildup of DNP. We further show that the relative importance of quadrupolar effects grows as the magnitude of the applied magnetic field is increased. The competition between HF and quadrupolar interactions imprints clear signatures in the steady-state electron spin evolution, providing an experimental tool to measure the strength of quadrupolar couplings in a QD. Our results show that accounting for higher nuclear spin is important not only for quantitative accuracy, but also for capturing important qualitative features of the DNP process in driven QD systems.

The following sections are structured as follows. In Sec. 5.2, we describe the system and Hamiltonian. In Sec. 5.3, we lay out the theoretical approach in detail for arbitrary nuclear spin $I$ and construct the equations that govern DNP for $I=1 / 2,1$, and $3 / 2$ nuclear spin baths. We present an analytical solution for the steady-state nuclear spin polarization distribution for $I=1 / 2$. In Sec. 5.4, we numerically compute steady-state polarization distributions for $I=1$ and $3 / 2$ and compare the results to the $I=1 / 2$ solution for various parameter choices. We also study the effect of DNP on the electron spin evolution. We conclude in Sec. 5.5.


Figure 5.1: The relevant level structure in the mode-locking experiments. $|x\rangle$ and $|\bar{x}\rangle$ are the electron spin states along the optical axis. These states are coupled by an external magnetic field along the $z$ direction. Circularly polarized light excites the ground electron spin states to excited trion levels $|T\rangle$ and $|\bar{T}\rangle$ with angular momentum projections $+3 / 2$ and $-3 / 2$, respectively. The selection rules are such that each ground state couples to only one excited state. The trion states decay via spontaneous emission with rate $\gamma_{e}$. In this work, we focus on left-circularly polarized driving.

### 5.2 System and Hamiltonian

Our focus in this work is on QD experiments in which a single electron is periodically pumped by a train of optical pulses [132, 133, 134, 135, 136, 137, 138, 182]. Each pulse excites the electron to a trion state (a bound state of an electron and an exciton), which then decays back to the electronic ground state manifold via spontaneous emission. The full Hamiltonian of the nuclear spin bath and the driven electron is given by

$$
\begin{equation*}
H(t)=H_{0, e}+H_{0, n}+H_{c}(t)+H_{r e s}+H_{H F}+H_{Q} . \tag{5.1}
\end{equation*}
$$

Here, $H_{0, e}$ describes the electronic degrees of freedom in the QD in the absence of driving:

$$
\begin{equation*}
H_{e, 0}=\omega_{e} \hat{S}_{z}+\omega_{\bar{T}}|\bar{T}\rangle\langle\bar{T}|, \tag{5.2}
\end{equation*}
$$

where $\omega_{e}$ is the electron spin Zeeman frequency, $\hat{S}_{z}$ is the spin operator in the electronic ground space, and $\omega_{\bar{T}}$ is the energy of the trion state $|\bar{T}\rangle$. We take the magnetic field to be oriented along the $z$ direction, while the optical axis lies in the $x$ direction (see Fig. 5.1). We neglect the second trion level $|T\rangle$ in $H_{0, e}$ because it is not excited by the laser polarization we are considering. This driving is described by the Hamiltonian

$$
\begin{equation*}
H_{c}(t)=\Omega(t)|\bar{x}\rangle\langle\bar{T}|+h . c . \tag{5.3}
\end{equation*}
$$

where we assume the drive laser is left-circularly polarized (red arrow in Fig. 5.1) with periodic temporal profile $\Omega\left(t+T_{R}\right)=\Omega(t)$, so that each pulse couples the electron spin state $|\bar{x}\rangle$ to the trion state $|\bar{T}\rangle$. The latter decays via spontaneous emission with rate $\gamma_{e}$. This process arises from interactions with a photonic bath, which is represented by the term $H_{\text {res }}$. We do not give an explicit expression for this term as it is not needed in what follows. The Zeeman splitting of the nuclear spins is given by $H_{0, n}=\omega_{n} \sum_{i} \hat{I}_{z}^{i}$.

The HF interaction is given by the contact term:

$$
\begin{equation*}
H_{H F}=\sum_{i=1}^{N} A_{i} \hat{S}_{z} \hat{I}_{z}^{i}+\sum_{i=1}^{N} A_{i} / 2\left(\hat{S}_{+} \hat{I}_{-}^{i}+\hat{S}_{-} \hat{I}_{+}^{i}\right) \tag{5.4}
\end{equation*}
$$

where $N$ is the number of nuclei that interact appreciably with the electron. The first term is referred to as the Overhauser term, and it gives rise to an effective magnetic field seen by the electron spin in the case of nonzero nuclear spin polarization. The second term generates flip-flop interactions under which the electron spin flips with a nuclear spin. These terms are responsible for transferring angular momentum from the electron onto the nuclei, while the Overhauser term is the primary mechanism for feedback between the nuclear spin polarization and the electron spin evolution. The HF couplings $A_{i}$ are determined by the magnitude of the electronic wavefunction at the location of the nuclear spin $I^{i}$. However,
on timescales short compared to $N / \mathcal{A} \sim \mu \mathrm{s}$, where $\mathcal{A}$ is the total HF interaction energy, the variations in these couplings do not significantly affect the electron spin evolution [183]. Here, we focus on fast optical driving where the electron reaches a steady state over a timescale of about 100 ns [151], which allows us to make the 'box model' approximation in which all the HF couplings are taken equal: $A_{i}=A \equiv \mathcal{A} / N[183,184]$. Further comments about this approximation are given in Sec. 5.5.

The quadrupolar interaction is given by [185, 186]

$$
\begin{equation*}
H_{Q}=\sum_{i=1}^{N} \frac{\nu_{Q}^{i}}{2}\left(\hat{I}_{z^{\prime}}^{i}{ }^{2}-\frac{I(I+1)}{3}\right) \tag{5.5}
\end{equation*}
$$

This interaction occurs due to the coupling of the nuclear quadrupole moment to electric field gradients caused by strain in the semiconductor lattice, and it is only present for $I>1 / 2$. The presence of quadrupolar interactions has lead to striking phenomena in various types of experiments conducted in QDs. A few examples include the anomalous Hanle effect [123] and suppression of spin diffusion [121]. Line-dragging phenomena have also been associated with the presence of quadrupolar interactions [141, 142, 152]. The coupling strength $\nu_{Q}$ is referred to as the nuclear quadrupole resonance frequency, which is estimated to be around 2.8 MHz for $\mathrm{As}[121]$. The quadrupole resonance frequency generally depends on the local strain in the vicinity of each nuclear spin, and so it generally varies across the material. Here, we assume that the strain remains roughly constant over the QD, and so we take all the frequencies to be equal: $\nu_{Q}^{i}=\nu_{Q}$. The operator $\hat{I}_{z^{\prime}}$ in Eq. (5.5) is the component of the nuclear spin operator along the principal axis of the electric field gradient. Our focus will be on the case of QDs with cylindrical symmetry in which the electric field gradient makes an angle $\theta$ with the magnetic field. Therefore, we have $\hat{I}_{z^{\prime}}=\hat{I}_{z} \cos \theta+\hat{I}_{x} \sin \theta$, which then
gives [185]:

$$
\begin{equation*}
H_{Q}=\frac{\nu_{Q}}{2} \sum_{i=1}^{N}\left[\left(\hat{I}_{z}^{i}\right)^{2} \cos ^{2} \theta-\frac{I(I+1)}{3}+\left(\hat{I}_{z}^{i} \hat{I}_{x}^{i}+\hat{I}_{x}^{i} \hat{I}_{z}^{i}\right) \sin \theta \cos \theta+\left(\hat{I}_{x}^{i}\right)^{2} \sin ^{2} \theta\right] \tag{5.6}
\end{equation*}
$$

When $\theta=0, H_{Q}$ creates non-uniform energy spacings between the nuclear spin levels. For $\theta \neq 0, H_{Q}$ has the additional effect of driving $\Delta m_{I}= \pm 1$ and $\Delta m_{I}= \pm 2$ nuclear spin-flip transitions, where $m_{I}$ is the eigenvalue of $\hat{I}_{z}$. Notice that the rate for $\Delta m_{I}= \pm 1$ transitions is maximal at $\theta=\pi / 4$, while the rate for $\Delta m_{I}= \pm 2$ transitions is largest for $\theta=\pi / 2$, which is also the value of $\theta$ where the non-uniformity in the energy level spacings is zero. Thus, we see that the role of $H_{Q}$ changes as $\theta$ varies from 0 to $\pi / 4$, and from $\pi / 4$ to $\pi / 2$. Because $H_{Q}$ is $\pi$-periodic in $\theta$, it suffices to focus on the range $0 \leq \theta \leq \pi / 2$.

In the case of $I=1 / 2$ nuclei, the underlying physical mechanism behind the formation of DNP can be understood as follows. Imagine that the electron spin starts in a pure (polarized) state and the nuclear spins are in a totally mixed (unpolarized) state. The HF interaction then transfers angular momentum from the electron onto the nuclei, creating DNP. In the absence of driving, this would lead to only a modest nuclear spin polarization, and this polarization would be short-lived because it would eventually be transferred back to the electron via the HF interaction. However, the laser pulses periodically reset the electron spin to a polarized state, enabling a net transfer of angular momentum from the laser, through the electron, and onto the nuclei. This basic mechanism can also underlie DNP in nuclear spin baths with $I>1 / 2$, however it is unclear what role the quadrupolar interactions play in this story. Answering this question is a main goal of this work.

It is worth noting that our Hamiltonian, Eq. (5.1), does not include inter-nuclear dipolar interactions. In self-assembled QDs, these interactions are weak compared to the hyperfine and quadrupolar interactions, and their main effect is to drive nuclear spin diffusion, which
gradually causes the decay of DNP. It has been shown experimentally, however, that this diffusion process is strongly suppressed in self-assembled QDs due to strain [163, 164, 165], leading to diffusion times in excess of several minutes. This is longer than the timescale for generating DNP ( $\sim$ seconds [133]). For this reason, we neglect nuclear dipolar interactions and diffusion in this work.

### 5.3 Nuclear spin-flip rates and kinetic equation

Before we describe our approach in detail, we first give an overview of the general strategy and main ingredients. Our framework is summarized in Fig. 5.2. The overall strategy is similar to that introduced in Refs. [150, 151]. However, significant modifications are needed to allow for higher nuclear spin. Also, here we present a non-perturbative approach, whereas Refs. $[150,151]$ relied on perturbation theory. Therefore, the theoretical model presented in this section has overlap with, but supersedes, that of Refs. [150, 151]. Readers who are only interested in the results and not the approach could skip ahead to Sec. 5.4.

We are dealing with a system that is both open and driven. An efficient way to treat nonunitary evolution is to use dynamical maps [150, 151, 187, 188, 189]. In this approach, the non-unitary evolution of a system from an initial state $\rho$ to a final state $\rho^{\prime}$ is implemented by applying a set of operators and summing the results:

$$
\begin{equation*}
\rho^{\prime}=\sum_{k} E_{k} \rho E_{k}^{\dagger} . \tag{5.7}
\end{equation*}
$$

The operators $E_{k}$ are known as Kraus operators, and they constitute a generalization of the usual unitary operators that evolve closed quantum systems to the case of non-unitary evolution in open systems. The condition $\sum_{k} E_{k}^{\dagger} E_{k}=\mathbb{1}$ ensures that the trace of the


Figure 5.2: Schematic depiction of the self-consistent formalism we use to model DNP with feedback. We exploit a hierarchy of timescales to first solve for the joint evolution of the electron coupled to a single nuclear spin. Under a Markovian approximation, the electron spin state is reset after each drive period. The resulting nuclear spin evolution yields nuclear spin-flip rates that are then fed into a kinetic equation governing the dynamics of the multinuclear spin polarization distribution. The flip rates depend on the effective electron spin precession frequency, including the Overhauser field contribution for self-consistency. The solution to the kinetic equation is then used to update the electron steady state, closing the feedback loop.
density matrix is always unity. The advantage of Kraus operators is that they allow one to incorporate effects due to the transient occupation of excited states using operators that live purely in the ground space of the system. In the present problem, we use these operators to describe the effect of each optical pulse on the electron spin state. The entire process of optical excitation, subsequent decay, and rotation is captured by an appropriate set of Kraus operators (given in Section 5.6) without having to explicitly include excited states or a photonic bath into the formalism. The dynamical map description works well so long as the population returns regularly to the electron spin ground states, as is the case for the periodic driving used in the mode-locking experiments. These Kraus operators can then be used to obtain the electron spin steady state in the absence of nuclei, as shown in Section 5.7.

Of course, we are interested in the case where the electron spin is coupled to a nuclear spin bath through HF interactions while it is being driven. Under the condition that the electron is being pumped fast enough (which indeed is the case for the mode-locking experiments [132, $133,134,135,136,137,138]$ ), the electron reaches its steady state on a much faster timescale compared to the electron-nuclear interaction dynamics and the electron spin decoherence time. This allows us to use a Markovian approximation in which we first solve for the driven electron steady state and then incorporate the effects due to the electron-nuclear couplings on top of this.

To bring the nuclei into the framework, we first solve for the joint evolution of one nuclear spin coupled to the driven electron spin. Although the HF interaction generates unitary dynamics, this is disrupted periodically by the pulses, and this in turn leads to an effective non-unitary dynamical map for the nuclear spin that depends on the electron steady state under the Markovian approximation. We extract nuclear spin-flip rates from this effective nuclear spin evolution operator; these rates provide information about the movement of population between the different nuclear spin levels.

We calculate the steady state of the entire nuclear spin bath using a rate equation that depends on the spin-flip rates obtained from the single-nucleus solution. A critical step is that we build in self-consistent system-environment feedback by modifying the flip rates. To understand this, we first need to describe the Overhauser effect [190], which is the main feedback mechanism between the electron and nuclei. A polarized nuclear spin bath acts as an effective magnetic field and therefore shifts the Zeeman frequency of the electron. However, the interaction between the electron and the nuclear spin bath is reciprocal; not only will the state of the electron change under the Overhauser field, but the nuclear spins will also be affected by the Knight field [191], i.e., the effective magnetic field due to polarization of the electron. The Knight field is given by the electron steady state spin vector, and so it enters into the nuclear spin flip rates, as explained above. The electron steady state (and hence the Knight field) in turn depends on the total magnetic field, which includes the Overhauser field due to nuclear polarization. These interdependencies constitute a complete feedback loop that must be treated self-consistently. We do this by making the nuclear spin-flip rates depend on the net nuclear polarization of the bath. The steady-state of the rate equation then gives the polarization distribution of the nuclear spin bath with feedback included. Finally, we use this nuclear polarization distribution to perform the Overhauser shift on the Zeeman frequency of the electron and update the nuclear-bath-averaged electron spin steady-state self-consistently.

The framework we have just outlined can be thought of as a self-consistent dynamical meanfield approach. In the remainder of this section, we use this approach to compute the dynamical map for a single nuclear spin as well as the nuclear spin flip rates. We then construct the kinetic equations that govern the dynamics of the full nuclear spin bath. Our method is quite general and can be applied to baths of any nuclear spin. Here, we focus on the cases $I=1 / 2,1$, and $3 / 2$.

### 5.3.1 Effective dynamical map for one nuclear spin

Given the electron spin steady state (Eq. (5.49)), we can proceed to construct an effective dynamical map for a single nuclear spin. We do this by first constructing the evolution operator in the spin vector (SV) representation that describes the joint evolution of the electron and nuclear spins over one driving period. We then apply the Markovian approximation and reset the electron spin to its steady state at the end of the period. Tracing out the electron then leaves an effective dynamical map for the nuclear spin.

To start, we must choose a basis of Hermitian matrices $\hat{\lambda}_{k}$ of dimension $2 I+1$, where $k=1, \ldots,(2 I+1)^{2}$, in order to define the nuclear SV. Unlike in the spin $I=1 / 2$ case considered in Refs. [150, 151], for $I>1 / 2$ we have much more freedom in how to choose this basis, and the choice we make can have a substantial impact on the complexity of the analysis that follows. We choose the first $2 I+1$ of these matrices to be diagonal, each with a single nonzero component equal to one. The remaining $2 I(2 I+1)$ matrices each have two nonzero components, and these matrices are purely real or purely imaginary. For example, in the case of $I=3 / 2$, we have 16 basis matrices:

$$
\begin{array}{ll}
\hat{\lambda}_{k, a b}=\delta_{a k} \delta_{b k}, \quad k=1 \ldots 4, \\
\hat{\lambda}_{5, a b}=\frac{1}{\sqrt{2}}\left(\delta_{a 1} \delta_{b 2}+\delta_{a 2} \delta_{b 1}\right), & \hat{\lambda}_{6, a b}=\frac{-i}{\sqrt{2}}\left(\delta_{a 1} \delta_{b 2}-\delta_{a 2} \delta_{b 1}\right), \\
\hat{\lambda}_{8, a b}=\frac{-i}{\sqrt{2}}\left(\delta_{a 1} \delta_{b 3}-\delta_{a 3} \delta_{b 1}\right), \quad \hat{\lambda}_{9, a b}=\frac{1}{\sqrt{2}}\left(\delta_{a 1} \delta_{b 4}+\delta_{a 4} \delta_{b 1}\right), \quad \hat{\lambda}_{10, a b}=\frac{-i}{\sqrt{2}}\left(\delta_{a 1} \delta_{b 3} \delta_{b 4}-\delta_{a 3} \delta_{b 1}\right), \\
\left.\hat{\lambda}_{11, a b} \delta_{b 1}\right), \\
\hat{\lambda}_{14, a b}=\frac{1}{\sqrt{2}}\left(\delta_{a 2} \delta_{b 3}+\delta_{a 3} \delta_{b 2}\right), \quad \hat{\lambda}_{12, a b}=\frac{-i}{\sqrt{2}}\left(\delta_{a 2} \delta_{b 3}-\delta_{a 3} \delta_{b 2}\right), \quad \hat{\lambda}_{13, a b}=\frac{1}{\sqrt{2}}\left(\delta_{a 2} \delta_{b 4}\right), \quad \hat{\lambda}_{15, a b}=\frac{1}{\sqrt{2}}\left(\delta_{a 3} \delta_{b 4}+\delta_{a 4}\right),  \tag{5.8}\\
\left.\delta_{b 3}\right), & \hat{\lambda}_{16, a b}=\frac{-i}{\sqrt{2}}\left(\delta_{a 3} \delta_{b 4}-\delta_{a 4} \delta_{b 3}\right) .
\end{array}
$$

These matrices are normalized such that $\operatorname{Tr}\left[\hat{\lambda}_{j} \hat{\lambda}_{k}\right]=\delta_{j k}$. Denoting the nuclear spin density
matrix as $\rho_{n}$, the components of the nuclear SV $\mathcal{S}_{n}$ are then given by

$$
\begin{equation*}
\mathcal{S}_{n, k}=\operatorname{Tr}\left[\rho_{n} \lambda_{k}\right] . \tag{5.9}
\end{equation*}
$$

Note that the populations, $\rho_{n, i i}$, are the first four components of $\mathcal{S}_{n}$. We will see that this feature simplifies the process of computing flip rates.

Let us denote the density matrix that describes the total electron-nuclear spin state at the beginning of a driving period by $\varrho$. We expand this in terms of an operator basis formed from tensor products of the nuclear spin operators $\hat{\lambda}_{k}$ with the electron spin Pauli matrices $\hat{\sigma}_{j}:$

$$
\begin{equation*}
\hat{G}_{(2 I+1)^{2} j+k}=\hat{\sigma}_{j} \otimes \hat{\lambda}_{k}, \tag{5.10}
\end{equation*}
$$

with $j=0, . ., 3, k=1, \ldots,(2 I+1)^{2}$, and where we define $\hat{\sigma}_{0}=\mathbb{1}_{2 \times 2}$. We use this set of $4(2 I+1)^{2}$ operators as a basis for the SV of the joint system: $\mathcal{S}_{\ell}=\operatorname{Tr}\left(\varrho \hat{G}_{\ell}\right)$. This SV evolves over one driving period according to $\mathcal{S}^{\prime}=\mathcal{Y} \mathcal{S}$, where the SV evolution operator $\mathcal{Y}$ is given by

$$
\begin{equation*}
\mathcal{Y}_{\ell \ell^{\prime}}=\frac{1}{2} \operatorname{Tr}\left[\hat{G}_{\ell} \mathcal{U} \hat{G}_{\ell^{\prime}} \mathcal{U}^{\dagger}\right] \tag{5.11}
\end{equation*}
$$

where $\mathcal{U}=\exp \left\{-i\left(\omega_{e} \hat{S}_{z}+\omega_{n} \hat{I}_{z}+H_{H F}^{N=1}+H_{Q}^{N=1}\right) T_{R}\right\}$ describes the joint evolution of the electron spin and single nuclear spin under precession and the HF and quadrupolar interactions. At this point, we invoke the Markovian approximation: Because the electron reaches its steady state, $\mathcal{S}_{e}^{s s}$, quickly compared to the timescales for nuclear spin and HF dynamics, we reset the electron SV to its steady state value at the beginning/end of each period: $\mathcal{S}=\mathcal{S}_{e}^{s s} \otimes \mathcal{S}_{n}$. We then obtain an effective nuclear spin dynamical map, $\mathcal{Y}_{n}$, by acting with the full evolution operator, $\mathcal{Y}$, on the tensor product $\mathcal{S}_{e}^{s s} \otimes \mathcal{S}_{n}$ and reading off the coefficients
of the components of the nuclear $\mathrm{SV}, \mathcal{S}_{n}$, from the resulting $\mathcal{S}^{\prime}$ :

$$
\begin{equation*}
\mathcal{Y}_{n, j k}=\frac{d}{d \mathcal{S}_{n, k}}\left[\mathcal{Y}\left(\mathcal{S}_{e}^{s s} \otimes \mathcal{S}_{n}\right)\right]_{j} \tag{5.12}
\end{equation*}
$$

Here, $j, k=1, \ldots,(2 I+1)^{2}$, that is, we only retain the components of $\mathcal{S}^{\prime}$ that correspond to the basis operators $\hat{G}_{k}=\mathbb{1}_{2 \times 2} \otimes \hat{\lambda}_{k}$, i.e., the components that correspond to purely nuclear spin degrees of freedom. Note that although the joint evolution operator $\mathcal{Y}$ describes unitary evolution, the nuclear spin dynamical map, $\mathcal{Y}_{n}$, implements non-unitary evolution. This non-unitarity is a consequence of the Markovian approximation, which is itself due to the non-unitary driving of the electron spin.

### 5.3.2 Single-nucleus flip rates

We can use the nuclear spin dynamical map, $\mathcal{Y}_{n}$, that we found in the previous subsection to find the flip rates for a single nuclear spin interacting with the electron spin. These flip rates govern the movement of population from one nuclear spin state to another. Such processes are described by the following kinetic equation:

$$
\begin{equation*}
\frac{d p_{m}}{d t}=\sum_{n \neq m} \mathrm{w}_{n}^{m} p_{n}-\sum_{n \neq m} \mathrm{w}_{m}^{n} p_{m} \tag{5.13}
\end{equation*}
$$

where $p_{m}$ is the population of level $m$, and $\mathrm{w}_{n}^{m}$ is the rate to flip from state $n$ to $m$, which in general differs from the rate to flip from $m$ to $n, \mathrm{w}_{m}^{n}$. Which transitions are allowed depends on the type of interactions present in the Hamiltonian. For instance, the HF flipflop terms only cause $\Delta m_{I}= \pm 1$ transitions, while the quadrupolar interaction also drives $\Delta m_{I}= \pm 2$ transitions. We can combine the rate equations (5.13) into a matrix equation. We exemplify this in the $I=3 / 2$ case, where we denote the four states $|+3 / 2\rangle,|+1 / 2\rangle$,


Figure 5.3: Single-nucleus spin-flip rates for (a,b) $I=1 / 2$, (c,d) $I=1$, (e,f) $I=3 / 2$ as a function of the magnetization $m$ of the nuclear spin bath. Flip rates are shown in (a), (c), (e), while flip rate differences are shown in (b), (d), (f). The parameter values are $T_{R}=13.2 \mathrm{~ns}, N A=10 \mathrm{GHz}, N=1000, \omega_{e 0}=0.5 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}$, $q_{0}=0.3, \phi=-\pi / 2$. For (c-f), we set the quadrupolar parameters to $\nu_{Q}=2.8 \mathrm{MHz}$ and $\theta=0$. Only the nonzero flip rates are shown.
$|-1 / 2\rangle,|-3 / 2\rangle$, by the shorthand $\{++,+,-,--\}$. The matrix equation is then $\dot{\mathcal{P}}=\mathcal{M} \mathcal{P}$, where $\mathcal{P}=\left(p_{++}, p_{+}, p_{-}, p_{--}\right)$, and
$\mathcal{M}=\left[\begin{array}{cccc}-\left(\mathrm{w}_{++}^{+}+\mathrm{w}_{++}^{-}+\mathrm{w}_{++}^{--}\right) & \mathrm{w}_{+}^{++} & \mathrm{w}_{-}^{++} & \mathrm{w}_{--}^{++} \\ \mathrm{w}_{++}^{+} & -\left(\mathrm{w}_{+}^{++}+\mathrm{w}_{+}^{-}+\mathrm{w}_{+}^{--}\right) & \mathrm{w}_{-}^{+} & \mathrm{w}_{--}^{+} \\ \mathrm{w}_{++}^{-} & \mathrm{w}_{+}^{-} & -\left(\mathrm{w}_{-}^{++}+\mathrm{w}_{-}^{+}+\mathrm{w}_{-}^{--}\right) & \mathrm{w}_{--}^{-} \\ \mathrm{w}_{++}^{--} & \mathrm{w}_{+}^{--} & \mathrm{w}_{-}^{--} & -\left(\mathrm{w}_{--}^{+}+\mathrm{w}_{--}^{-}+\mathrm{w}_{--}^{++}\right)\end{array}\right]$.

It is clear that this equation satisfies the condition that the sum of the components of the probability vector $\mathcal{P}$ should be unity at all times. This is guaranteed by the property that the sum of the rows of $\mathcal{M}$ vanishes.

To determine the flip rates, we need to connect the generic kinetic equation, Eq. (5.13), to the nuclear spin evolution operator, Eq. (5.12), derived earlier. This can be done by starting
from the evolution over one driving period:

$$
\begin{equation*}
\mathcal{S}_{n}\left(t+T_{R}\right)=\mathcal{Y}_{n} \mathcal{S}_{n}(t) \tag{5.15}
\end{equation*}
$$

The fact that the nuclear spin evolution is much slower than the driving period $T_{R}$ allows us to coarse-grain this equation to arrive at a continuous evolution equation:

$$
\begin{equation*}
\frac{d}{d t} \mathcal{S}_{n}=\frac{1}{T_{R}}\left(\mathcal{Y}_{n}-\mathbb{1}\right) \mathcal{S}_{n} \tag{5.16}
\end{equation*}
$$

Because we have defined $\mathcal{S}_{n}$ such that its first four components are just the populations of the nuclear spin states, we can identify this equation with $\dot{\mathcal{P}}=\mathcal{M P}$ and therefore read off the flip-rate matrix components from the nuclear spin evolution matrix:

$$
\begin{equation*}
\mathcal{M}_{i j}=\frac{1}{T_{R}}\left(\mathcal{Y}_{n}-\mathbb{1}\right)_{i j}, \quad i, j=1 \ldots 2 I+1 \tag{5.17}
\end{equation*}
$$

This allows us to read off the flip rates from the nuclear spin dynamical map. It is worth noting that $\mathcal{Y}_{n}$ contains not only terms that mix the populations of the different nuclear spin levels but also terms that mix populations and nuclear spin coherences. Here, we are neglecting the influence of the latter on the late-time populations. In numerical simulations, we find that these terms have a negligible effect on the flip rates. Moreover, they will be further suppressed by nuclear spin dephasing [92, 112], which happens quickly compared to nuclear spin flips. This simplification allows us to obtain non-perturbative expressions for the flip rates.

In the case of $I=1 / 2$ nuclei, the flip rates can be obtained analytically following the above procedure:

$$
\begin{equation*}
\mathrm{w}_{ \pm}=\frac{A^{2}\left(1 \pm S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \sqrt{\left(\omega_{e}-\omega_{n}\right)^{2}+A^{2}} / 2\right)}{2 T_{R}\left[\left(\omega_{e}-\omega_{n}\right)^{2}+A^{2}\right]} \tag{5.18}
\end{equation*}
$$

where we use the shorthand notation $\mathrm{w}_{+} \equiv \mathrm{w}_{-1 / 2}^{+1 / 2}$ and $\mathrm{w}_{-} \equiv \mathrm{w}_{+1 / 2}^{-1 / 2}$. Note that unlike in Refs. [150, 151], here we did not have to resort to perturbation theory in the HF interaction to obtain an analytical expression for the flip rates. The flip rates for $I=1$ and $I=3 / 2$ can also be obtained analytically in the case of zero quadrupolar coupling, $\nu_{Q}=0$. In this case, there are four nonzero flip rates for $I=1$ :

$$
\begin{align*}
& \mathrm{w}_{-1}^{0}=\frac{A^{2}\left(1+S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{-}^{(1)} / 2\right)}{T_{R}\left(\Omega_{-}^{(1)}\right)^{2}}, \\
& \mathrm{w}_{0}^{-1}=\frac{A^{2}\left(1-S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{-}^{(1)} / 2\right)}{T_{R}\left(\Omega_{-}^{(1)}\right)^{2}}, \\
& \mathrm{w}_{0}^{+1}=\frac{A^{2}\left(1+S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{+}^{(1)} / 2\right)}{T_{R}\left(\Omega_{+}^{(1)}\right)^{2}}, \\
& \mathrm{w}_{+1}^{0}=\frac{A^{2}\left(1-S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{+}^{(1)} / 2\right)}{T_{R}\left(\Omega_{+}^{(1)}\right)^{2}}, \tag{5.19}
\end{align*}
$$

with

$$
\begin{equation*}
\Omega_{ \pm}^{(1)}=\sqrt{\left(\omega_{e}-\omega_{n}\right)^{2} \pm A\left(\omega_{e}-\omega_{n}\right)+9 A^{2} / 4} \tag{5.20}
\end{equation*}
$$

while there are six nonzero flip rates for $I=3 / 2$ :

$$
\begin{align*}
\mathrm{w}_{+}^{++} & =\frac{3 A^{2}\left(1+S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{+1}^{(3 / 2)} / 2\right)}{2 T_{R}\left(\Omega_{+1}^{(3 / 2)}\right)^{2}}, \\
\mathrm{w}_{++}^{+} & =\frac{3 A^{2}\left(1-S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{+1}^{(3 / 2)} / 2\right)}{2 T_{R}\left(\Omega_{+1}^{(3 / 2)}\right)^{2}}, \\
\mathrm{w}_{-}^{+} & =\frac{A^{2}\left(1+S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{0}^{(3 / 2)} / 2\right)}{T_{R}\left(\Omega_{0}^{(3 / 2)}\right)^{2}}, \\
\mathrm{w}_{+}^{-} & =\frac{A^{2}\left(1-S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{0}^{(3 / 2)} / 2\right)}{T_{R}\left(\Omega_{0}^{(3 / 2)}\right)^{2}}, \\
\mathrm{w}_{--}^{-}= & \frac{3 A^{2}\left(1+S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{-1}^{(3 / 2)} / 2\right)}{2 T_{R}\left(\Omega_{-1}^{(3 / 2)}\right)^{2}}, \\
\mathrm{w}_{-}^{--}= & \frac{3 A^{2}\left(1-S_{e, z}^{s s}\right) \sin ^{2}\left(T_{R} \Omega_{-1}^{(3 / 2)} / 2\right)}{2 T_{R}\left(\Omega_{-1}^{(3 / 2)}\right)^{2}} \tag{5.21}
\end{align*}
$$

with

$$
\begin{equation*}
\Omega_{\eta}^{(3 / 2)}=\sqrt{\left(\omega_{e}-\omega_{n}\right)^{2}+2 \eta A\left(\omega_{e}-\omega_{n}\right)+4 A^{2}} \tag{5.22}
\end{equation*}
$$

In the absence of quadrupolar interactions, only $\Delta m_{I}= \pm 1$ transitions (i.e., transitions between adjacent spin levels) are allowed, as follows directly from the form of the HF flipflop interaction. When the quadrupolar coupling is nonzero, we can no longer obtain an analytical expression for the flip rates, but these are still easily obtained numerically by computing $\mathcal{Y}_{n}$ for specific parameter values.

### 5.3.3 Multi-nuclear flip rates

We can convert the single-nucleus flip rates obtained above into multi-nuclear flip rates by making them dependent on the magnetization of the entire nuclear spin bath. This dependence comes from the Overhauser effect in which nuclear spin polarization acts as an


Figure 5.4: Single-nucleus spin-flip rates as a function of nuclear spin bath magnetization $m$ for $I=1$ and for different values of the quadrupolar angle $\theta$. (a) Flip rate for the $\Delta m_{I}=1$ transition $|0\rangle \rightarrow|+1\rangle$. (b) Flip rate difference for the $|0\rangle \leftrightarrow|+1\rangle$ transitions. (c) Flip rate for the $\Delta m_{I}=2$ transition $|-1\rangle \rightarrow|+1\rangle$. The parameter values are $T_{R}=13.2 \mathrm{~ns}$, $N A=10 \mathrm{GHz}, N=1000, \omega_{e 0}=0.5 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}, q_{0}=0.3$, $\phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}$.
effective magnetic field seen by the electron spin. We incorporate this effect by adding a magnetization-dependent shift to the precession frequency of the electron:

$$
\begin{equation*}
w_{i}^{j}(m)=\mathrm{w}_{i}^{j}\left(\omega_{e} \rightarrow \omega_{e 0}+m A\right) \tag{5.23}
\end{equation*}
$$

where $\omega_{e 0}$ denotes the contribution to the precession frequency due purely to the external magnetic field, and where we use $w_{i}^{j}(m)$ to denote the rate to flip from state $i$ to state $j$ in the presence of nuclear spin magnetization $m$. For nuclei of spin $I$, we can express this magnetization in terms of occupation numbers, $N_{\ell}$, for each of the nuclear spin states:

$$
\begin{equation*}
m=\sum_{\ell=-I}^{I} \ell N_{\ell} \tag{5.24}
\end{equation*}
$$

Fig. 5.3 shows the dependence of the flip rates on the net magnetization $m$ for $I=1 / 2,1$, and $3 / 2$. In this figure, results for zero quadrupolar angle, $\theta=0$, are shown in the $I>1 / 2$ cases. Even though the quadrupolar coupling is nonzero, $\nu_{Q}>0$, only $\Delta m_{I}= \pm 1$ transitions are permitted in this case because when $\theta=0$, the only effect of the quadrupolar interaction is to modify the energy splittings between nuclear spin levels, and so the selection rules are


Figure 5.5: Single-nucleus spin-flip rates as a function of nuclear spin bath magnetization $m$ for $I=3 / 2$ and for different values of the quadrupolar angle $\theta$. (a) Flip rate for the $\Delta m_{I}=1$ transition $|-1 / 2\rangle \rightarrow|+1 / 2\rangle$. (b) Flip rate for the $\Delta m_{I}=3$ transition $|-3 / 2\rangle \rightarrow|+3 / 2\rangle$. (c) Flip rate for the $\Delta m_{I}=1$ transition $|-3 / 2\rangle \rightarrow|-1 / 2\rangle$. (d) Flip rate for the $\Delta m_{I}=2$ transition $|-3 / 2\rangle \rightarrow|+1 / 2\rangle$. (e) Flip rate difference for the $|-3 / 2\rangle \leftrightarrow|-1 / 2\rangle$ transitions. (f) Flip rate difference for the $|-3 / 2\rangle \leftrightarrow|+1 / 2\rangle$ transitions. The parameter values are $T_{R}=13.2 \mathrm{~ns}, N A=10 \mathrm{GHz}, N=1000, \omega_{e 0}=0.5 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}$, $q_{0}=0.3, \phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}$.
still determined solely by the HF interaction. We discuss the effect of nonzero $\theta$ below.

The salient features evident in Fig. 5.3 can be understood from the non-perturbative expressions for the flip rates given above. First of all, the flip rates are strongly peaked at magnetization $m \approx-\omega_{e 0} / A$. In the spin $1 / 2$ case, the precise location of the peak is the value of $m$ at which the argument of the sine in Eq. (5.18) vanishes since the flip rates are essentially given by squared sinc functions. For low to moderate external magnetic field strengths and large $N$, the terms involving $\omega_{n}$ and $A^{2}$ can be neglected, leaving $m \approx-\omega_{e 0} / A$. Similar statements hold for $I=1$ and $I=3 / 2$ in the absence of quadrupolar effects, as is clear from Eqs. (5.19) and (5.21). The fact that the flip rates are maximal at $m \approx-\omega_{e 0} / A$ can be understood from energy conservation: At these values, the effective Zeeman energy of the electron is almost zero, and thus so is the energy mismatch between the electron and nucleus. This in turn reduces the energy penalty for flip-flops, accelerating the transfer of
polarization. Conversely, the overall decay of the flip rates away from $m \approx \omega_{e 0} / A$ is due to the HF interaction becoming inefficient at overcoming the large energy mismatch between the electronic and nuclear spin splittings.

It is also evident in Fig. 5.3 that the flip rates vanish periodically as a function of $m$. The periodicity is also controlled by the arguments of the sine functions in the flip rates. These zeros correspond to values of $\omega_{e}$ for which complete flip-flops between the electronic and nuclear spins occur-polarization is transferred back and forth between the electron and nucleus an integer number of times within a single drive period $T_{R}$. Because there is no net polarization transfer, the flip rate vanishes. For $I>1 / 2$, the locations of these zeros depend on which pair of adjacent spin levels we consider, although this dependence fades away in the large $N$ limit, where $A \rightarrow 0$. In the next section, we show that these flip-rate zeros play a central role in the phenomenon of mode-locking.

Each pair of flip rates describing transitions between the same two spin levels are almost equal [see panels (b), (d), (f) of Fig. 5.3]. As can be seen from Eqs. (5.18)-(5.21), the differences of these flip rates are proportional to $S_{e, z}^{s s}(m)$, and this component of the electron steady state is suppressed near $m \approx-\omega_{e 0} / A$ because it is proportional to $\omega_{e}$ (see Eq. (5.49)). This is a reflection of the fact that when $\omega_{e}=0$, the electron steady state becomes polarized along the optical pulse axis (the $x$ direction), where it is no longer affected by the pulses and is thus stabilized. In the figure, we see that this combination of accelerated flip-flops and the suppression of $S_{e, z}^{s s}(m)$ near $m \approx-\omega_{e 0} / A$ results in flip rate differences that are more than two orders of magnitude smaller than the flip rates themselves.

The effect of a nonzero quadrupolar angle $\theta$ on the flip rates is shown in Figs. 5.4 and 5.5 for $I=1$ and $3 / 2$, respectively. In the case $I=1$, it is evident that $\theta$ has a negligible effect on the $\Delta m_{I}= \pm 1$ flip rates. On the other hand, sufficiently large values of the angle, $\theta \gtrsim \pi / 4$, give rise to $\Delta m_{I}= \pm 2$ transitions that are not otherwise present. Although the rates for these
transitions are two orders of magnitude smaller than those of the $\Delta m_{I}= \pm 1$ transitions, they are still large enough to affect the polarization distribution of the nuclear spin bath, as we show in Sec. 5.4. Similar but somewhat more prominent effects are evident for $I=3 / 2$ in Fig. 5.5. Here, larger values of $\theta$ produce small but noticeable changes in $\Delta m_{I}= \pm 1$ flip rates, significant $\Delta m_{I}= \pm 2$ transition rates, and even $\Delta m_{I}= \pm 3$ transitions. A striking feature evident in both Figs. 5.4 and 5.5 is that the flip rates for $\Delta m_{I}= \pm 2$ transitions do not decay as $m$ moves away from $m=-\omega_{e 0} / A$. This is consistent with the fact that spin flips caused by the quadrupolar interaction do not require the electron and nuclear spin Zeeman energies to be equal. Unlike HF spin flips, quadrupolar spin flips depend weakly on the bath magnetization. On the other hand, the $\Delta m_{I}= \pm 3$ flip rates are sensitive to $m$ (see Fig. 5.5(b)), because these arise from a higher-order process that combines HF and quadrupolar spin flips.

### 5.3.4 Kinetic equations for multi-nuclear spin polarization distributions

In this subsection, we use the flip rates obtained in the previous subsection to construct kinetic rate equations that govern the evolution of the polarization distribution of the entire nuclear spin bath. We do this for each of the three values of nuclear total spin $I$ considered in this work. Although the kinetic equation for $I=1 / 2$ has been discussed in detail elsewhere [150, 151], here we present an analytical solution to this equation that was not previously known. The kinetic equations for $I=1$ and $3 / 2$ will be solved numerically in the next section to obtain nuclear spin polarization distributions in these cases. Detailed comparisons of the polarization distributions that result in all three cases for various parameter values are given below in Sec. 5.4. In that section, these distributions are then used to compute the effect on the electron spin evolution with and without quadrupolar interactions.

## Kinetic equation for spin $I=1 / 2$ nuclei

The polarization of a spin $1 / 2$ nuclear bath in a definite configuration with occupation numbers $N_{+}$and $N_{-}$(the number of spins in the $|+1 / 2\rangle$ and $|-1 / 2\rangle$ states, respectively) is given by $m=\left(N_{+}-N_{-}\right) / 2$. The total number of spins is $N=N_{+}+N_{-}$. Knowledge of the polarization $m$ is sufficient to determine the two occupation numbers, $N_{+}$and $N_{-}$. This in turn means that the probability of each bath configuration is equal to the polarization probability distribution $P(m)$. We may write down a kinetic equation governing the dynamics of this distribution [150, 151]:

$$
\begin{equation*}
\frac{d}{d t} P(m)=-\sum_{ \pm}\left[w_{ \pm}(m) \frac{N \mp 2 m}{2}\right] P(m)+\sum_{ \pm} w_{\mp}(m \pm 1)\left[\frac{N \pm 2 m}{2}+1\right] P(m \pm 1) \tag{5.25}
\end{equation*}
$$

A close look at this kinetic equation reveals that the right-hand side is comprised of two terms that are related to each other by shifting $m \rightarrow m+1$ :

$$
\begin{equation*}
\frac{d}{d t} P(m)=F(m+1)-F(m) \tag{5.26}
\end{equation*}
$$

where $F(m)=w_{-}(m)(m+N / 2) P(m)-w_{+}(m-1)(-m+1+N / 2) P(m-1)$. Therefore, in the steady state where $d P(m) / d t=0$, we find $F(m)=F(m+1)=$ constant. Since we must have $P(N+1)=0$, it follows that this constant is zero. The equation $F(m)=0$ then yields a two-term recursion relation $[150,151]$ :

$$
\begin{equation*}
P(m)=\frac{N-2 m+2}{N+2 m} \frac{w_{+}(m-1)}{w_{-}(m)} P(m-1) . \tag{5.27}
\end{equation*}
$$

This relation can easily be solved iteratively starting from an arbitrary value for $P(-N)$ and then imposing the normalization condition $\sum_{m} P(m)=1$. This approach was used to produce numerical results for the polarization distribution in Refs. [150, 151]. Notice that
this procedure yields the unique steady state of the kinetic equation, Eq. (5.25). Because of this uniqueness, the steady state must be stable. This is evident from the kinetic equation, where a positive fluctuation that takes $P(m)$ away from its steady-state value results in $d P(m) / d t<0$, which indicates that the steady state will be subsequently restored. An analogous statement holds for a negative fluctuation as well. The kinetic equations for $I>1 / 2$ described below also possess this property.

Here, we obtain an analytical solution for $P(m)$ by exploiting the explicit, non-perturbative expressions we obtained for the flip rates in Eq. (5.18). First of all, an expression for $P(m)$ follows immediately from Eq. (5.27):

$$
\begin{align*}
P(m) & =\mathcal{N}^{-1} \prod_{k=1-N / 2}^{m} \frac{N-2 k+2}{N+2 k} \frac{w_{+}(k-1)}{w_{-}(k)} \\
& =\frac{\mathcal{N}^{-1} N!}{(N / 2+m)!(N / 2-m)!} \prod_{k=1-N / 2}^{m} \frac{w_{+}(k-1)}{w_{-}(k)} \tag{5.28}
\end{align*}
$$

where $\mathcal{N}$ is a normalization factor. Next, we use the fact that the two flip rates only differ by the sign in front of $S_{e, z}^{s s}(m)$, which leads to a cascade of cancellations between the numerator and denominator in the product. We are left with

$$
\begin{align*}
P(m)= & \frac{\mathcal{N}^{-1}}{(N / 2+m)!(N / 2-m)!} \prod_{k=1-N / 2}^{m} \frac{1+S_{e, z}^{s s}(k-1)}{1-S_{e, z}^{s s}(k)} \\
& \times \frac{\left(\omega_{e 0}-\omega_{n}+A m\right)^{2}+A^{2}}{\sin ^{2}\left(T_{R} \sqrt{\left(\omega_{e 0}-\omega_{n}+A m\right)^{2}+A^{2}} / 2\right)}, \tag{5.29}
\end{align*}
$$

where we have absorbed additional constants into $\mathcal{N}$. The first, combinatoric factor in $P(m)$ corresponds to a Gaussian-like envelope that quickly approaches a Gaussian as $N$ increases: $[(N / 2)!]^{2} /[(N / 2+m)!(N / 2-m)!] \rightarrow e^{-2 m^{2} / N}$ as $N \rightarrow \infty$. The second factor in Eq. (5.29)
produces sharp spikes at values of $m$ that correspond to the zeros of the flip rates. These values of $m$ satisfy

$$
\begin{equation*}
\sqrt{\left(\omega_{e 0}-\omega_{n}+A m\right)^{2}+A^{2}} \approx \frac{2 \pi p}{T_{R}} \tag{5.30}
\end{equation*}
$$

where $p$ is an integer. The concentration of probability near these special values of $m$ produces mode-locking: Nuclear polarization shifts the electron Zeeman frequency to values where HF flip-flops stop transferring polarization between the electronic and nuclear spins. This happens because an integer number of flip-flops occur during each drive period. Using that $\omega_{n} \ll \omega_{e 0}$ and assuming $N$ is sufficiently large that $A \ll \omega_{e 0}$, these values of $m$ correspond to the electron precession becoming commensurate with the pulse train: $\omega_{e}=\omega_{e 0}+A m \approx 2 \pi p / T_{R}$, which is the primary signature of mode-locking seen in experiments $[132,133,134,135,136,137,138]$.

The middle factor (the product) in Eq. (5.29) is primarily responsible for the average magnetization of the nuclear spin bath, $\langle m\rangle=\sum_{m} m P(m)$. This factor is also where additional pulse parameters such as the rotation angle $\phi$ and the residual ground state population $q_{0}$ influence the polarization distribution. If $\phi$ is equal to 0 or $\pi$ or if $q_{0}$ is zero, then $S_{e, z}^{s s}(k)=0$ for all $k$, in which case the final factor in Eq. (5.29) reduces to 1 . In this case, the combinatoric factor, which is centered about $m=0$, ensures that the average magnetization will be small, $\langle m\rangle \approx 0$. On the other hand, if $\phi \neq 0$ and the external magnetic field is sufficiently large, then $\langle m\rangle$ can be significant, and its sign depends on the sign of $\phi$ and on the orientation of the external field. If $\phi>0$, then $S_{e, z}^{s s}(m)$ is more often positive than negative for $m<-\omega_{e 0} / A$, which in turn means that $1+S_{e, z}^{s s}(m-1) / 1-S_{e, z}^{s s}(m)$ is biased toward values larger than 1 , and so the product grows as $m$ increases. Once $m$ passes $-\omega_{e 0} / A, S_{e, z}^{s s}(m)$ now tends to more negative values, and the product shrinks as $m$ increases. Thus, we see that for $\phi>0$, the product in Eq. (5.29) is peaked at $m \approx-\omega_{e 0} / A$, and so the average magnetization will lie between 0 and $-\omega_{e 0} / A$. On the other hand, if $\phi<0$, then the same reasoning leads
to the conclusion that the product in Eq. (5.29) has a dip at $m \approx-\omega_{e 0} / A$, and thus the net magnetization is driven away from this point and will have a sign that coincides with that of $\omega_{e 0}$. These features are borne out in plots of Eq. (5.29), as shown below in Sec. 5.4.

## Kinetic equation for $\operatorname{spin} I=1$ nuclei

Before we write down the kinetic equation for $I=1$ nuclei, we first introduce the notation we use to distinguish different bath configurations. We denote the occupation numbers of the three spin states by $N_{-1}, N_{0}$, and $N_{1}$. The bath polarization for a given configuration is then $m=+1 \times N_{1}+0 \times N_{0}-1 \times N_{-1}$. We see immediately that there is an important difference compared to the $I=1 / 2$ case considered above: The polarization does not uniquely specify a configuration of the bath. For instance, in the case of two $I=1$ spins with $m=0$, we can have either $N_{1}=1=N_{-1}$ and $N_{0}=0$ or $N_{1}=0=N_{-1}$ and $N_{0}=2$. This is in contrast to the $I=1 / 2$ case, where each value of $m$ corresponds to a unique configuration. As the number of spins increases, the number and orders of such "degeneracies" grow quickly. Because the polarization does not uniquely specify a configuration, we must combine it with one of the occupation numbers to uniquely label different configurations. We choose to use $N_{0}$ and express the probability of a given configuration by $P\left(m, N_{0}\right)$. Unlike in the spin $1 / 2$ case, this quantity is now distinct from the polarization probability distribution; the latter is obtained by summing over all possible values of $N_{0}$ that are consistent with the given value of $m$ :

$$
\begin{equation*}
P(m)=\sum_{N_{0}} P\left(m, N_{0}\right) . \tag{5.31}
\end{equation*}
$$

We can write down a kinetic equation for $P\left(m, N_{0}\right)$ :

$$
\begin{align*}
\frac{d}{d t} P\left(m, N_{0}\right)= & F\left(m, N_{0}\right)+G\left(m+1, N_{0}-1\right) \\
& -G\left(m, N_{0}\right)-F\left(m+1, N_{0}+1\right) \tag{5.32}
\end{align*}
$$

where

$$
\begin{align*}
& F\left(m, N_{0}\right)=-w_{0}^{-1} P\left(m, N_{0}\right) N_{0}+w_{-1}^{0}(m-1) P\left(m-1, N_{0}-1\right) N_{-}\left(m-1, N_{0}-1\right), \\
& G\left(m, N_{0}\right)=w_{1}^{0} P\left(m, N_{0}\right) N_{+}\left(m, N_{0}\right)-w_{0}^{1}(m-1) P\left(m-1, N_{0}+1\right)\left(N_{0}+1\right) . \tag{5.33}
\end{align*}
$$

Here $N_{ \pm}\left(m, N_{0}\right) \equiv(1 / 2)\left(N \pm m-N_{0}\right)$. In the kinetic equation above we have only considered the $\Delta m_{I}= \pm 1$ transitions. Including transitions that change the angular momentum by more than 1 (for instance due to quadrupolar interactions) leads to additional terms not shown above. Such terms are illustrated for the case of $I=3 / 2$ nuclei in the next section. Returning to the spin 1 case, the steady state of the above kinetic equation,

$$
\begin{equation*}
F\left(m, N_{0}\right)-G\left(m, N_{0}\right)=F\left(m+1, N_{0}+1\right)-G\left(m+1, N_{0}-1\right) \tag{5.34}
\end{equation*}
$$

does not yield a recursion relation as in the $I=1 / 2$ case. We solve this equation (and its generalization for nonzero quadrupolar interactions) numerically in Sec. 5.4.

## Kinetic equation for spin $I=3 / 2$ nuclei

We again adopt the notation $\{++,+,-,--\}$ to label quantities associated with the four spin quantum numbers $m_{I}=\{+3 / 2,+1 / 2,-1 / 2,-3 / 2\}$ of a spin $3 / 2$ nucleus. For a nuclear spin bath comprised of $N=N_{++}+N_{+}+N_{-}+N_{--}$spins, the magnetization of the system (Eq. (5.24)) is $m=\left(3 N_{++}+N_{+}-N_{-}-3 N_{--}\right) / 2$. In the $I=3 / 2$ case, we need two more
quantities in addition to $m$ to uniquely label different multi-spin configurations. We choose these to be $N_{++}$and $N_{--}$. The remaining two occupation numbers are then determined by these three quantities for a fixed total number of spins:

$$
\begin{align*}
& N_{+}=\frac{1}{2}\left(2 m+N-4 N_{++}+2 N_{--}\right)  \tag{5.35}\\
& N_{-}=\frac{1}{2}\left(-2 m+N+2 N_{++}-4 N_{--}\right) . \tag{5.36}
\end{align*}
$$

The probabilities $P\left(m, N_{++}, N_{--}\right)$that the nuclear spin bath is in the various configurations labeled by $m, N_{++}$, and $N_{--}$obey the following set of kinetic equations:

$$
\begin{align*}
\frac{d}{d t} P\left(m, N_{++}, N_{--}\right)= & F\left(m, N_{++}, N_{--}\right)+G\left(m, N_{++}, N_{--}\right)+H\left(m, N_{++}, N_{--}\right) \\
& +I\left(m, N_{++}, N_{--}\right)+J\left(m, N_{++}, N_{--}\right) \\
& -F\left(m+1, N_{++}+1, N_{--}\right)-G\left(m+1, N_{++}, N_{--}-1\right)-H\left(m+1, N_{++}, N_{--}\right) \\
& -I\left(m-2, N_{++}-1, N_{--}\right)-J\left(m+2, N_{++}, N_{--}-1\right) \tag{5.37}
\end{align*}
$$

where

$$
\begin{align*}
F\left(m, N_{++}, N_{--}\right)= & +w_{+}^{++}(m-1) P\left(m-1, N_{++}-1, N_{--}\right) N_{+}\left(m-1, N_{++}-1, N_{--}\right) \\
& -w_{++}^{+}(m) P\left(m, N_{++}, N_{--}\right) N_{++}  \tag{5.38}\\
G\left(m, N_{++}, N_{--}\right)= & +w_{--}^{-}(m-1) P\left(m-1, N_{++}, N_{--}+1\right)\left(N_{--}+1\right) \\
& -w_{-}^{--}(m) P\left(m, N_{++}, N_{--}\right) N_{-}\left(m, N_{++}, N_{--}\right) \\
H\left(m, N_{++}, N_{--}\right)= & +w_{-}^{+}(m-1) P\left(m-1, N_{++}, N_{--}\right) N_{-}\left(m-1, N_{++}, N_{--}\right) \\
& -w_{+}^{-}(m) P\left(m, N_{++}, N_{--}\right) N_{+}\left(m, N_{++}, N_{--}\right)  \tag{5.39}\\
I\left(m, N_{++}, N_{--}\right)= & +w_{++}^{-}(m+2) P\left(m+2, N_{++}+1, N_{--}\right)\left(N_{++}+1\right) \\
& -w_{-}^{++}(m) P\left(m, N_{++}, N_{--}\right) N_{-}\left(m, N_{++}, N_{--}\right)  \tag{5.40}\\
J\left(m, N_{++}, N_{--}\right)= & +w_{--}^{+}(m-2) P\left(m-2, N_{++}, N_{--}+1\right)\left(N_{--}+1\right) \\
& -w_{+}^{--}(m) P\left(m, N_{++}, N_{--}\right) N_{+}\left(m, N_{++}, N_{--}\right) \tag{5.41}
\end{align*}
$$

Here, we have included $\Delta m_{I}= \pm 1$ and $\Delta m_{I}= \pm 2$ transitions. Although $\Delta m_{I}= \pm 3$ transitions cannot be directly driven by either the HF interaction or the quadrupolar interaction to first order in their respective coupling strengths, they can potentially arise from higherorder effects as we saw from the flip rates in Fig. 5.5. Now that we have the kinetic equations governing the nuclear polarization, the next step is to solve them.


Figure 5.6: Structure of the matrix $\mathcal{R}$ defining the linear system of equations governing the steady-state solution of the multi-nuclear kinetic equation for spin $I=1$ for (left) $N=3$ spins and (right) $N=20$ spins in the absence of quadrupolar interactions.


Figure 5.7: Steady-state nuclear spin polarization distribution of a bath with $N=200$ nuclear spins for (a) $I=1 / 2$, (b) $I=1$, and (c) $I=3 / 2$. The parameter values are $T_{R}=13.2 \mathrm{~ns}, N A=10 \mathrm{GHz}, \omega_{e 0}=0.5 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}, q_{0}=0.3$, $\phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}$. In the case of $I=3 / 2$ and $I=1$ the quadrupolar angle is $\theta=0$.


Figure 5.8: Extrapolation of the average nuclear spin bath polarization $\langle m\rangle$ to larger bath sizes $N$ for two values of total spin: $I=1$ (red circles) and $I=3 / 2$ (blue diamonds). The points are obtained by solving the respective kinetic equations, Eqs. (5.32) and (5.37). The lines are linear fits. The parameter values are $T_{R}=13.2 \mathrm{~ns}, N A=10 \mathrm{GHz}, \omega_{e 0}=0.5 \mathrm{GHz}$, $\omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}, q_{0}=0.3, \phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}, \theta=0$.


Figure 5.9: Steady-state nuclear spin polarization distribution of a bath with $N=150$ nuclear spins for four different values of the quadrupolar angle $\theta$ for (a) $I=1$ and (b) $I=3 / 2$. The parameter values are $T_{R}=13.2 \mathrm{~ns}, N A=10 \mathrm{GHz}, \omega_{e 0}=0.5 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}$, $\gamma_{e}=0.5 \mathrm{GHz}, q_{0}=0.3, \phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}$.

### 5.4 Nuclear polarization distribution and feedback

### 5.4.1 Steady-state polarization distributions

For $I=1$ and $I=3 / 2$, we solve the respective kinetic equations numerically to obtain steady-state polarization distributions. This is done by first setting the time derivatives to zero: $\frac{d}{d t} P\left(m, N_{++}, N_{--}\right)=0$. The resulting algebraic equations are then collected together and written as a matrix $\mathcal{R}$ acting on a vector $\mathcal{V}$ of the probabilities $P\left(m, N_{++}, N_{--}\right)$such that $\mathcal{R} \mathcal{V}=0$. Thus, the steady-state polarization distribution is the unique null vector of $\mathcal{R}$. The matrix $\mathcal{R}$ depends on the Overhauser-shifted flip rates and occupation numbers for each configuration. The linear dimension of this matrix is equal to the number of distinct multi-spin configurations. For $N$ spins of total spin $I$, the number of configurations is given by the simplicial polytopic numbers $\binom{N+2 I}{2 I}$. For $I=1 / 2,1$, and $3 / 2$, this gives $N+1$, $(N+1)(N+2) / 2$, and $(N+1)(N+2)(N+3) / 6$, respectively. Therefore, in the case of $I=1$, we must compute the null vector of a matrix that grows quadratically with the number of nuclei, while for $I=3 / 2$, we must do the same for a matrix that grows like $N^{3}$. The matrix $\mathcal{R}$ is quite sparse in both cases (see Fig. 5.6), especially in the absence of quadrupolar interactions. This allows us to employ the Arnoldi method to compute the steady-state polarization distribution for hundreds of spins with $I=3 / 2$ and thousands of spins with $I=1$.

Fig. 5.7 compares results for the steady-state nuclear spin polarization for $N=200$ for all three values of $I$. In the $I>1 / 2$ cases, we set the quadrupolar angle to zero, $\theta=0$; however, the nonzero quadrupolar interaction $\nu_{Q}>0$ still modifies the energy splittings between the nuclear spin levels. In all three cases, the polarization distribution exhibits multiple narrow peaks at values of $m$ that correspond to the mode-locking frequencies, i.e., these values of $m$ are such that $\omega_{e 0}+A m=2 \pi p / T_{R}$ where $p$ is an integer (for an analytical
derivation of the $I=1 / 2$ case see Section 5.3.4). As discussed in Sec. 5.3.2, the flip rates approximately vanish at these values of $m$. (Note that the spacing of the peaks in Fig. 5.7 is five times smaller than the spacing of the flip-rate zeros in Figs. 5.3, 5.4, and 5.5 because this spacing is proportional to $1 / A=N / \mathcal{A}$, and $N$ is five times smaller in Fig. 5.7.) The steady-state probabilities $P\left(m, N_{++}, N_{--}\right)$are largest at these magnetization values because they are multiplied by nearly vanishing flip rates in the kinetic equations; the probabilities must compensate for the smallness of the flip rates such that the product of the two is finite and comparable to terms of similar size in the kinetic equations. This trend can be seen explicitly from the analytical solution in the $I=1 / 2$ case, Eq. (5.29), where it is evident that $P(m)$ depends inversely on the flip rates. In Fig. 5.7, we see that this also occurs for $I>1 / 2$. For all values of $I$, we can physically understand the formation of probability peaks at flip-rate zeros as resulting from the fact that, at these magnetization values, the joint electron-nuclear spin evolution under the HF interaction becomes commensurate with the driving pulses. Consequently, the pulses do not cause a net polarization transfer between the electron and nuclear spins. Thus, these values of the magnetization $m$ provide a point of stability in the electron-nuclear feedback mechanism. We also see from Fig. 5.7(a), and to some degree from Fig. 5.7(b), that the polarization distribution is suppressed in the vicinity of $m=-\omega_{e 0} / A$ (which corresponds to $m=-10$ for the parameters used in the figure). This is due to the fact that the flip rates are largest near these magnetization values and therefore drive population away from these values.

Another striking feature of the polarization distributions in Fig. 5.7 is that the distributions for $I>1 / 2$ exhibit broad envelopes in addition to the mode-locking peaks. This is a consequence of the fact that there are multiple distinct flip rates for $I>1 / 2$, as shown in Eqs. (5.19) and (5.21). These flip rates oscillate with $\omega_{e}$ at distinct frequencies that differ from each other by an amount proportional to $A$. Therefore, they do not all vanish at
the same values of $\omega_{e}$, dulling the sharpness of the mode-locking peaks. This effect becomes diminished at larger $N$, because in this limit $A$ decreases, and all the flip-rate zeros approach the values of $m$ at which $\omega_{e 0}+A m=2 \pi p / T_{R}$, where $p$ is an integer, producing a more comblike distribution. The broadening of the distribution at smaller values of $N$ is an important feature that is missed when $I=1 / 2$ spins are used to model $I>1 / 2$ spin baths. In the example of Fig. 5.7, we see that it also leads to an increase in the average magnetization $\langle m\rangle$ due to the enhanced weight of the distribution at positive magnetizations. This enhancement is more pronounced for $I=3 / 2$ compared to $I=1$. Fig. 5.8 examines the behavior of $\langle m\rangle$ as a function of $N$. The points are obtained by solving the respective kinetic equations, Eqs. (5.32) and (5.37). In the $I=1$ case, it is possible to obtain results for much larger bath sizes because the $\mathcal{R}$ matrix is much smaller in this case. For both $I=1$ and $I=3 / 2$, the points are well described by a linear relationship between $\langle m\rangle$ and $N$, as shown in the figure. We find that for the parameters considered and for large $N$, the average polarization for $I=3 / 2$ is approximately two times larger compared to that of an $I=1$ bath, with the net polarization in this case approaching $9 \%$.

The effects of nonzero quadrupolar angle on the polarization distribution for $I=1,3 / 2$ are illustrated in Fig. 5.9. Here, we set $N=150$, because nonzero $\theta$ reduces the sparsity of the $\mathcal{R}$ matrix, making the numerical computation more intensive than before, especially for $I=3 / 2$. From Fig. 5.9(a), we see that for $I=1$, nonzero $\theta$ leads to quantitative changes in the heights of the mode-locking peaks, along with a slight redistribution of the probability to negative magnetizations for intermediate values of $\theta$. Similar behavior occurs for $I=3 / 2$, as shown in Fig. 5.9(b). The redistribution can be understood from the fact that, in the absence of the HF interaction, the quadrupolar coupling produces a Gaussian distribution centered around $m=0$. This is discussed in more detail below. The fact that this redistribution is strongest near $\theta=\pi / 4$ suggests that the $\Delta m_{I}= \pm 1$ quadrupolar-


Figure 5.10: Steady-state nuclear spin polarization distribution of a bath with $N=1000$ $I=1$ nuclear spins for four different values of the quadrupolar angle $\theta$. The other parameter values are $T_{R}=13.2 \mathrm{~ns}, N A=10 \mathrm{GHz}, \omega_{e 0}=0.5 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}$, $q_{0}=0.3, \phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}$.
driven transitions play an important role in this process. This effect constitutes another way in which the quadrupolar interaction can make the DNP process for $I>1 / 2$ depart significantly from what is predicted for an $I=1 / 2$ bath. Also notice that in both panels of Fig. 5.9, the polarization distributions are still suppressed near $m=-\omega_{e 0} / A$ even for $\theta>0$. This indicates that the HF contributions to the flip rates remain an important factor in shaping the overall distribution.

Fig. 5.10 again shows the effect of nonzero $\theta$ for $I=1$, but now for a bath of size $N=1000$. For $\theta=0$, there is a distinct comb-like structure that is the hallmark of mode-locking. However, for $\theta>0$, this structure quickly disappears and is replaced by an almost Gaussian distribution centered around zero magnetization. A Gaussian distribution is in fact what occurs in the absence of the HF interaction, because the flip rates are then purely due to the quadrupolar coupling, which means that they are independent of $m$ and are equal for


Figure 5.11: The average polarization $\langle m\rangle$ of a nuclear spin bath with $N=1000$ nuclei of total spin $I=1$ for several values of the quadrupolar angle in the range of $0 \leq \theta \leq \pi / 2$. The inset color map shows the steady-state nuclear spin polarization distribution over the same range of quadrupolar angles. The other parameter values are $T_{R}=13.2 \mathrm{~ns}, N A=10 \mathrm{GHz}$, $\omega_{e 0}=0.5 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}, q_{0}=0.3, \phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}$.


Figure 5.12: The feedback effect of nuclear spin polarization on the $x$ component of the electron spin steady state as a function of time over one drive period $T_{R}=13.2 \mathrm{~ns}$. (a) $N=150$ nuclei of spin $I=1 / 2,1$ and $3 / 2$, and (b) $N=1000$ nuclei of spin $I=1 / 2$ and 1. The parameter values are $N A=10 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}, q_{0}=0.3$, $\phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}$.


Figure 5.13: The feedback effect of $N=1000 I=1$ nuclear spins on the $x$ component of the electron spin steady state as a function of time over one drive period $T_{R}=13.2 \mathrm{~ns}$. Here the quadrupolar angles $\theta=0$ and $\theta=\pi / 2$ are considered for different bare electron Zeeman frequencies of (a) 0.5 GHz , (b) 2.45 GHz and (c) 15.19 GHz . The electron Zeeman frequencies chosen for (b) and (c) correspond to the local minima shown in Fig. 5.14 and the nuclear spin polarization distribution for (a) is shown in Fig. 5.10. The parameter values are $N A=10 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}, q_{0}=0.3, \phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}$.
$\Delta m_{I}>0$ and $\Delta m_{I}<0$. This shows that the quadrupolar interaction plays a much more important role compared to the HF interaction for the case considered in Fig. 5.10. This is because the larger value of $N$ corresponds to a reduction in the HF coupling $A$, and hence in the magnitude of the flip rates (see Eq. (5.19)). This in turn increases the relative importance of the quadrupolar interaction. This can be seen from Fig. 5.4, where it is evident that as $\theta$ increases, the flip rate for the $\Delta m_{I}=2$ transition quickly surpasses the difference in the flip rates for the $\Delta m_{I}= \pm 1$ transitions. As a consequence, the probability distribution is no longer sensitive to the detailed features of the $\Delta m_{I}= \pm 1$ transitions, which are responsible for both the comb-like mode-locking structure and the suppression near $m=-\omega_{e 0} / A$. This shows that even small values of $\theta$ can have a dramatic effect on the DNP process for large numbers of nuclei. This is quantified in Fig. 5.11, which shows how the nuclear spin polarization distribution and average magnetization, $\langle m\rangle$, depend on $\theta$. The latter quickly decays with increasing $\theta$. As is evident from the inset in Fig. 5.11, the distribution itself exhibits mode-locking fringes at small $\theta$ that become blurred at larger $\theta$. The sensitivity of mode-locking to the quadrupolar interaction suggests that it could be used as a diagnostic tool to estimate the quadrupolar coupling strength and angle in experiments. This is further supported in the next section, where we show how the steady-state electron spin vector in the presence of DNP feedback depends on the quadrupolar angle.

### 5.4.2 Feedback on electron spin

Once we obtain the steady-state polarization distribution of the nuclear spin bath, the final step is to update the steady state of the electron by applying the Overhauser shift to the Zeeman frequency:

$$
\begin{equation*}
\bar{S}_{e, i}^{s s}\left(t, \omega_{e 0}\right)=\sum_{m} P(m) S_{e, i}^{s s}\left(t, \omega_{e 0}+m A\right) \tag{5.42}
\end{equation*}
$$



Figure 5.14: The effect of the $I=3 / 2$ nuclear feedback on the $x$ component of the steadystate electron spin vector. The red filled circles indicate local minima of $S_{e, x}^{s s}$ (shown in gray) for several values of the electron Zeeman frequency $\omega_{e 0}$ without nuclear feedback. The other points indicate the values of $\bar{S}_{e, x}^{S s}\left(\omega_{e 0}\right)$ at the same values of $\omega_{e 0}$, but now with feedback included as in Eq. (5.42). Results for four different values of the quadrupolar angle $\theta$ are shown. Other parameter values are $N=150, T_{R}=13.2 \mathrm{~ns}, N A=10 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}$, $\gamma_{e}=0.5 \mathrm{GHz}, q_{0}=0.3, \phi=-\pi / 2, \nu_{Q}=2.8 \mathrm{MHz}$.


Figure 5.15: The nuclear spin polarization distributions corresponding to five of the electron Zeeman frequency values from Fig. 5.14 for quadrupolar angles (a) $\theta=0$, (b) $\theta=\pi / 8$, (c) $\theta=\pi / 4$, and (d) $\theta=\pi / 2$ for an $I=3 / 2$ nuclear bath. Other parameter values are $N=150, T_{R}=13.2 \mathrm{~ns}, N A=10 \mathrm{GHz}, \omega_{n}=-0.5 \mathrm{MHz}, \gamma_{e}=0.5 \mathrm{GHz}, q_{0}=0.3, \phi=-\pi / 2$, $\nu_{Q}=2.8 \mathrm{MHz}$.

Here the summation is over all possible values of $m$, and $t$ is the time elapsed since the last pulse. We obtain the time-evolved electron steady state by starting from the expression for the steady state immediately after a pulse, Eq. (5.49), and evolving it under Larmor precession with frequency $\omega_{e 0}+m A$ for time $t$. Fig. 5.12 shows the resulting DNP-modified electron steady state over one drive period for different species of nuclear spins. Fig. 5.12(a) compares $I=1 / 2,1$ and $3 / 2$ species for $N=150$, where the two latter DNP distributions are shown in Fig. 5.9 for $\theta=0$. Fig. 5.12(b) compares the cases $I=1 / 2$ and 1 for $N=1000$. It is evident that the total spin of the nuclei can have a significant effect on the electron spin precession in the steady state. Because the mode-locking effect is stronger in the spin $1 / 2$ case (see Fig. 5.7), the electron spin precession is closer to a sinusoid due to the fact that only a few discrete values of the Overhauser field contribute to the sum in Eq. (5.42). On the other hand, for larger spin, the nuclear polarization distribution is broader, giving rise to a beating in the electron spin vector over each driving period.

The role of quadrupolar interactions in the feedback is examined in Fig. 5.13, which shows the resulting DNP-modified electron steady state over one drive period for six different $N=1000$, $I=1$ polarization distributions. Two of these are distributions shown in Fig. 5.10-the ones corresponding to $\theta=0$ and $\theta=\pi / 2$. The modified steady states for these two cases are shown in Fig. 5.13(a), where it is evident that a large quadrupolar angle suppresses oscillations, both in the vicinity of the driving pulses and in the "echo" that occurs in the middle of the drive period near $t=T_{R} / 2$, which is 6.6 ns for the chosen parameter values. Similar behavior occurs for other values of the external magnetic field, as is demonstrated in Figs. 5.13(b), (c). It should be noted that the amplitude of these oscillations are used to identify the presence of mode-locking $[132,133,134,135,136,137,138]$, and so the suppression of these oscillations can provide an experimental indicator of substantial quadrupolar effects.

The electron steady state, Eq. (5.49), is a rapidly oscillatory function of the applied magnetic
field. In Ref. [151], it was found using perturbation theory that for $I=1 / 2$, nuclear feedback suppresses the amplitudes of these oscillations. In particular, it was shown that the $x$ component of the electron steady-state SV approaches unity for all values of the external magnetic field as a consequence of mode-locking: The SV becomes synchronized with the pulses such that it lies parallel to the optical axis at the pulse times. Here, we examine how this effect is modified by the presence of quadrupolar interactions. This is illustrated in the case of $I=3 / 2$ in Fig. 5.14, where we show the $x$-component of the electron steady state immediately after a pulse, $S_{e, x}^{s s}$, for ten different values of the electron Zeeman frequency with and without feedback. We are primarily interested in the amplitude of the electron steady-state oscillations, so we choose the ten different Zeeman frequencies that correspond to minima of the oscillations in the absence of feedback (red dots in Fig. 5.14). To find how the envelope of the electron spin oscillations is affected by the feedback process, we compute the nuclear spin polarization distributions for each of these minima. These distributions then alter the values of these minima according to Eq. (5.42) (with $t=0$ ). As can be seen from Fig. 5.14, the amplitude of the electron steady-state oscillations is suppressed (i.e., the minima increase up toward unity) in the presence of DNP, and the degree of this suppression varies weakly and nonmonotonically with the quadrupolar angle $\theta$. To understand this behavior better, in Fig. 5.15 we show the polarization distributions for five of the minima from Fig. 5.14 for four different quadrupolar angles. It is clear that for all values of $\theta$, as the electron spin Zeeman frequency due to the external magnetic field, $\omega_{e 0}$, is increased, the polarization distributions gravitate toward $m=0$. This is because larger values of the electron Zeeman frequency suppress HF flip-flops, as the violation of energy conservation becomes more pronounced in this case. This is why the $\theta=0$ curve in Fig. 5.14 monotonically decreases with increasing $\omega_{e 0}$. On the other hand, quadrupole-induced nuclear spin flips do not depend on the electron Zeeman frequency, and so these gradually begin to dominate as both $\theta$ and $\omega_{e 0}$ increase. This in turn causes the curves in Fig. 5.14 to
become essentially independent of $\omega_{e 0}$ as $\theta$ increases. This is another manifestation of how quadrupolar interactions suppress mode-locking effects.

### 5.5 Conclusions

In this work, we developed a general theoretical framework to describe the dynamics of an electron trapped in a self-assembled quantum dot that is driven by a periodic train of optical pulses and coupled to a nuclear spin bath. Using a dynamical, self-consistent, mean-field type approach, we calculated the steady-state dynamic nuclear polarization, as well as its influence on the evolution of the electron spin. Our framework is non-perturbative, applies to nuclei of arbitrary total spin $I$, and includes quadrupolar effects that arise for $I>1 / 2$. We showed that the phenomenon of mode-locking, or DNP-induced frequency-focusing, seen in experiments $[132,133,134,135,136,137,138]$ emerges naturally from our formalism. It can be understood as originating from the structure of the rates for the electron and nuclear spins to flip with one another under the hyperfine interaction. The flip rates vanish when the effective electron precession frequency (including the DNP-driven Overhauser shift) becomes commensurate with the optical pulse train, because in this case the pulses do not interrupt the joint electron-nuclear evolution, and so no polarization is transferred from the electron spin to the nuclei. The vanishing of the flip rates then leads to sharp peaks in the nuclear polarization distribution at magnetization values that satisfy the commensurability condition. Our exact result for the nuclear spin probability distribution in the $I=1 / 2$ case makes this connection explicit, since the distribution depends inversely on the flip rates. In addition to mode-locking, we showed that hyperfine flip-flops also give rise to a net nuclear spin polarization that appears to grow linearly with the number of nuclei.

It is worth considering how the mode-locking peaks determined by the commensurability
condition (Eq. (5.30)) might be modified if we were to go beyond the box model and include a distribution of hyperfine couplings. First we note that, for generic choices of the applied magnetic field, only the nuclei that are closest to the center of the electronic wavefunction contribute to mode-locking. This is because $P(m)$ is concentrated near relatively small values of $m$ (for $I=1 / 2$ this is due to the Gaussian-like factor in Eq. (5.29)), so the smallness of the hyperfine couplings of the nuclei that are farther away cannot be compensated by larger values of $m$ to satisfy the commensurability condition. Small variations in the hyperfine couplings of nuclei close to the center could be incorporated using a "wedding cake" model in which the nuclei are separated into groupings defined by distinct values of the hyperfine coupling. These groupings could be treated as smaller, independent spin baths, each with its own mode-locking condition. Distinct but nearly-equal values of $A$ will give rise to modelocking peaks at almost the same magnetization values $m$, and collectively these closelyspaced peaks will form broader mode-locking features in $P(m)$. The fact that clear signatures of frequency-focusing are seen in experiments [132, 133, 134, 135, 136, 137, 138] suggests that this broadening is a relatively small effect.

Our formalism includes not only hyperfine-driven phenomena, but also quadrupolar effects that can arise for $I>1 / 2$. We found that the importance of quadrupolar interactions depends sensitively on the quadrupolar angle $\theta$ between the applied magnetic field and the principal axis of strain in the dot. For $\theta<\pi / 8$, hyperfine interactions tend to dominate, leading to clear signatures of mode-locking. However, for $\theta \geq \pi / 8$, quadrupole-induced nuclear spin flips begin to dominate, which leads to a suppression of mode-locking and a reduction of the net nuclear polarization. We also showed that quadrupolar effects become more pronounced when the applied magnetic field is increased, because hyperfine flip-flops are suppressed by the increasingly large Zeeman energy mismatch between the electron and nuclei. These effects are clearly visible in the nuclear spin polarization distributions for
both $I=1$ and $I=3 / 2$, and they translate to experimentally detectable signatures that are encoded in the presence or absence of electron spin oscillations in the steady state. Hyperfine flip-flops lead to coherent oscillations in the vicinity of each pulse and halfway between pulses, while quadrupolar interactions act to suppress these oscillations. These signatures offer a potential method to measure the strength of quadrupolar interactions in quantum dots.

The framework we have presented constitutes an efficient, quantitative approach to describing the dynamics of a driven spin coupled to a spin bath. Going forward, it would be interesting to see if some of the simplifying assumptions made here can be relaxed to enhance quantitative accuracy. For example, can we go beyond the box model limit and allow for non-uniform hyperfine couplings, perhaps using a "wedding cake" model in which the electronic wavefunction envelope is approximated by a piecewise-constant function? Such a generalization would also allow for the inclusion of multiple nuclear species, which is relevant for common semiconductor QD compounds such as InGaAs. It would also be interesting to extend this method beyond the independent nuclei approximation, perhaps using a clusterbased approach in which inter-nuclear interactions are included gradually within clusters of increasing size [100, 192]. In terms of applications, our framework could be employed to design driving protocols to achieve desired bath polarization states to either mitigate decoherence or utilize the bath as a quantum memory $[167,168,169,170]$. Finally, we note that the theory we developed is quite general and could be applied to other problems involving a driven system coupled to a quantum bath.

### 5.6 Kraus operators for optical pumping of the electron

The existence of a hierarchy of timescales in mode-locking experiments allows us to first solve for the electron spin dynamics without having to include nuclear spin effects. This is
due to the fact that the nuclear spin dynamics are slow compared to those of the electron. Given that the nuclear spins are the main source of decoherence for the electron, this means we can also neglect electron spin decoherence effects. In addition, the optical pumping and spontaneous emission are fast compared to the pulse period, $\gamma_{e} T_{R} \gg 1$, which ensures that the excited population returns fully to the ground state before the next pulse comes. This allows us to treat the evolution of the electron over each period in terms of a dynamical map that acts only on the electron spin ground state subspace, as in Eq. (5.7).

The Kraus operators, $E_{k}$, that make up the dynamical map can be found by explicitly computing the non-unitary part of the evolution for an arbitrary initial density matrix and comparing the initial and final density matrices [151]. To compute the non-unitary part of the evolution due to the sequence of pulses $H_{c}(t)$, we only need the electronic parts of the full Hamiltonian in Eq. (5.1): $H_{e}(t)=H_{0, e}+H_{c}(t)$. The fact that the pulse is much shorter than the spin precession period allows us to ignore the precession during the action of the pulse. Therefore $|\bar{x}\rangle$ and $|\bar{T}\rangle$ can be considered as an effective two-level system, where the evolution operator due to the pulse in the $|x\rangle,|\bar{x}\rangle,|\bar{T}\rangle$ basis is

$$
U_{p}=\left[\begin{array}{ccc}
1 & 0 & 0  \tag{5.43}\\
0 & u_{\bar{x} \bar{x}} & -u_{\bar{T} \bar{x}}^{*} \\
0 & u_{\bar{T} \bar{x}} & u_{\bar{x} \bar{x}}^{*}
\end{array}\right]
$$

After the pulse, a fraction $\left|u_{\bar{T} \bar{x}}\right|^{2}$ of the population remains in the trion state. We can describe the decay of this population due to spontaneous emission using the Liouville-von Neumann equation with appropriately chosen Lindblad operators $\mathcal{L}: \dot{\rho}=i\left[\rho, H_{0, e}\right]+\mathcal{L}(\rho)$, where the first term includes the Larmor precession of the ground spin states during the decay. Solving this equation for an arbitrary initial state then yields the following Kraus operators in the
$|x\rangle,|\bar{x}\rangle$ basis [151]:

$$
E_{1}=\left[\begin{array}{ll}
1 & 0  \tag{5.44}\\
0 & q
\end{array}\right], \quad E_{2}=\left[\begin{array}{rr}
0 & a_{1} \\
0 & -a_{2}
\end{array}\right], \quad E_{3}=\left[\begin{array}{ll}
0 & 0 \\
0 & \kappa
\end{array}\right]
$$

where $q=u_{\bar{x} \bar{x}} \equiv q_{o} e^{i \phi}, a_{1}=\omega_{e} \sqrt{\left(1-q_{o}^{2}\right) / 2\left(4 \gamma_{e}^{2}+\omega_{e}^{2}\right)}, a_{2}=i \gamma_{e} \sqrt{2} \sqrt{\left(1-q_{o}^{2}\right) /\left(4 \gamma_{e}^{2}+\omega_{e}^{2}\right)}$, and $\kappa=\sqrt{1-q_{o}^{2}-a_{1}^{2}-\left|a_{2}\right|^{2}}$. These Kraus operators guarantee the unity of the trace of the density matrix by satisfying $\sum_{k} E_{k}^{\dagger} E_{k}=\mathbb{1}$. The parameter $q_{o}$ quantifies the amount of population remaining in the spin state $|\bar{x}\rangle$ after the pulse is applied, and $\phi$ is the angle about the $x$ axis by which the pulse rotates the electron spin. These two parameters can be computed given a specific pulse shape, but in the following we leave these parameters arbitrary. Note that these Kraus operators capture the evolution of the electronic spin from the beginning of the pulse until a steady state is reached under the combined action of precession and spontaneous emission. This steady state is reached on timescales large compared to $1 / \gamma_{e}$.

### 5.7 Electron spin steady state

We can use the Kraus operators from above to compute the electron spin steady state. Rather than work directly with the Kraus operators, it is more convenient to switch to the spin vector (SV) representation, especially since finding the steady state requires applying the Kraus operators an infinite number of times. In general, a SV $S$ transforms under non-unitary evolution as follows:

$$
\begin{equation*}
S^{\prime}=Y S+K \tag{5.45}
\end{equation*}
$$

where $Y$ is a matrix that generally both rotates and shrinks the SV , while $K$ corresponds to the non-unital part of the evolution (i.e., a loss or gain of population in the subspace described by $S$ ). If $K$ is nonzero, then a nontrivial steady state is possible. As shown in Ref. [151], for spin $1 / 2$ these quantities can be obtained from the Kraus operators using the following formulas:

$$
\begin{align*}
K_{i} & =\frac{1}{2} \operatorname{Tr} \sum_{k} \hat{\sigma}_{i} \mathcal{E}_{k} \mathcal{E}_{k}^{\dagger}  \tag{5.46}\\
Y_{i j} & =\frac{1}{2} \operatorname{Tr} \sum_{k} \hat{\sigma}_{i} \mathcal{E}_{k} \hat{\sigma}_{j} \mathcal{E}_{k}^{\dagger} \tag{5.47}
\end{align*}
$$

where the $\hat{\sigma}_{i}$ are Pauli matrices. In the case of the mode-locking experiment, the Kraus operators $\mathcal{E}_{k}$ evolve the electron spin over one period, that is, they include both the non-unitary dynamics $\left(E_{k}\right)$ generated by a pulse and also the unitary precession under the magnetic field over time $T_{R}: \mathcal{E}_{k}=E_{k} e^{-i \omega_{e} T_{R} \hat{S}_{z}}$. In concatenating these two parts of the evolution in this way, we are assuming that the drive period is much longer than the time it takes the electron to reach a steady state following the pulse. This in turn requires $T_{R} \gamma_{e} \gg 1$, which is typically satisfied in mode-locking experiments [132, 133, 134, 135, 136, 137, 138]. To find the steady state, it is convenient to combine both $Y$ and $K$ into a single $4 \times 4$ matrix:

$$
\mathcal{Y}_{e}=\left[\begin{array}{cccc}
1 & 0 & 0 & 0  \tag{5.48}\\
K_{x} & Y_{x x} & Y_{x y} & Y_{x z} \\
K_{y} & Y_{y x} & Y_{y y} & Y_{y z} \\
K_{z} & Y_{z x} & Y_{z y} & Y_{z z}
\end{array}\right]
$$

where the evolution of the electron SV over one period is now given by $\mathcal{S}_{e}^{\prime}=\mathcal{Y}_{e} \mathcal{S}_{e}$. Here, the first component of the 4 -component $\mathrm{SV} \mathcal{S}_{e}$ is always fixed to 1 , while the remaining three
components constitute the usual spin $1 / 2 \mathrm{SV}$. In this representation it is easy to see that the steady state $\mathcal{S}_{e}^{s s}=\left(1, S_{e, x}^{s s}, S_{e, y}^{s s}, S_{e, z}^{s s}\right)$ is the eigenvector of $\mathbb{1}-\mathcal{Y}_{e}$ with eigenvalue zero. Transforming the Kraus operators of Eq. (5.44) from the $x$ basis to the $z$ basis, plugging the result into Eq. (5.48), and computing the null vector of $\mathcal{Y}_{e}$ leads to the following steady state electron SV [151]:
$S_{e, x}^{s s}=a_{1}\left(a_{1} q_{o}\left(q_{o}-\cos \phi\right) \cos \left(\omega_{e} T_{R}\right)-i a_{2}\left(q_{o} \cos \phi-1\right) \sin \left(\omega_{e} T_{R}\right)-a_{1} q_{o} \cos \phi+a_{1}\right) \times \mathcal{C}^{-1}$, $S_{e, y}^{s s}=a_{1}\left(a_{1} q_{o}\left(\cos \phi-q_{o}\right) \sin \left(\omega_{e} T_{R}\right)-i a_{2}\left(q_{o} \cos \phi-1\right)\left(\cos \left(\omega_{e} T_{R}\right)-1\right)\right) \times \mathcal{C}^{-1}$, $S_{e, z}^{s s}=a_{1} q_{o} \sin \phi\left(a_{1} \sin \left(\omega_{e} T_{R}\right)-i a_{2}\left(\cos \left(\omega_{e} T_{R}\right)-1\right)\right) \times \mathcal{C}^{-1}$,
where

$$
\begin{align*}
\mathcal{C}= & \left(a_{1}^{2}+q_{o}^{2}-1\right) \cos \left(\omega_{e} T_{R}\right)-a_{1} q_{o} \cos \phi\left[i a_{2} \sin \left(\omega_{e} T_{R}\right)\right. \\
& \left.+a_{1} \cos \left(\omega_{e} T_{R}\right)+a_{1}\right]+i a_{1} a_{2} \sin \left(\omega_{e} T_{R}\right)+\left(a_{1}^{2}-1\right) q_{o}^{2}+1 . \tag{5.50}
\end{align*}
$$

These are the components of the electron SV immediately after each pulse. The steady state at other times during the driving period can be obtained by rotating this vector about the $z$ axis by angle $\omega_{e} T_{R}$ (to account for the Larmor precession).

## Chapter 6

## Magnetic impurities in topological <br> insulators

Band theory is an effective tool for the categorization of several phases of matter. Two common examples of such phases are the conducting states and the insulating states with large energy gaps. A topological insulator (TI) is a special type of electronic material that behaves as an insulator with a band gap in its bulk, but has protected conducting states on its edges. In this chapter first, we will give a very brief introduction to the physical foundations of TIs in Section 6.1. In Sections 6.2-6.12 (partial reprint of "Topological insulator ring with magnetic impurities" by Arian Vezvaee, Antonio Russo, Sophia E. Economou, and Edwin Barnes [193]), we will study the problem of HF interaction between magnetic impurities and the edge states in a TI ring.

### 6.1 Topological band theory and $\mathbb{Z}_{2}$ topological insulators

The birth of TIs came about from applications of topology to the classification of different insulators. The goal of this section is to classify topologically distinct Hamiltonians. The key concept here is that if there is a boundary between two topologically distinct systems,


Figure 6.1: The quantum Hall effect and the cyclotron (chiral) motion of electrons in the bulk-left (along the edge-right).
somewhere along this boundary the topological invariant should change. Put another way, if there are two intersecting insulators with two different topologies, along the boundary of the two the energy gap should close, otherwise the two phases are identical. This closing of the energy gap leads to the appearance of low-energy electronic states in the boundary of the two materials. These gapless states can also be characterized and in fact, the topology of the bulk materials is related to the topology of these gapless edge states as well [194]. This is known as the bulk-boundary correspondence. It is therefore necessary to study the topological invariants of insulating systems. The simplest case of these topological invariants first appeared in the quantum Hall effect $[195,196]$. Semiclassically, we can understand the origin of this effect by considering what happens to a two-dimensional electron gas when an external magnetic field is applied (see Fig. 6.1). The electrons in the bulk of this system precess due to the external magnetic and form Landau levels. At the boundary, however, the electrons 'bounce off' of the edge of the sample and consequently, an electric field leads to the drift of the cyclotron orbits, which in turn leads to a quantized Hall conductivity. We will discuss the role of topology in this system in the following.

### 6.1.1 The Chern invariant

In this section, we introduce the Chern invariant [197] and its physical meaning. Here, as in Chapter 2, we assume a translation symmetry of the system that allows us to use the Bloch theorem wavefunctions $u_{\vec{k}}(\vec{r})$ and the corresponding Berry phase [198]: Due to the intrinsic phase ambiguity of wavefunctions, Bloch states are invariant as,

$$
\begin{equation*}
u_{\vec{k}}(\vec{r}) \rightarrow e^{i \phi(\vec{k})} u_{\vec{k}}(\vec{r}) . \tag{6.1}
\end{equation*}
$$

In analogy to electromagnetic gauge transformations one may define the Berry connection

$$
\begin{equation*}
\vec{A}=-i u_{\vec{k}}^{\dagger}(\vec{r}) \nabla_{\vec{k}} u_{\vec{k}}(\vec{r}) \tag{6.2}
\end{equation*}
$$

which transforms as $\vec{A} \rightarrow \vec{A}+\nabla_{\vec{k}} \phi(\vec{k})$. Notice that $\vec{A}$ is not gauge invariant. However, the Berry phase (which can be thought of as the analog of the magnetic flux), is invariant for any closed loop $C$ :

$$
\begin{equation*}
\oint_{C} \vec{A} \cdot d \vec{k}=\int_{S} d^{2} \vec{k} \nabla \times \vec{A}, \tag{6.3}
\end{equation*}
$$

where we have assumed that $\vec{k}$ is two dimensional. The Berry phase is closely related the Chern invariant: The Chern invariant is the total Berry phase in the Brillouin zone [7]:

$$
\begin{equation*}
\nu=\frac{1}{2 \pi} \int d^{2} \vec{k} \nabla \times \vec{A} \tag{6.4}
\end{equation*}
$$

The Chern invariants can explain the gapless conducting states at the interface of an integer quantum Hall state with a vacuum. These states are protected from backscattering off of
smooth disorders at the edge and are known as chiral modes, in the sense that they only propagate in one direction. Thouless, Kohmoto, Nightingale, and den Nijs (TKNN) showed that the integer in the quantized Hall effect is in fact the Chern number $\nu$ [199]. In terms of the topology of a quantum Hall state with $n=1$ and a trivial insulator (vacuum) with $n=0$, it can be imagined that somewhere along the interface of the two, the band gap has to close because the topological invariant has to change. The corresponding 'gapless' modes are the chiral edge states. Therefore it can be concluded that quantum Hall systems with different Chern numbers $\nu \in \mathbb{Z}$ form different classes of insulators; the so-called $\mathbb{Z}$ classification.

We may revisit the bulk-boundary correspondence with the Chern number concept. If we imagine a domain wall between two bulk insulators with differing Chern numbers $\nu_{L}$ and $\nu_{R}$, the bulk gap must vanish at the interface and the number of corresponding excitations at the interface is the difference in Chern number $\nu_{L}-\nu_{R}$.

### 6.1.2 $\mathbb{Z}_{2}$ topological insulators

The quantum Hall systems are not invariant under time-reversal (TR) symmetry; the chiral edge mode moves in a particular direction. This TR breaking led to an idea by Kane and Mele in 2005 [200] to propose a quantum spin Hall insulator by taking two copies of a quantum Hall system with opposite Chern number and chirality. This ensures the invariance of the system under TR symmetry and in this model, the total Chern number will vanish $\nu_{\downarrow}=-\nu_{\uparrow} \in \mathbb{Z}$. The original idea was to implement this on two layers of honeycomb lattices where the magnetic field of the quantum Hall effect is replaced by the spin-orbit interaction between the two layers. However, the interaction between the two spin layers is ignored. In a follow-up work [201], the same authors showed that in the presence of spin interactions a topological distinction exists between even and odd insulators; i.e., all insulators with even
invariant belong to the same class (known as the topologically trivial insulators), and all insulators with odd invariant belong to another class (known as 2D topological insulators). As such, this is known as the $\mathbb{Z}_{\mathbb{2}}=\mathbb{Z} / 2 \mathbb{Z}$ classification.

The $\mathbb{Z}_{\mathbb{2}}$ classification can be understood qualitatively in terms of Kramer's theorem. The TR symmetry operator for an electron with spin $1 / 2$ is defined as $\mathcal{T}_{1 / 2}=i \sigma_{y} \Theta$ where $\Theta$ is the complex conjugation operator. It is trivial to see that $\mathcal{T}_{1 / 2}^{2}=-1$ for any spin $1 / 2$ particle. As a result, Kramer's theorem states that all eigenstates of a TR invariant Hamiltonian (that is $\left.\left[H, \mathcal{T}_{1 / 2}\right]=0\right)$ are at least twofold degenerate. This can be proven by contradiction: Let us consider a non-degenerate state $|\psi\rangle$ for which $\mathcal{T}_{1 / 2}|\psi\rangle=c|\psi\rangle$. Therefore we should have $\mathcal{T}_{1 / 2}^{2}|\psi\rangle=|c|^{2}|\psi\rangle$, which is contradictory since $|c|^{2} \neq-1$.

To understand how the twofold degeneracy of Kramer's theorem leads to the two classes of trivial and topological insulators, it is helpful to consider Fig. 6.2 where two possible electronic edge states of 2D TR invariant insulators, as a function of the crystal momentum along the edge, are shown. Due to Kramer's theorem, these edge states should be degenerate at $\vec{k}=0$ and $\vec{k}=\pi / a$ (denoted by $\Gamma_{a}$ and $\Gamma_{b}$ in the figure) as these points are invariant under TR. Between these two points, the degeneracy is lifted by the spin-orbit interaction. Depending on how these states reconnect at TR invariant points, we can have trivial insulators or topological insulators: If the reconnection is pairwise (Fig. 6.2(a)), the bands intersect $E_{F}$ an even number of times and the edge states can be eliminated by pushing the bound states away from the gap (a trivial insulator). If the reconnection is anti-pairwise (Fig. 6.2(b)) the bands intersect $E_{F}$ an odd number of times and one cannot move away from the edge states out of the gap (a topological insulator). The topological class of the bulk band structure determines which of these scenarios takes place. Finally, from the bulkboundary correspondence, we can determine that since each intersection at point $k$ is related to $-k$ by TR , the number of Kramers pairs of edge modes of the $\mathbb{Z}_{2}$ invariants across the


Figure 6.2: Two possible reconnection scenarios of Kramer edge states for a TR invariant insulator (only half of the Brillouin zone is shown since the other half is related through TR). (a) Pairwise reconnection which leads to a trivial insulator. (b) Anti-pairwise reconnection which leads to topological insulators. Figure taken from Ref. [7].
interface should be $\Delta \nu \bmod 2$.

### 6.1.3 Realization of 2D topological insulators in $\mathrm{HgTe} / \mathrm{CdTe}$

The original proposal for the realization of TIs was to use graphene. However, due to weak spin-orbit coupling in graphene, attention was taken to materials with strong spin-orbit couplings. Bernevig, Hughes, and Zhang (BHZ) [8] suggested a setup that involves a HgTe layer sandwiched between two CdTe layers, which was later confirmed by König et al. [202]. The band structure of CdTe is similar to those discussed in Chapter 2; the conduction band has $\Gamma_{6}$ symmetry with $s$-type orbitals and the valence band has $\Gamma_{8}$ symmetry with $p$-type orbitals (as before, the split-off $\Gamma_{7}$ band has much lower energy and is therefore ignored). However, the HgTe layer has an inverted band structure: The $p$-like $\Gamma_{8}$ levels have higher energy than the $s$-like $\Gamma_{6}$ levels (Fig. 6.3). BHZ showed that for HgTe thicknesses smaller than a critical value ( $d_{c}=6.3 \mathrm{~nm}$ ), the quantum well structure of $\mathrm{HgTe} / \mathrm{CdTe}$ has a normal band structure since CdTe will dominate. For $d>d_{c}$ the band structure will be inverted


Figure 6.3: (a) Band structure of bulk HgTe and CdTe . (b) The quantum well structure of $\mathrm{HgTe} / \mathrm{CdTe}$ in the normal $\left(d<d_{c}\right)$ regime and inverted $\left(d>d_{c}\right)$ regime. Figure taken from Ref. [8].
since HgTe will dominate. By varying the thickness of the HgTe layer the thickness of the HgTe layer a phase transition between a trivial insulator and a topological insulator occurs since the $s$ and $p$ bands will cross each other at $d_{c}$ without an avoided crossing, as a result of which the energy gap closes.

### 6.2 Magnetic impurities and backscatterings: Motivation

We now turn to studying of the role magnetic impurities in TIs, starting from this section to Section 6.12. The work presented in these sections are a partial reprint of "Topological insulator ring with magnetic impurities" by Arian Vezvaee, Antonio Russo, Sophia E. Economou, and Edwin Barnes [193].

As we discussed in the preceding section, TIs behave as insulators in the bulk while exhibiting conducting helical surface or edge states [200, 201, 203]. TIs are invariant under TR symmetry, and their surface or edge states are topologically protected provided this symmetry remains unbroken [200, 204, 205]. These states have spin and momentum locked orthogonally to each other, and hence states of opposite momentum have opposite spin so that full backscattering cannot occur without a spin-flipping mechanism [204]. One of the best known examples of TIs are HgTe quantum wells, first predicted by Bernevig, Hughes and Zhang [8] and later confirmed in various experiments [202, 206, 207, 208]. However, imperfect conductance has been measured in experiments performed on longer HgTe samples [202, 206, 209, 210], suggesting that TR-violating scatterers such as intrinsic nuclear spins or magnetic impurities may become important in such devices [10, 211, 212, 213, 214, 215, 216, 217, 218, 219, 220]. Similar considerations are also relevant for 3D TI candidates such as $\mathrm{Bi}_{2} \mathrm{Se}_{3}[221,222]$ and
$\mathrm{Sb}_{2} \mathrm{Te}_{3}[221]$, which also include spinful nuclear isotopes and likely carry magnetic impurities as well [223]. Both theoretical and experimental evidence that spinful nuclei lead to not only backscattering of helical modes but also dynamic nuclear polarization has also appeared recently [215, 219, 224]. Despite this progress, it remains challenging to observe the precise role of nuclear spins or magnetic impurities in experiments because of the numerous other factors present in these devices.

In the remainder of the chapter, we investigate the impact of nuclear spins or magnetic impurities coupled to helical edge states in topological insulator nanorings. The introduction of a magnetic flux threading the ring provides an additional control knob to facilitate the study of the helical electron-impurity interaction. Quantum nanorings and disks have drawn a significant amount of attention over the past decade [225, 226, 227, 228], in part because they are ideal systems in which to study quantum interference phenomena such as the AharonovBohm (AB) effect $[229,230,231,232,233]$ and other geometrical phase effects. Persistent currents in quantum rings due to the AB effect [234] were soon proposed after the AB effect itself and were confirmed experimentally [235]. Since then, persistent currents have been one of the most active fields of research in this context [146, 236, 237, 238, 239, 240, 241, 242, 243]. Research on quantum rings is also expanding due to their various applications in spintronics. A few examples include spin entanglement control [244], spin filtering [245], spin beam splitting [246] and spin current pumping [247]. Possible applications to quantum information processing have also been proposed [248]. Additional applications are possible in rings possessing a significant spin-orbit interaction [249, 250, 251, 252], and this has in part motivated recent investigations of TI quantum rings both theoretically [253] and experimentally [254]. Furthermore, ring-like distribution of helical edge states in the context of TI quantum dots have been observed as well $[255,256]$. The AB effect in such systems has been worked out theoretically and observed experimentally in both 2D and 3D TIs [257, 258, 259, 260].

The bound-state spectrum of clean 2D TI quantum rings based on the BHZ model of HgTe quantum wells, in the presence of a magnetic field, has been calculated [261], but the effect of a magnetic impurity on this spectrum remains an open problem.

Here, we calculate the spectrum of helical edge states on a 2D TI ring coupled to a nuclear spin or magnetic impurity of arbitrary spin and with a magnetic flux threading the ring. Using a generalized time-reversal symmetry under which both the electronic and impurity spins are reversed, along with spin conservation, we derive a universal formula for the spectrum as a function of magnetic field that depends only on the amplitude of transmission through the impurity. Thus our results apply for any spatial profile of the electron-impurity interaction region. We show that the solution for an arbitrary-spin impurity can be built up using solutions for spin $1 / 2$ and spin 1 impurities, which we obtain explicitly. We show that, in a certain energy regime, the spectrum becomes effectively independent of the magnetic flux for sufficiently strong impurity coupling, leading to sizable energy gaps. In addition, we calculate the entanglement entropy of the helical states as a function of magnetic field, finding that the electronic and impurity spins become maximally entangled near the spectral gaps.

The rest of the chapter is structured as follows. In Section 6.3 we describe the TI ringimpurity model and discuss the symmetries present in this model and their consequences. We show that a generalized version of time-reversal symmetry allows us to decompose the model for an arbitrary-spin impurity into decoupled spin $1 / 2$ and spin 1 sectors. Following this result, we solve the scattering problem for these two special cases in Sections 6.4 and 6.5, respectively. In each case, we obtain a universal formula for the energy spectrum in terms of the transmission amplitude. In Section 6.6, we discuss how our results generalize to the case of several impurities on the ring. In Section 6.7, we calculate the spin current and entanglement entropy of the system. In Section 6.8, we study the dependence of the
spectrum on the ring geometry and impurity couplings and obtain approximate formulas for energy bandwidths and gaps for a square impurity potential. The last three sections contain additional technical details pertaining to our derivations.

### 6.3 Hamiltonian and symmetries

### 6.3.1 TI ring in a magnetic field

We take the non-interacting Hamiltonian on the ring to be the effective Hamiltonian for ring-like TI quantum dot edge states, where the edge states are distributed near the dot boundary [256]:

$$
\begin{equation*}
H_{0}=v_{0} \hat{p}_{y} \sigma_{z}, \tag{6.5}
\end{equation*}
$$

where $v_{0}$ is the Fermi velocity, $\hat{p}_{y}$ is the (angular) momentum operator, and $\sigma_{z}$ acts on the spin subspace. The eigenstates of this Hamiltonian are spin-momentum locked plane waves: $\psi_{+}=e^{i p_{y} y}|\uparrow\rangle, \psi_{-}=e^{-i p_{y} y}|\downarrow\rangle$, where for a ring of circumference $d$, the momenta are quantized: $p_{y}=2 \pi n / d$. We model the interaction between the electron spin and an impurity spin $I$ as

$$
\begin{equation*}
H_{S, I}=F(y)\left[A^{z} \sigma_{z} I_{z}+A^{\perp}\left(\sigma_{-} I_{+}+\sigma_{+} I_{-}\right)\right] \tag{6.6}
\end{equation*}
$$

where we follow Ref. [10] and use an interaction that has been averaged over impurity spin locations within the edge state. This interaction occurs over a finite region of width $w$ on the ring, with a specific spatial profile set by $F(y)$, and we allow for the longitudinal and transverse spin-spin coupling constants $A^{z}$ and $A^{\perp}$ to be different. This interaction breaks TR symmetry and provides a mechanism for backscattering that is assisted by electronimpurity spin flip-flops generated by the transverse terms in Eq. (6.6).

In the presence of an applied magnetic flux $\Phi_{B}$ that threads the ring (see Fig. 6.4), the momentum operator in Eq. (6.5) is shifted according to: $\hat{p}_{y} \rightarrow \hat{p}_{y}+p_{B}$, where $p_{B} d=$ $2 \pi \Phi_{B} / \Phi_{0}$, where $\Phi_{0}$ is the magnetic flux quantum. This shift can be effectively undone by introducing an ansatz for the eigenstate wavefunction that includes a global phase:

$$
\begin{equation*}
\psi_{B}(y)=e^{-i p_{B} y} \psi(y) \tag{6.7}
\end{equation*}
$$

Here, $\psi(y)$ is a solution to the Hamiltonian without the vector potential, Eq. (6.5), but now with a nontrivial boundary condition imposed on it as required to ensure single-valuedness of the wavefunction, $\psi_{B}(0)=\psi_{B}(d)$ :

$$
\begin{equation*}
\psi(d)=e^{i p_{B} d} \psi(0)=e^{2 \pi i \Phi_{B} / \Phi_{0}} \psi(0) \tag{6.8}
\end{equation*}
$$

The first equality resembles a Bloch constraint in one dimension, with $d$ interpreted as the lattice spacing and $p_{B}$ as the crystal momentum. Thus, we can treat the impurity scattering problem with nonzero magnetic flux as though we are solving a Kronig-Penney model with Hamiltonian $H=H_{0}+H_{S, I}$ without magnetic flux. The magnetic flux dependence is restored by replacing the crystal momentum by $p_{B}$. This observation reflects the general connection between the AB problem on a ring with a non-uniform potential and the Kronig-Penney model [234, 262, 263]. We may thus think of the energy spectrum dependence on $p_{B}$ or $\Phi_{B}$ as an effective band structure.

### 6.3.2 Spin conservation and generalized time-reversal symmetry

Although TR symmetry is broken by the coupling to the impurity, Eq. (6.6), this interaction does preserve a generalized time-reversal (GTR) symmetry that flips both the electronic and


Figure 6.4: A schematic illustration of a TI ring of circumference $d$ with a single impurity of width $w$. The edge states are indicated with arrows and the magnetic field associated with the $A B$ effect is localized inside the ring.
impurity spins. By exploiting this symmetry along with conservation of total spin, we can achieve a better understanding of the boundary matching problem that we must solve in order to obtain the energy spectrum. In fact, the consequences of these two symmetries together lead to a universal result for the energy spectrum in terms of only one variable, which we take to be the amplitude of transmission through the impurity.

The total Hamiltonian, $H=H_{0}+H_{S, I}$, commutes with the total spin operator $J_{z}=S_{z}+I_{z}$. Thus, the total wavefunction describing both the electron and impurity breaks into sectors labeled by $J_{z}$. Two of these sectors (the ones corresponding to maximum and minimum $J_{z}$ ) are one-dimensional, while the rest are two-dimensional, as is illustrated in Fig. 6.5.

To understand the consequence of GTR, first note that this operation maps the electronimpurity spin state $|\uparrow\rangle|n\rangle$ to $|\downarrow\rangle|-n\rangle$. The former state has $J_{z}=n+1 / 2$, while the latter has $J_{z}=-n-1 / 2$, so we see that GTR mixes different total spin sectors. Importantly, it mixes only these two sectors, so that the Hilbert space breaks up into "blocks", where each


Figure 6.5: Decomposition of the wavefunction into its sectors and blocks (here we suppress the spatial dependence). $|\uparrow\rangle$ and $|\downarrow\rangle$ are electron spin states, and $|n\rangle$ labels impurity spin states. Two components with the same total spin $J_{z}$ that form one sector are connected with blue arrows. Red arrows show the components that mix under GTR. We see that the two sectors $J_{z}=n+1 / 2$ and $J_{z}=-n-1 / 2$ mix and form the block labeled by $J_{z}=|n+1 / 2|$.
block consists of two sectors of opposite $J_{z}$ and thus can be labeled by $\left|J_{z}\right|$ (Fig. 6.5) ${ }^{1}$. Of course, when $J_{z}=0$ there is only a single sector in the block; this type of block only occurs for half-integer-spin impurities. In terms of scattering eigenstates, GTR relates an eigenstate incoming from one side of the impurity to an eigenstate incoming from the opposite side, and it allows us to solve for these eigenstates by separately solving the matching problem in each block. In the next two sections, we exploit this fact to derive general relations between the reflection and transmission amplitudes for a single impurity. We then use these relations to obtain a universal formula for the energy spectrum; we find that the same formula arises in every block regardless of the value of $\left|J_{z}\right|$. Thus, the entire spectrum for an arbitrary-spin impurity can be obtained from this formula after the transmission amplitudes in each sector are calculated for a given interaction region profile $F(y)$.

[^2]
### 6.4 Spin-1/2 impurity

In this section, we solve the scattering problem for a single impurity with spin $m=1 / 2$. We begin with ansatz eigenstate wavefunctions on the left (L) and right (R) sides of the interaction region:

$$
\begin{align*}
& \psi_{L}(y)=e^{i p y}\left(\begin{array}{l}
\alpha \\
\beta \\
0 \\
0
\end{array}\right)+e^{-i p y}\left(\begin{array}{c}
0 \\
0 \\
\alpha^{\prime} \\
\beta^{\prime}
\end{array}\right), \\
& \psi_{R}(y)=e^{i p y}\left(\begin{array}{c}
\alpha^{\prime \prime} \\
\beta^{\prime \prime} \\
0 \\
0
\end{array}\right)+e^{-i p y}\left(\begin{array}{c}
0 \\
0 \\
\alpha^{\prime \prime \prime} \\
\beta^{\prime \prime \prime}
\end{array}\right) . \tag{6.9}
\end{align*}
$$

Here, the basis states for the spinors are (from top to bottom) $|\uparrow, 1 / 2\rangle,|\uparrow,-1 / 2\rangle,|\downarrow, 1 / 2\rangle$, $|\downarrow,-1 / 2\rangle$, where the arrows denote the electron spin, and $\pm 1 / 2$ refers to the impurity spin. The coefficients $\alpha$ and $\beta$ specify the "initial" impurity spin state for an eigenstate incoming from the left, while $\alpha^{\prime \prime \prime}$ and $\beta^{\prime \prime \prime}$ give the initial impurity state for an eigenstate incoming from the right. The remaining coefficients $\alpha^{\prime}, \beta^{\prime}, \alpha^{\prime \prime}, \beta^{\prime \prime}$ correspond to reflection or transmission coefficients, depending on the direction from which the incident wave originates.

The standard approach to solving this type of scattering problem is to also solve for the wavefunction inside the interaction region and to then enforce continuity of the wavefunction at the boundaries of this region. Imposing the additional constraint, Eq. (6.8), generally gives rise to a relation between the energy and the magnetic flux, leading to the effective band structure $E\left(\Phi_{B}\right)$.


Figure 6.6: Scattering off a single impurity (depicted here as a square barrier, which is assumed to carry nonzero spin $I$ ). Incident waves from the left (with coefficient $A$ ) and right (with coefficient $B$ ) and their corresponding transmitted and reflected waves are shown. We assume the impurity is in an eigenstate of $I_{z}$ with eigenvalue $|n|<I$. Each arrow corresponds to one component of the wavefunction (on left or right) with a particular spin state. Combinations of these waves form the wavefunctions in Eq. (6.10).

Here, we describe a more economical approach to obtaining the energy spectrum that yields a universal formula for arbitrary-spin impurities with the help of GTR symmetry. We first demonstrate this for a spin $1 / 2$ impurity and postpone discussion of larger spins to the next section. First note that the wavefunction spinor components with $J_{z}= \pm 1$ do not mix with each other or with any other components under the electron-impurity interaction, Eq. (6.6), and simply acquire phase factors as a consequence of the impurity. These phases have no bearing on the energy spectrum, and thus the $\left|J_{z}\right|=1$ block can be safely neglected. (The impact of GTR on these phases is discussed in Section 6.10.) We therefore focus only on the $J_{z}=0$ block. If our impurity scattering problem were formulated in an infinite 1D channel instead of on a finite ring, then every left-incoming eigenstate would be degenerate with a right-incoming state, and any superposition of these would also be an eigenstate. On the
left and right side of the impurity barrier (see Fig. 6.6), we could then write

$$
\psi(0)=\left[\begin{array}{c}
A  \tag{6.10}\\
r_{\rightarrow} A+t_{\leftarrow} B
\end{array}\right], \quad \psi(d)=\left[\begin{array}{c}
t_{\rightarrow} A+r_{\leftarrow} B \\
B
\end{array}\right],
$$

where $A$ and $B$ are the coefficients of the left-incoming and right-incoming states in the superposition, and we only keep the $J_{z}=0$ wavefunction spinor components $|\uparrow,-1 / 2\rangle$ and $|\downarrow, 1 / 2\rangle$. The subscript arrows on the reflection and transmission amplitudes indicate the direction of the corresponding incoming wave. If we now return to the ring geometry by identifying $y=0$ and $y=d$ and imposing the single-valuedness constraint, Eq. (6.8), then we find that only one of these superpositions is a valid eigenstate, and the magnetic flux is determined by the scattering amplitudes:

$$
\begin{align*}
B / A & =\frac{1-r_{\leftarrow} r_{\rightarrow}-t_{\leftarrow} t_{\rightarrow} \pm \sqrt{\left(1-r_{\leftarrow} r_{\rightarrow}+t_{\leftarrow} t_{\rightarrow}\right)^{2}-4 t_{\leftarrow} t_{\rightarrow}}}{2 r_{\leftarrow} t_{\leftarrow}},  \tag{6.11}\\
e^{2 \pi i \Phi_{B} / \Phi_{0}} & =\frac{1-r_{\leftarrow} r_{\rightarrow}+t_{\leftarrow} t_{\rightarrow} \pm \sqrt{\left(1-r_{\leftarrow} r_{\rightarrow}+t_{\leftarrow} t_{\rightarrow}\right)^{2}-4 t_{\leftarrow} t_{\rightarrow}}}{2 t_{\leftarrow}} . \tag{6.12}
\end{align*}
$$

The two solutions distinguished by the sign in front of the square root correspond to currents circulating in opposite directions around the ring, as we discuss further in Sec. 6.7. These two solutions are degenerate and are related to each other by GTR symmetry. It is important to note that Eq. (6.12) holds for any barrier shape $F(y)$; the only assumption we have made is that the barrier vanishes at $y=0$ and $y=d$. Once the scattering amplitudes are obtained for a given barrier shape, Eq. (6.12) can be used to obtain the corresponding energy spectrum. In the absence of GTR or any other symmetry, the reflection and transmission amplitudes $r_{\rightarrow}$, $r_{\leftarrow}, t_{\rightarrow}, t_{\leftarrow}$ would all be independent of each other (aside from the normalization condition). However, as explained in detail in Section 6.10, GTR symmetry imposes two relations among
these amplitudes:

$$
\begin{equation*}
t_{\leftarrow}=t_{\rightarrow} \equiv t \equiv|t| e^{i \phi_{t}}, \quad r_{\rightarrow} r_{\leftarrow}=\left(|t|^{2}-1\right) e^{2 i \phi_{t}} . \tag{6.13}
\end{equation*}
$$

These relations dramatically simplify Eq. (6.12) and allow us to express the magnetic flux in terms of only the transmission amplitude:

$$
\begin{equation*}
e^{2 \pi i \Phi_{B} / \Phi_{0}}=\cos \phi_{t} /|t| \pm \sqrt{\left(\cos \phi_{t} /|t|\right)^{2}-1} \tag{6.14}
\end{equation*}
$$

Since the left-hand-side is a pure phase, the right-hand-side must also be a pure phase in order for a solution to exist. This then leads to the following condition for a state to exist at a given energy:

$$
\begin{equation*}
\cos ^{2} \phi_{t}<|t|^{2} \tag{6.15}
\end{equation*}
$$

Energy ranges where $t(E)$ violates this condition correspond to gaps in the spectrum. In ranges where states exist, the two different branches of the square root in Eq. (6.14) simply correspond to the fact that the energy is independent of the sign of the magnetic flux: $E\left(\Phi_{B}\right)=E\left(-\Phi_{B}\right)$. For reasons that will become clear in the next section, we refer to this feature as a 2-fold "flux degeneracy", i.e., the number of distinct values of the flux $\Phi_{B}$ that give rise to a state of a given energy $E$. Fig. 6.7(a) shows an example band structure obtained from this formula for the case of a square barrier using typical experimental parameters. The scattering amplitudes for the square barrier are derived in Section 6.11. One salient feature of the spectrum is that band edges always occur at half-integer multiples of the flux quantum. This generally holds for any spin $1 / 2$ impurity regardless of couplings or potential shape and follows directly from the band edge condition $\cos \phi_{t}= \pm|t|$ and Eq. (6.14). We will see in the next section that band edges can occur at other values of $\Phi_{B}$ for higher-spin impurities. The most striking consequence of the impurity is the occurrence of nearly flat bands in the vicinity of $E=-A^{z}=-0.05 \mathrm{eV}$, with gaps of size 8 meV between them. We explain
the origin of these bands and study their dependence on the impurity couplings and ring geometry in Sec. 6.8. We further analyze the spectrum for a spin $1 / 2$ impurity quantitatively for a range of realistic device parameters in the same section.

The constraint in Eq. (6.15) can be visualized in terms of the complex $t$ plane (Fig. 6.7(b)). Scanning through values of the energy corresponds to tracing out a curve in this plane, and whenever the curve enters one of the yellow regions, which indicate values of $t$ that violate Eq. (6.15), a gap occurs in the spectrum. Large loops give rise to dispersive bands, while the flat bands correspond to loops concentrated close to the origin. If the parametric curve tangentially touches the yellow region at $\operatorname{Re}[t]= \pm 1$, then a band touching point appears in the spectrum at $\Phi_{B}=0$ or $\pm 1 / 2$. Such points can only occur at values of the energy for which $|t|=1$, i.e., for which the impurity is effectively transparent. We show in Sec. 6.8 that a discrete set of energies satisfy this condition in the case of a square impurity barrier.

### 6.5 Arbitrary-spin impurity

In this section, we solve the scattering problem for an arbitrary-spin impurity by following an approach that is similar to what we used for a spin $1 / 2$ impurity in the previous section. Since GTR symmetry allows us to solve the problem in each block of the Hilbert space separately, we only need to solve the case of a spin 1 impurity to obtain the solution in the general case. The spin 1 case has only one block consisting of two GTR-coupled sectors. Impurities with larger integer spins will break into a series of blocks, all with the same structure as the spin 1 case, allowing us to solve these cases in terms of multiple copies of the spin 1 impurity solution. Moreover, half-integer spins will also reduce to one spin $1 / 2$ block (with $J_{z}=0$ ) and several spin 1 blocks (with $J_{z} \neq 0$ ). As a result, we can solve the problem for an arbitrary-spin impurity by combining the solutions for the spin $1 / 2$ and spin


Figure 6.7: (a) Energy spectrum (solid blue lines) as a function of magnetic flux for a spin $1 / 2$ impurity with square potential. The dashed lines indicate the spectrum without the impurity. (b) Parametric plot of the transmission amplitude as a function of energy. Yellow regions indicate values where the condition $\cos ^{2} \phi_{t}<|t|^{2}$ does not hold and thus correspond to gaps in the spectrum. Colored dots map certain energies of the energy spectrum to points on the parametric plot. The parameters are $d=1000 \AA, w=130 \AA, A^{z}=A^{\perp}=0.05 \mathrm{eV}$ $[9], v_{0}=2.4 \mathrm{eV} \AA[10]$.

1 cases.

Out of the six components of the electron-impurity wavefunction for a spin 1 impurity, the two with maximal $\left|J_{z}\right|$ (i.e., $J_{z}= \pm 3 / 2$ ) are again decoupled from each other and from all other components, while the remaining four components form the block with $\left|J_{z}\right|=1 / 2$. From now on we distinguish all variables of this block with $\pm$ signs to indicate the sector to which it belongs according to the sign of $J_{z}$. As in the previous section, we consider leftincoming waves in each sector with coefficients $A^{ \pm}$superposed with right-incoming waves with coefficients $B^{ \pm}$. The analog of Eq. (6.10) becomes

$$
\psi(0)=\left[\begin{array}{c}
A^{+}  \tag{6.16}\\
A^{-} \\
r_{\rightarrow}^{+} A^{+}+t_{\leftarrow}^{+} B^{+} \\
r_{\rightarrow}^{-} A^{-}+t_{\leftarrow}^{-} B^{-}
\end{array}\right], \quad \psi(d)=\left[\begin{array}{c}
t_{\rightarrow}^{+} A^{+}+r_{\leftarrow}^{+} B^{+} \\
t_{\rightarrow}^{-} A^{-}+r_{\leftarrow}^{-} B^{-} \\
B^{+} \\
B^{-}
\end{array}\right],
$$

where the basis states are now $|\uparrow, 0\rangle,|\uparrow,-1\rangle,|\downarrow, 1\rangle,|\downarrow, 0\rangle$. Since $\psi(0)$ and $\psi(d)$ are each essentially just two copies of the analogous expressions in the spin $1 / 2$ case, Eq. (6.10), when we impose the single-valuedness condition, Eq. (6.8), we obtain two copies of the band structure equation, Eq. (6.12), one for each sector labeled by $\pm$.

We may again invoke GTR symmetry to simplify these expressions using relations between the scattering amplitudes. However, since GTR now couples two distinct sectors, this process is different from the spin $1 / 2$ case, for which there was only a single sector. The details are given in Section 6.12. The resulting relations among the amplitudes are as follows:

$$
\begin{align*}
& t_{\rightarrow}^{+}=t_{\leftarrow}^{-}, \quad t_{\leftarrow}^{+}=t_{\rightarrow}^{-}, \quad r_{\rightarrow}^{+}=r_{\rightarrow}^{-}, \quad r_{\leftarrow}^{+}=r_{\leftarrow}^{-}, \\
& \left|t_{\rightarrow}^{ \pm}\right|=\left|t_{\leftarrow}^{ \pm}\right| \equiv|t|, \quad\left|r_{\rightarrow}^{ \pm}\right|=\left|r_{\leftarrow}^{ \pm}\right| \equiv|r|, \quad|r|^{2}+|t|^{2}=1, \tag{6.17}
\end{align*}
$$


(b)



Figure 6.8: Spin 1 impurity. (a) Energy spectrum as a function of magnetic flux. Each of the two total spin sectors yields a different state (distinguished by red and blue) at each energy. (b) Zoom-in of the spectrum shown in (a) (upper panel) and the condition for states to exist (lower panel). States occur at energies where the transmission amplitudes satisfy $\left|\cos \phi_{t}^{\circ}\right|<|t|$. (c) Parametric plot of transmission amplitude $t=|t| e^{i \phi_{t}^{\circ}}$ for the energy range shown in (b). The colored dots indicate the corresponding energies shown in (b). The parameters are $d=1000 \AA, w=130 \AA, A^{z}=A^{\perp}=0.05 \mathrm{eV}[9], v_{0}=2.4 \mathrm{eV} \AA[10]$.
and additionally we have

$$
\begin{equation*}
\phi_{t^{ \pm}}^{\circ}=\phi_{r^{ \pm}}^{\circ}+\pi / 2, \tag{6.18}
\end{equation*}
$$

where we have defined,

$$
\begin{equation*}
\phi_{x}^{\circ} \equiv\left(\phi_{x \rightarrow}+\phi_{x \leftarrow}\right) / 2, \quad \text { for } \quad x=r^{ \pm}, t^{ \pm}, \tag{6.19}
\end{equation*}
$$

where $\phi_{x \rightarrow}, \phi_{x \leftarrow}$ are the phases of the corresponding scattering amplitudes. It follows from Eq. (6.17) that $\phi_{t^{+}}^{\circ}=\phi_{t^{-}}^{\circ}$, which allows us to drop the sector labels $\pm$ in these quantities: $\phi_{t^{+}}^{\circ}=\phi_{t^{-}}^{\circ} \equiv \phi_{t}^{\circ}$. These relations allow us to simplify Eq. (6.12) down to the result

$$
\begin{equation*}
e^{2 \pi i \frac{\Phi_{B}}{\Phi_{0}}}=e^{i\left(\phi_{t_{\vec{~}}}-\phi_{t_{\leftarrow}^{\text {土 }}}\right) / 2}\left[\cos \phi_{t}^{\circ} /|t| \pm \sqrt{\left(\cos \phi_{t}^{\circ} /|t|\right)^{2}-1}\right] . \tag{6.20}
\end{equation*}
$$

The overall phase factor in this expression depends on the sector as can be seen from Eq. (6.17); the two phases in fact differ only by a sign: $\phi_{t_{\rightarrow}^{+}}-\phi_{t \_}=-\left(\phi_{t_{\hookrightarrow}}-\phi_{t \leftarrow}\right)$. An important consequence of the overall phase is that there is now a 4-fold flux degeneracy instead of a 2-fold degeneracy like we have for a spin $1 / 2$ impurity. This 4 -fold degeneracy comes from the two sectors and the two branches of the square root in Eq. (6.20). Because the overall phase differs by only a sign between the two sectors, it remains true that $E\left(-\Phi_{B}\right)=E\left(\Phi_{B}\right)$, or in other words the spectrum remains symmetric about $\Phi_{B}=0$. An example spectrum for a spin 1 impurity is shown in Fig. 6.8(a), where the additional degeneracy is evident. Also notice that, unlike in the spin $1 / 2$ impurity case, the band edges can occur at arbitrary values of the flux; this is due to the extra phase $e^{i\left(\phi_{t \rightarrow}-\phi_{t_{\rightleftarrows}^{ \pm}}\right) / 2}$ appearing in Eq. (6.20). We also note that flat bands are again apparent in the region near $E=-A^{z}$.

Also notice the similarity of Eq. (6.20) to Eq. (6.14). Aside from the overall phase factor, the only other difference is that $\phi_{t}$ has been replaced by the average phase $\phi_{t}^{\circ}$. Eq. (6.14) can be understood as a special case of Eq. (6.20) where the self-duality of the $J_{z}=0$ sector

GTR symmetry enforces $t_{\rightarrow}=t_{\leftarrow}$, so that the overall phase factor in Eq. (6.20) vanishes, and $\phi_{t}^{\circ}$ reduces to $\phi_{t}$.

All of our analysis here was based only on the fact that two sectors are mixed by GTR symmetry (except when $J_{z}=0$ where there is only one self-symmetric sector). This is true for any value of $J_{z}$ so that in fact Eq. (6.20) holds regardless of the spin of the impurity. Thus we conclude that the flux degeneracy for an impurity of $\operatorname{spin} m$ is equal to twice the number of distinct nontrivial sectors (i.e., those with $\left|J_{z}\right| \neq m+1 / 2$ ), since each such sector contributes two solutions corresponding to the two branches of the square root in Eq. (6.20). Hence the flux degeneracy is $2(2 m+1)-2=4 m$.

Note that even though the spectrum formula, Eq. (6.20), holds regardless of the spin of the impurity, the resulting energy spectra still depend sensitively on the spin and potential of the impurity since these details strongly affect the transmission amplitudes (i.e., the $t$ 's) that enter this formula. These amplitudes are determined by diagonalizing of the Hamiltonian, which depends on the impurity couplings and potential. As an example, consider a spin $3 / 2$ impurity. In this case, there are three sectors ( $J_{z}=0$ and $J_{z}= \pm 1$ ), and each produces a unique spectrum formula condition like Eq. (6.20). To obtain the spectrum, it is necessary to diagonalize the total Hamiltonian in each of the two blocks ( $J_{z}=0$ and $\left|J_{z}\right|=1$ ), extract the transmission amplitudes, and plug them into Eq. (6.20).

### 6.6 Multiple impurities

To treat the case of $N$ impurities on the ring, we can proceed in the same way as for a single impurity. In particular, we can begin by writing down ansatz wavefunctions at $y=0$ and $y=d$, each of which now contains $2(2 I+1)^{N}$ spinor components assuming each impurity has the same total spin $I$. The Hilbert space again divides into sectors labeled by total


Figure 6.9: Energy spectrum for two spin $1 / 2$ impurities as a function of magnetic flux for parameters $d=1000 \AA, A^{z}=0.05 \mathrm{eV}, A^{\perp}=0.05 \mathrm{eV}, w=130 \AA$, and $v_{0}=2.4 \mathrm{eV} \AA$.
spin $J_{z}$, where now the dimensions of the sectors $\mathcal{D}_{J_{z}}$ depend on $J_{z}$. In each sector, we take the ansatz wavefunctions to be superpositions of left-incoming and right-incoming states, and we express these wavefunctions in terms of reflection and transmission coefficients as in Eq. (6.18). We then apply the single-valuedness condition, Eq. (6.8), separately in each sector. Doing so will yield a polynomial in $e^{2 \pi i \Phi_{B} / \Phi_{0}}$ for each sector, where the order of this polynomial is the dimension of that sector, $\mathcal{D}_{J_{z}}$. As an example, consider $N$ spin $1 / 2$ impurities, for which the dimension of each sector is the binomial coefficient

$$
\begin{equation*}
\mathcal{D}_{J_{z}}=\binom{N+1}{J_{z}+\frac{N+1}{2}} . \tag{6.21}
\end{equation*}
$$

For instance for $N=2$, the number of states with $J_{z}=+1 / 2$ is $\binom{3}{2}=3$, which means that we have to solve a cubic equation in order to find the spectrum (recall that for one impurity, the resulting polynomial was quadratic and led to Eq. (6.14)). As we add more impurities, the order of this polynomial grows exponentially, and it quickly becomes necessary to solve for the spectrum numerically. An example of a spectrum for two spin $1 / 2$ impurities (both with square potentials of equal size $w$ and with equal couplings) computed in this way is shown in Fig. 6.9. In this case, there is up to a six-fold flux degeneracy depending on the energy. Unlike in the case of a single impurity, here the spectrum no longer exhibits flat bands in the vicinity of $E=-A^{z}$.

Notice that for several impurities on the ring a Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction mediated by impurities can lead to an indirect exchange among the impurities. At low temperature this effect leads to ferromagnetic ordering of impurities [213, 214, 221, 265, 266, 267, 268, 269]. However, for our case, such RKKY couplings will be small due to the mesoscopic separation between the impurities on the ring. For example, for two impurities on the ring with circumference $d=1000$ Athe separation between between the two impurities will be $500 \AA$. This will lead to an indirect exchange coupling between the
two impurities in the order of $10^{-4}-10^{-5} \mathrm{eV}[213,214]$. Since this coupling is much smaller than the direct coupling between the impurities and the edge states $\left(A^{z}=A^{\perp}=0.05 \mathrm{eV}\right)$ we can safely neglect this interaction.

### 6.7 Spin current and entanglement entropy

### 6.7.1 Probability and spin currents

In this section, we show that the symmetries of the TI ring-impurity system also lead to a universal formula for the ratio of spin and probability currents. The probability current is given by

$$
\begin{equation*}
j_{p}=v_{0} \psi(y)^{\dagger} \sigma_{z} \psi(y) \tag{6.22}
\end{equation*}
$$

while the spin current is

$$
\begin{equation*}
j_{s}=\left(v_{0} / 2\right) \psi(y)^{\dagger} \psi(y) \tag{6.23}
\end{equation*}
$$

In the case of a spin $1 / 2$ impurity, it is straightforward to find the ratio of these two quantities using the wavefunction ansatzes in Eq. (6.10) in conjunction with the single-valuedness condition, Eq. (6.8):

$$
\begin{equation*}
j_{s}=j_{p} \frac{1}{2} \frac{\left|1-e^{2 \pi i \Phi_{B} / \Phi_{0}} t\right|^{2}+|r|^{2}}{\left|1-e^{2 \pi i \Phi_{B} / \Phi_{0}} t\right|^{2}-|r|^{2}} . \tag{6.24}
\end{equation*}
$$

Note that this expression is independent of the coefficients $A$ and $B$ that we introduced in Eq. (6.10). In this expression, $e^{2 \pi i \Phi_{B} / \Phi_{0}}$ also depends on $r$ and $t$, and therefore the right hand side of the equation only depends on energy. For a spin 1 impurity (or more generally for one block of a larger-spin impurity), a similar expression can be derived (see Section 6.12):

$$
\begin{equation*}
j_{s}=j_{p} \frac{1}{2} \frac{\left|1-e^{2 \pi i \Phi_{B} / \Phi_{0}} t_{\leftarrow}^{+}\right|^{2}+|r|^{2}}{\left|1-e^{2 \pi i \Phi_{B} / \Phi_{0}} t_{\leftarrow}^{+}\right|^{2}-|r|^{2}} . \tag{6.25}
\end{equation*}
$$



Figure 6.10: Comparison of the energy spectrum (right) with the electron-impurity entanglement entropy (left) for a spin $1 / 2$ impurity with square potential. Near the spectral gaps, the entanglement reaches its maximum possible value of $\log 2$. The parameters are $d=1000$ $\AA, w=130 \AA, A^{z}=A^{\perp}=0.05 \mathrm{eV}[9], v_{0}=2.4 \mathrm{eV} \AA[10]$.

Although it appears that the right-hand side depends on the sector (i.e., on total spin $J_{z}$ ), this is in fact not the case, as is shown in Section 6.12. Hence, the currents in both sectors of the block are identical. A detailed discussion of spin current pumping in TIs can be found in Ref. [270]. We are assuming that the magnetic field is fully localized inside the ring so that there is no Zeeman interaction for either the electronic or impurity spins. It is readily apparent from Eqs. (6.24) and (6.25) that the ratio of the currents can be controlled directly by adjusting the magnetic flux.

### 6.7.2 Entanglement entropy of nuclear spin and electron

Next, we analyze the entanglement between the electron and impurity as a function of magnetic flux. We can define a position-independent entanglement entropy between electron and impurity spins in the following way. We begin by writing the TI ring-impurity eigenstates
as

$$
\begin{equation*}
|\Psi\rangle=\int d y \sum_{i j} \psi_{i j}(y)|y, i, j\rangle, \tag{6.26}
\end{equation*}
$$

where the index $i$ denotes the electron spin state and $j$ the nuclear spin state. The full density matrix is given by $\rho_{I, e}=|\Psi\rangle\langle\Psi|$. After tracing out the electronic spin, $\rho_{I}=\sum_{i}\langle i| \rho_{I, e}|i\rangle$, this quantity will only depend on the impurity spin and the electron position, which we integrate out:

$$
\begin{equation*}
\rho_{I}=\sum_{i} \int d y \sum_{j, j^{\prime}} \psi_{i j}(y) \bar{\psi}_{i j^{\prime}}(y)|j\rangle\left\langle j^{\prime}\right| . \tag{6.27}
\end{equation*}
$$

Since the spin states $|\uparrow, 1 / 2\rangle$ and $|\downarrow,-1 / 2\rangle$ do not mix with other states, we drop them and focus on the spin states in the $J_{z}=0$ sector:

$$
\rho_{I}=\left(\begin{array}{cc}
\int_{0}^{d} d y\left|\psi_{\uparrow,-1 / 2}\right|^{2} & 0  \tag{6.28}\\
0 & \int_{0}^{d} d y\left|\psi_{\downarrow, 1 / 2}\right|^{2}
\end{array}\right)
$$

To simplify this result further, notice that the probability current can be written as,

$$
\begin{equation*}
\left(1 / v_{0}\right) j_{p}=\left|\psi_{\uparrow,-1 / 2}\right|^{2}-\left|\psi_{\downarrow, 1 / 2}\right|^{2} \tag{6.29}
\end{equation*}
$$

which should be constant over the entire ring. This in turn implies

$$
\begin{equation*}
\left(d / v_{0}\right) j_{p}=\int_{0}^{d} d y\left|\psi_{\uparrow,-1 / 2}\right|^{2}-\int_{0}^{d} d y\left|\psi_{\downarrow, 1 / 2}\right|^{2} \tag{6.30}
\end{equation*}
$$

Using this equation and the fact that wavefunction is normalized (assuming the $J_{z}= \pm 1$ components are zero), we can write Eq. (6.28) as

$$
\begin{equation*}
\rho_{I}=\frac{1}{2}\left[\mathbf{1}+\frac{d}{v_{0}} j_{p} \sigma_{z}\right] . \tag{6.31}
\end{equation*}
$$

The probability current vanishes in the energy gaps, which therefore implies that the entanglement entropy, $S=-\operatorname{Tr} \rho_{I} \log \rho_{I}$, reaches its maximum value of $\log 2$ at the band edges ${ }^{2}$. This finding is consistent with numerical results, as demonstrated in Fig. 6.10. One would expect that the spectral gaps occur at values of the magnetic flux where the electronic and impurity spins interact most strongly, and the fact that the entanglement is greatest near these values is consistent with this picture.

### 6.8 Origin of flat bands and parameter dependence for spin $1 / 2$ impurity

In this section, we focus on the case of a single spin $1 / 2$ impurity with a square potential, and we investigate quantitatively how the spectrum depends on the system parameters. As we discussed in Sections 6.4 and 6.5 , the energy spectrum is completely determined by the transmission amplitude $t$ (see Eq. (6.14)). As we show in Section 6.11, for a square potential this coefficient can be written as,

$$
\begin{equation*}
t(E)=\frac{e^{i E(d-w) / v_{0}} v_{0} q}{v_{0} q \cos (q w)-i\left(A^{z}+E\right) \sin (q w)} \tag{6.32}
\end{equation*}
$$

where $v_{0} q=\sqrt{\left(E+A^{z}\right)^{2}-\left(A^{\perp}\right)^{2}}$. The magnitude of the transmission amplitude is minimal at $E=-A^{z}$, at which it assumes the value $|t|=\operatorname{sech}\left(w A^{\perp} / v_{0}\right)$. Thus, the transmission is exponentially suppressed as the width $w$ or height $A^{\perp}$ of the impurity barrier are increased, or as the electron velocity $v_{0}$ is reduced. The dependence of $|t|$ on $A^{\perp}$ is demonstrated in Fig. 6.11, where it is evident that $|t|$ flattens out close to zero over a broad range of energies that grows as $A^{\perp}$ is increased. This behavior gives rise to the flat bands that occur

[^3]

Figure 6.11: Magnitude of the transmission coefficient as a function of energy for a single spin $1 / 2$ impurity with square potential for several different values of $A^{\perp}$. From top to bottom: $A^{\perp}=0.025,0.05,0.075,0.1 \mathrm{eV}$. The remaining parameters are $d=1000 \AA, w=130$ $\AA, A^{z}=0.05 \mathrm{eV}[9], v_{0}=2.4 \mathrm{eV} \AA$.
in the middle of Fig. 6.7(a). To see this, recall that the condition for a band to occur is $\left|\cos \phi_{t}\right|<|t|$, so that for $|t| \ll 1$, the range of phases satisfying this condition becomes very narrow. For physical parameters, $\phi_{t}$ is dominated by the kinematic term $E(d-w) / v_{0}$, which is why the parametric plot trajectories shown in Figs. 6.7(b) and 6.8(b) are nearly circular. Here, we can use this observation to estimate the smallest bandwidth $\sigma_{E}$ and the largest band gap $\Delta E$, which occur near $E=-A^{z}: \sigma_{E} \approx 2 v_{0} /(d-w) \arcsin \left(\operatorname{sech}\left(w A^{\perp} / v_{0}\right)\right)$, $\Delta E \approx \pi v_{0} /(d-w)$. Notice that $\sigma_{E}$ depends sensitively on the impurity coupling while $\Delta E$ does not. For the typical experimental parameters used in Fig. 6.7, these quantities evaluate to $\sigma_{E} \approx 0.7 \mathrm{meV}$ and $\Delta E \approx 8 \mathrm{meV}$, corresponding to the flat bands in the vicinity of $E=-0.05 \mathrm{eV}$ in Fig. 6.7(a).

To further elucidate the dependence on system parameters, we show the energy spectrum for several sets of parameters in Fig. 6.12. In Fig. 6.12(a), we increase the strength of the impurity coupling by an order of magnitude relative to Fig. 6.7, where the most striking consequence is that the bands for $E<0$ become significantly flatter ( $\sigma_{E} \sim 10^{-14} \mathrm{eV}$ near


Figure 6.12: Dependence of energy spectrum on ring geometry and impurity couplings for a spin $1 / 2$ impurity with square potential. The parameters are (a) $d=1000 \AA, w=130 \AA$, $A^{z}=A^{\perp}=0.05 \mathrm{eV}(\mathrm{b}) d=1000 \AA, w=130 \AA, A^{z}=0.03, A^{\perp}=0.05 \mathrm{eV}$ (c) $d=2000 \AA$, $w=130 \AA, A^{z}=A^{\perp}=0.05 \mathrm{eV}$ and (d) $d=1000 \AA, w=200 \AA, A^{z}=A^{\perp}=0.05 \mathrm{eV}$. In all cases, $v_{0}=2.4 \mathrm{eV} \AA$. For the larger ring circumference case shown in (c), the magnetic field range spans several Brillouin zones.
$E=-A^{z}=-0.5 \mathrm{eV}$ in this case). Notice that the band gaps remain approximately the same, as is consistent with our finding that these are insensitive to the impurity coupling. In addition, the flat bands continue over a 1 eV range in this case, all the way down to $E \approx-1$ eV , since now $A^{\perp}=0.5 \mathrm{eV}$.

In Fig. 6.12(b), we consider a situation in which $A^{z} \neq A^{\perp}$. In particular, we keep $A^{\perp}=0.05$ eV as in Fig. 6.7, but now reduce the longitudinal coupling to $A^{z}=0.03 \mathrm{eV}$. This shifts the flat band region upward in energy but does not affect $\sigma_{E}$ or $\Delta E$. An additional consequence of $A^{z} \neq A^{\perp}$ is that the behavior near $E=0$ is modified. When $A^{z}=A^{\perp}$ and $E=0$, it follows from Eq. (6.32) that $t=v_{0} /\left(v_{0}-i A^{\perp} w\right)$, which saturates the band condition $\left|\cos \left(\phi_{t}\right)\right|=|t|$. Thus, $E=0$ always corresponds to a band edge in the case of an isotropic interaction, while this property is lost in the anisotropic case. This behavior is evident from a comparison of Fig. 6.12(b) with the other panels of that figure.

Figs. 6.12(c),(d) show the dependence of the spectrum on the geometry of the ring. Increasing the ring circumference changes the number of "Brillouin zones" that fit within a given range of magnetic field, as shown in Fig. 6.12(c). Here, we increase the circumference by a factor of 2 relative to Fig. 6.7, so that now two full Brillouin zones fit instead of only half of one. In addition, the density of states increases by a factor of 2 , as follows from the inverse dependence of $\sigma_{E}$ and $\Delta E$ on the circumference.

The spectra shown in Fig. 6.12 contain a number of very small gaps, raising the question of whether the gaps ever close completely to form a Dirac point. As was mentioned in Sec. 6.4, band touching points can arise if the band edge condition, $\left|\cos \phi_{t}\right|=|t|$, and the transparency condition, $|t|=1$, are simultaneously satisfied. In the case of a square impurity barrier, we see from Eq. (6.32) that $|t|=1$ when $q=n \pi / w$ for arbitrary nonzero integer $n$,
which corresponds to the following energies

$$
\begin{equation*}
E_{n}^{ \pm}=-A^{z} \pm \sqrt{\left(A^{\perp}\right)^{2}+n^{2} \pi^{2} v_{0}^{2} / w^{2}} \tag{6.33}
\end{equation*}
$$

At these energies, the transmission amplitude reduces to a pure phase:

$$
\begin{equation*}
t\left(E_{n}^{ \pm}\right)= \pm e^{i E_{n}^{ \pm}(d-w) / v_{0}} \tag{6.34}
\end{equation*}
$$

Notice that these energies are always guaranteed to lie within a band since $\left|\cos \phi_{t}\right| \leq|t|$ is automatically satisfied. Imposing the band edge condition, $\phi_{t}=m \pi$ for integer $m$, then leads to the following set of discrete values of the ring circumference for which Dirac points appear in the spectrum:

$$
\begin{equation*}
d=w+m \pi v_{0} / E_{n}^{ \pm} \tag{6.35}
\end{equation*}
$$

Any choice of $m$ will yield a Dirac point at energy $E_{n}^{ \pm}$. Although the particular form of Eq. (6.35) only holds in the idealized case of a square impurity potential, an analogous expression should arise for other potential shapes.

### 6.9 Conclusion

In conclusion, we analyzed the problem of a topological insulator ring in which the helical edge states are coupled to magnetic impurities or spinful nuclei of arbitrary spin. This interaction breaks time-reversal symmetry and enables the backscattering of electrons. We considered the case where the ring is threaded by a magnetic flux, and we showed that the energy spectrum as a function of this flux is given by a universal formula that depends only on the amplitude of transmission through the impurity. We found that the impurity can give rise to sizable spectral gaps and flat bands, and we calculated the gap sizes and
bandwidths for a variety of experimentally relevant parameter regimes. We further showed that the entanglement between the electronic and impurity spins is maximal near these gaps, while at energies far away from these gaps, little entanglement develops, and the helical edge states remain unaffected by the impurity. Our results can be tested with quantum interference measurements in nanorings, providing a new approach to understanding the role of magnetic impurities in topological insulator transport.

### 6.10 Generalized time-reversal relations for spin $1 / 2$ impurities

GTR symmetry mixes eigenstates incoming from the left of the impurity with those incoming from the right. In this section, we exploit this fact to obtain a simple expression for the eigenstate spectrum in terms of the scattering transmission amplitude. This result is universal in the sense that it does not depend on the spatial profile (barrier shape) of the impurity or any other details of the system. We begin by supposing that the initial impurity state is an eigenstate of $I_{z}$, and we write the wavefunctions in terms of the transmission and reflection amplitudes. For example, we denote the left-incoming scattering eigenstate with initial impurity state $|-1 / 2\rangle$ by $\left|\phi_{\rightarrow,-1 / 2}^{(p)}\right\rangle$ (total $J_{z}=0$ and momentum $p$ ). This state corresponds to the $A$ wave in Fig. 6.6. The state on each side of the impurity takes the form ( $x$ denoting left side of the barrier and $x^{\prime}$ the right side)

$$
\left\langle x \mid \phi_{\rightarrow,-1 / 2}^{(p)}\right\rangle=\left[\begin{array}{c}
0  \tag{6.36}\\
e^{i p x} \\
r_{\rightarrow} e^{-i p x} \\
0
\end{array}\right], \quad\left\langle x^{\prime} \mid \phi_{\rightarrow,-1 / 2}^{(p)}\right\rangle=\left[\begin{array}{c}
0 \\
t_{\rightarrow} e^{i p x^{\prime}} \\
0 \\
0
\end{array}\right] .
$$

Similarly, for the $B$ wave (incident from the right with the impurity initially in $|1 / 2\rangle$ ) we have,

$$
\left\langle x \mid \phi_{\leftarrow, 1 / 2}^{(p)}\right\rangle=\left[\begin{array}{c}
0  \tag{6.37}\\
0 \\
t_{\leftarrow} e^{-i p x} \\
0
\end{array}\right], \quad\left\langle x^{\prime} \mid \phi_{\leftarrow, 1 / 2}^{(p)}\right\rangle=\left[\begin{array}{c}
0 \\
r_{\leftarrow} e^{i p x^{\prime}} \\
e^{-i p x^{\prime}} \\
0
\end{array}\right] .
$$

We also have an eigenstate corresponding to a left-incoming electron with the impurity initially in state $|1 / 2\rangle$ :

$$
\left\langle x \mid \phi_{\rightarrow, 1 / 2}^{(p)}\right\rangle=\left[\begin{array}{c}
e^{i p x}  \tag{6.38}\\
0 \\
0 \\
0
\end{array}\right], \quad\left\langle x^{\prime} \mid \phi_{\rightarrow, 1 / 2}^{(p)}\right\rangle=\left[\begin{array}{c}
\mathcal{P}_{\rightarrow} e^{i p x^{\prime}} \\
0 \\
0 \\
0
\end{array}\right]
$$

and similarly for $\left|\phi_{\leftarrow,-1 / 2}^{(p)}\right\rangle$.
We define the TR operator as $\mathcal{T}_{1 / 2}=i \sigma_{y} \Theta$ where $\Theta$ is the complex conjugation operator. The corresponding operator for the GTR symmetry in the case of a spin $1 / 2$ impurity is then $\mathcal{T}_{G T R}=\mathcal{T}_{1 / 2} \otimes \mathcal{T}_{1 / 2}$. It is easy to see that $\mathcal{T}_{G T R}\left|\phi_{\rightarrow, 1 / 2}^{(p)}\right\rangle$ must be proportional to $\left|\phi_{\leftarrow,-1 / 2}^{(p)}\right\rangle$, from which we conclude that for "passing" states like Eq. (6.38),

$$
\begin{equation*}
\mathcal{P}_{\rightarrow}=\mathcal{P}_{\leftarrow} \equiv \mathcal{P} \quad \text { with } \quad|\mathcal{P}|^{2}=1 \tag{6.39}
\end{equation*}
$$

Applying $\mathcal{T}_{G T R}$ on the two states with $J_{z}=0$, Eqs. (6.36) and (6.37), we see that the resulting state is a superposition of left-incoming and right-incoming states:

$$
\begin{equation*}
\mathcal{T}_{G T R}\left|\phi_{\rightarrow,-1 / 2}^{(p)}\right\rangle=a_{0}\left|\phi_{\rightarrow,-1 / 2}^{(p)}\right\rangle+b_{0}\left|\phi_{\leftarrow, 1 / 2}^{(p)}\right\rangle . \tag{6.40}
\end{equation*}
$$

For instance, $\mathcal{T}_{G T R}\left|\phi_{\rightarrow,-1 / 2}^{(p)}\right\rangle$ corresponds to the following left-side and right-side wavefunctions:

$$
\begin{align*}
\langle x| \mathcal{T}_{G T R}\left|\phi_{\rightarrow,-1 / 2}^{(p)}\right\rangle & =\left[\begin{array}{c}
0 \\
-\bar{r}_{\rightarrow} e^{i p x} \\
-e^{-i p x} \\
0
\end{array}\right], \\
\left\langle x^{\prime}\right| \mathcal{T}_{G T R}\left|\phi_{\rightarrow,-1 / 2}^{(p)}\right\rangle & =\left[\begin{array}{c}
0 \\
0 \\
-\bar{t}_{\rightarrow} e^{-i p x^{\prime}} \\
0
\end{array}\right] . \tag{6.41}
\end{align*}
$$

Imposing Eq. (6.40) to the wavefunction on the left side of the barrier will give us one equation per component. One of these equations implies that $a_{0}=-\bar{r}_{\rightarrow}$. We do the same on the right side, and the equation resulting from the third component implies $b_{0}=-\bar{t}_{\rightarrow}$. The remaining components (applied on both left and right) provide the following equations:

$$
\begin{align*}
& \bar{r}_{\rightarrow} t_{\rightarrow}+r_{\leftarrow} \bar{t}_{\rightarrow}=0,  \tag{6.42}\\
& \left|r_{\rightarrow}\right|^{2}+t_{\leftarrow} \bar{t}_{\rightarrow}=1 . \tag{6.43}
\end{align*}
$$

Combining Eq. (6.43) with the current conservation condition, $\left|r_{\rightarrow}\right|^{2}+\left|t_{\rightarrow}\right|^{2}=1$, we find that the two transmission amplitudes must be equal:

$$
\begin{equation*}
t_{\rightarrow}=t_{\leftarrow} \equiv t \equiv|t| e^{i \phi_{t}} . \tag{6.44}
\end{equation*}
$$

Multiplying Eq. (6.42) by $r_{\rightarrow} t$ and simplifying the result with the help of (6.43) and (6.44),
we also find

$$
\begin{equation*}
r_{\leftarrow} r_{\rightarrow}=\left(|t|^{2}-1\right) e^{2 i \phi_{t}} . \tag{6.45}
\end{equation*}
$$

Eqs. (6.44) and (6.45) are used to derive the remarkably simple expression for the band structure given in Eq. (6.14).

So far the only assumption we have made is that the Hamiltonian has GTR symmetry. If the impurity potential also possesses inversion symmetry (as a result of which $r_{\rightarrow}=r_{\leftarrow} \equiv$ $\left.r \equiv|r| e^{i \phi_{r}}{ }^{3}\right)$, then Eq. (6.42) implies $\bar{r} t=-r \bar{t}$, which in turn leads to $\phi_{t}=\phi_{r}+\pi / 2$. The presence of this symmetry of course has no impact on Eq. (6.14), but it does simplify the calculation of the wavefunction, for example in the case of a square barrier considered in Section 6.11.

### 6.11 Transmission and reflection amplitudes for square impurity barrier

In this section, we outline the general approach for finding reflection and transmission amplitudes for an arbitrary-spin impurity with square potential (i.e., $F(y)=\Theta(w / 2-|y|)$ in Eq. (6.6)). We do this by decomposing the Hilbert space into sectors of total spin $J_{z}$ and by separately solving for the scattering amplitudes in each sector. To better understand the structure of the Hamiltonian in each sector, we first consider the case of a spin $1 / 2$ impurity. Using a wavefunction ansatz that includes a chiral plane wave factor of the form $e^{i q y}$, the

[^4]full Hamiltonian $H=H_{0}+H_{S, I}$ inside the interaction region is
\[

H=\left[$$
\begin{array}{cccc}
A^{z}+q v_{0} & 0 & 0 & 0  \tag{6.46}\\
0 & q v_{0}-A^{z} & A^{\perp} & 0 \\
0 & A^{\perp} & -q v_{0}-A^{z} & 0 \\
0 & 0 & 0 & A^{z}-q v_{0}
\end{array}
$$\right]
\]

The middle block of this matrix corresponds to the $J_{z}=0$ sector $\left(M_{\left(J_{z}=0\right)}\right)$ and the other two states are $|\uparrow \uparrow\rangle$ and $|\downarrow \downarrow\rangle$, which do not couple to any other states and are irrelevant for calculating scattering amplitudes. For a general impurity spin, $H$ will be block diagonal (when the basis states are grouped according to $J_{z}$ ), all of which are two-dimensional except for the one-dimensional blocks corresponding to maximal $\left|J_{z}\right|$. Each two-dimensional block, which we denote by $M_{\left(J_{z}\right)}$, will have an associated set of scattering amplitudes $r_{\leftarrow}, r_{\rightarrow}, t_{\leftarrow}$, $t_{\rightarrow}$. The most general form of $M_{\left(J_{z}\right)}$ for arbitrary $J_{z}$ is

$$
\begin{align*}
M_{\left(J_{z}\right)} & =\left[\begin{array}{cc}
q v_{0}-u-m_{0} & h \\
h & -q v_{0}-u+m_{0}
\end{array}\right] \\
& =-u \mathbf{1}+\left(q v_{0}-m_{0}\right) \sigma_{z}+h \sigma_{x} \\
& =-u \mathbf{1}+\left[b \cos \theta \sigma_{z}+b \sin \theta \sigma_{x}\right] \tag{6.47}
\end{align*}
$$

where we have defined

$$
\left[\begin{array}{c}
\cos \theta  \tag{6.48}\\
\sin \theta
\end{array}\right]=\frac{1}{b}\left[\begin{array}{c}
q v_{0}-m_{0} \\
h
\end{array}\right] \quad \text { and } \quad b^{2}=h^{2}+\left(q v_{0}-m_{0}\right)^{2}
$$

The eigenvectors are

$$
\left[\begin{array}{c}
\cos \theta / 2  \tag{6.49}\\
\sin \theta / 2
\end{array}\right] \quad \text { and } \quad\left[\begin{array}{c}
-\sin \theta / 2 \\
\cos \theta / 2
\end{array}\right]
$$

and the eigenvalues are $E=-u \pm b$. We can solve for $q$ in terms of energy,

$$
\begin{equation*}
q_{ \pm}=\left(1 / v_{0}\right)\left(m_{0} \pm \sqrt{(E+u)^{2}-h^{2}}\right) \tag{6.50}
\end{equation*}
$$

Note that $q$ and hence $\theta$ may be complex depending on the energy.

To obtain the scattering amplitudes, we need to match wavefunction ansatzes inside and outside the impurity potential at the boundaries of the potential. Defining $a=d-w$, we match the wavefunctions at $y=a / 2$ and $y=a / 2+w$ (see Fig. 6.6), where

$$
\psi(0)=\left[\begin{array}{c}
A  \tag{6.51}\\
r_{\rightarrow} A+t_{\leftarrow} B
\end{array}\right], \quad \psi(d)=\left[\begin{array}{c}
t_{\rightarrow} A+r_{\leftarrow} B \\
B
\end{array}\right] .
$$

On the left side of the interaction region, this amounts to requiring

$$
\left[\begin{array}{c}
A e^{i p a / 2}  \tag{6.52}\\
\left(r_{\rightarrow} A+t_{\leftarrow} B\right) e^{-i p a / 2}
\end{array}\right]=\left[\begin{array}{c}
\cos \theta / 2 \\
\sin \theta / 2
\end{array}\right] c_{+}+\left[\begin{array}{c}
\sin \theta / 2 \\
\cos \theta / 2
\end{array}\right] c_{-},
$$

and on the right side,

$$
\left[\begin{array}{c}
\left(t_{\rightarrow} A+r_{\leftarrow} B\right) e^{-i p a / 2}  \tag{6.53}\\
B e^{i p a / 2}
\end{array}\right]=\left[\begin{array}{c}
\cos \theta / 2 \\
\sin \theta / 2
\end{array}\right] e^{i q_{+} w} c_{+}+\left[\begin{array}{c}
\sin \theta / 2 \\
\cos \theta / 2
\end{array}\right] e^{i q_{-} w} c_{-},
$$

where $p=E / v_{0}$. These equations, combined with the single-valuedness condition (6.8), allow us to eliminate $A, B$, and $c_{ \pm}$and to obtain the reflection and transmission amplitudes.

As an example, for the case of spin $1 / 2$ impurity where $m_{0}=0$ and $q_{ \pm}= \pm q$, we find

$$
\begin{align*}
\sqrt{r_{\leftarrow} r_{\rightarrow}} & =i e^{i p a} \frac{\sin \theta \sin q w}{e^{-i q w} \cos ^{2} \theta / 2-e^{i q w} \sin ^{2} \theta / 2}  \tag{6.54}\\
t_{\leftarrow}=t_{\rightarrow} & =e^{i p a} \frac{\cos \theta}{e^{-i q w} \cos ^{2} \theta / 2-e^{i q w} \sin ^{2} \theta / 2} . \tag{6.55}
\end{align*}
$$

With some simple algebraic manipulations, we can transform this transmission amplitude to the form of Eq. (6.32). In addition, we may write

$$
\begin{align*}
|r|^{2} & =\frac{\sin ^{2}(q w) \sin ^{2} \theta}{1-\cos ^{2}(q w) \sin ^{2} \theta}=\frac{\sin ^{2}(q w)}{1+\cot ^{2} \theta-\cos ^{2}(q w)} \\
& =\left[1+\frac{\cot ^{2} \theta}{\sin ^{2}\left(\left(h / v_{0}\right) w \cot \theta\right)}\right]^{-1} \tag{6.56}
\end{align*}
$$

where we have used

$$
\begin{equation*}
\sin \theta=\frac{h}{E+u}=\frac{A^{\perp}}{E+A^{z}}, \tag{6.57}
\end{equation*}
$$

where the first equality holds for any sector, while the second equality applies for $J_{z}=0$. Note that we can express all other variables in terms of this shifted (and dimensionless) energy $(E+u) / h$. For example, we may rewrite the transmission amplitude as

$$
\begin{equation*}
t=\frac{e^{i E(d-w) / v_{0}} v_{0} q}{v_{0} q \cos (q w)-i\left(A^{z}+E\right) \sin (q w)} . \tag{6.58}
\end{equation*}
$$

### 6.12 Generalized time-reversal relations for spin $1 \mathrm{im}-$ purities

In this section, we derive the consequences of GTR symmetry in the case of a spin 1 impurity. As explained in Section 6.3, the wavefunction decomposes into blocks spanned by basis states
with the same absolute value of total spin $\left|J_{z}\right|$. As discussed in Sec. 6.5, the spectrum for an arbitrary-spin impurity can be obtained by combining the solutions for spin $1 / 2$ (obtained in Sec. 6.4) and spin 1 impurities. In the case of a spin 1 impurity, the electron-impurity Hilbert space divides into two trivial one-dimensional subspaces corresponding to the states with $J_{z}= \pm 3 / 2$, and a four-dimensional block spanned by states with $J_{z}= \pm 1 / 2$. Following the procedure of Section 6.10, we consider left-incoming and right-incoming states for which the impurity is initially in an eigenstate of $I_{z}$. We label these states as e.g., $\left|\phi_{\rightarrow, 0}^{(p)}\right\rangle$, which represents an electron incoming from the left with momentum $p$ and with the impurity initially in state $|0\rangle$. We write the wavefunctions for each of these states on the left-side ( $x$ ) and right-side $\left(x^{\prime}\right)$ of the impurity in terms of reflection and transmission amplitudes:

$$
\begin{align*}
& \left\langle x \mid \phi_{\rightarrow, 0}^{(p)}\right\rangle=\left[\begin{array}{c}
e^{i p x} \\
0 \\
r_{\rightarrow}^{+} e^{-i p x} \\
0
\end{array}\right], \quad\left\langle x^{\prime} \mid \phi_{\rightarrow, 0}^{(p)}\right\rangle=\left[\begin{array}{c}
t_{\rightarrow}^{+} e^{i p x^{\prime}} \\
0 \\
0 \\
0
\end{array}\right],  \tag{6.59}\\
& \left\langle x \mid \phi_{\rightarrow,-1}^{(p)}\right\rangle=\left[\begin{array}{c}
0 \\
e^{i p x} \\
0 \\
r_{\rightarrow}^{-} e^{-i p x}
\end{array}\right], \quad\left\langle x^{\prime} \mid \phi_{\rightarrow,-1}^{(p)}\right\rangle=\left[\begin{array}{c}
0 \\
t_{\rightarrow}^{-} e^{i p x^{\prime}} \\
0 \\
0 \\
0 \\
0 \\
\left\langle x \mid \phi_{\leftarrow, 1}^{(p)}\right\rangle= \\
t_{\leftarrow}^{+} e^{-i p x} \\
0
\end{array}\right],  \tag{6.60}\\
& \left\langle x^{\prime} \mid \phi_{\leftarrow, 1}^{(p)}\right\rangle=\left[\begin{array}{c}
{\left[\begin{array}{c}
r_{\leftarrow}^{+} e^{i p x^{\prime}} \\
0 \\
e^{-i p x^{\prime}} \\
0
\end{array}\right],}
\end{array},\right. \tag{6.61}
\end{align*}
$$

$$
\left\langle x \mid \phi_{\leftarrow, 0}^{(p)}\right\rangle=\left[\begin{array}{c}
0  \tag{6.62}\\
0 \\
0 \\
t_{\leftarrow}^{-} e^{-i p x}
\end{array}\right], \quad\left\langle x^{\prime} \mid \phi_{\leftarrow, 0}^{(p)}\right\rangle=\left[\begin{array}{c}
0 \\
r_{\leftarrow}^{-} e^{i p x^{\prime}} \\
0 \\
e^{-i p x^{\prime}}
\end{array}\right] .
$$

Here, the basis states are $|\uparrow, 0\rangle,|\uparrow,-1\rangle,|\downarrow, 0\rangle,|\downarrow, 1\rangle$; we have left out the two "passing" states $|\uparrow, 1\rangle$ and $|\downarrow,-1\rangle$ since the action of GTR on these will be identical to that for the passing states in the spin $1 / 2$ case treated in Section 6.10, namely the impurity-induced phases on these states obey the relation $\mathcal{P}_{\rightarrow}=\mathcal{P}_{\leftarrow}=\mathcal{P}$.

In order to understand the action of GTR on these states, we must first generalize the definition of the GTR operator introduced in Section 6.10 to the case of a spin 1 impurity. We choose the following definition:

$$
\mathcal{T}_{1}=\left(\begin{array}{ccc}
0 & 0 & 1  \tag{6.63}\\
0 & -1 & 0 \\
1 & 0 & 0
\end{array}\right) \Theta
$$

where $\Theta$ is again the complex conjugation operator. The GTR operator is then $\mathcal{T}_{\text {GTR }}=$ $\mathcal{T}_{1 / 2} \otimes \mathcal{T}_{1}$. Acting with this operator on one of the states in Eqs. (6.59) - (6.62) yields a linear combination of two of the other states. For example,

$$
\begin{equation*}
\mathcal{T}_{G T R}\left|\phi_{\rightarrow, 0}^{(p)}\right\rangle=a^{-}\left|\phi_{\rightarrow,-1}^{(p)}\right\rangle+b^{-}\left|\phi_{\leftarrow, 0}^{(p)}\right\rangle, \tag{6.64}
\end{equation*}
$$

and

$$
\begin{equation*}
\mathcal{T}_{G T R}\left|\phi_{\rightarrow,-1}^{(p)}\right\rangle=a^{+}\left|\phi_{\rightarrow, 0}^{(p)}\right\rangle+b^{+}\left|\phi_{\leftarrow, 1}^{(p)}\right\rangle . \tag{6.65}
\end{equation*}
$$

By acting on the other two states in a similar fashion, we get a total of four equations like Eqs. (6.64) and (6.65), each of which yields two 4 -component spinor equations when we
restrict to the left- or right-side of the impurity. This gives a total of 32 complex equations, 16 of which are trivial, and 8 more can be used to solve for the 8 coefficients $a^{-}, b^{-}$, etc. The remaining 8 complex equations constrain the scattering amplitudes and can be written as

> (i) $1=\bar{t}_{\rightarrow}^{+} t_{\leftarrow}^{-}+\bar{r}_{\rightarrow}^{+} r_{\rightarrow}^{-}$
> (ii) $\bar{t}_{\rightarrow}^{+} r_{\leftarrow}^{-}=-\bar{r}_{\rightarrow}^{+} t_{\rightarrow}^{-}$
> (iii) $1=\bar{t}_{\rightarrow}^{-} t_{\leftarrow}^{+}+\bar{r}_{\rightarrow}^{-} r_{\rightarrow}^{+}$
> (iv) $\bar{r}_{\rightarrow}^{-} t_{\rightarrow}^{+}=-\bar{t}_{\rightarrow}^{-} r_{\leftarrow}^{+}$
> (v) $\bar{t}_{\leftarrow}^{+} r_{\rightarrow}^{-}=-\bar{r}_{\leftarrow}^{+} t_{\leftarrow}^{-}$
> (vi) $1=\bar{t}_{\leftarrow}^{+} t_{\rightarrow}^{-}+\bar{r}_{\leftarrow}^{+} r_{\leftarrow}^{-}$
> (vii) $\bar{t}_{\leftarrow}^{-} r_{\rightarrow}^{+}=-\bar{r}_{\leftarrow}^{-} t_{\leftarrow}^{+}$
> (viii) $1=\bar{t}_{\leftarrow}^{-} t_{\rightarrow}^{+}+\bar{r}_{\leftarrow}^{-} r_{\leftarrow}^{+}$

Equations (i) and (ii) come from solving Eq. (6.64) on the left- and right-side of the impurity, equations (iii) and (iv) come from solving Eq. (6.65), and equations (v), (vi) and (vii), (viii) come from solving similar equations involving $\mathcal{T}_{G T R}\left|\phi_{\leftarrow 1}\right\rangle$ and $\mathcal{T}_{G T R}\left|\phi_{\leftarrow 0}\right\rangle$, respectively. An instant consequence of these eight equations is,

$$
\begin{align*}
& \left|t_{\rightarrow}^{-}\right|=\left|t_{\rightarrow}^{+}\right|=\left|t_{\leftarrow}^{+}\right|=\left|t_{\leftarrow}^{-}\right|=|t|,  \tag{6.66}\\
& \left|r_{\rightarrow}^{-}\right|=\left|r_{\rightarrow}^{+}\right|=\left|r_{\leftarrow}^{+}\right|=\left|r_{\leftarrow}^{-}\right|=|r| . \tag{6.67}
\end{align*}
$$

Applying this constraint to equations (ii), (iv), (vi) and (viii) gives a relation between the
phases. Adding the resulting equations from (ii) and (viii) gives

$$
\begin{equation*}
\phi_{t \_}-\phi_{t_{\leftrightarrows}^{+}}=\phi_{t \rightrightarrows}-\phi_{t \leftharpoondown}, \tag{6.68}
\end{equation*}
$$

and adding the resulting equations from (vi) and (viii) gives

$$
\begin{equation*}
\phi_{r_{\rightrightarrows}^{+}}-\phi_{r_{亡}^{ \pm}}=\phi_{r_{\rightrightarrows}^{-}}-\phi_{r_{\rightleftarrows}^{-}} . \tag{6.69}
\end{equation*}
$$

Furthermore, solving the all eight equations with the constraint that $|t|^{2}+|r|^{2}=1$, provides us with

$$
\begin{align*}
& \phi_{t \rightrightarrows}=\phi_{t \rightleftarrows}=\phi_{r \rightrightarrows}+\phi_{r \rightleftarrows}-\phi_{t \leftharpoondown}+\pi \\
& \phi_{t \leftharpoondown}=\phi_{t_{\rightarrow}^{+}} \\
& \phi_{r_{\rightarrow}^{+}}=\phi_{r_{\rightarrow}^{-}} \\
& \phi_{r_{\leftarrow}}=\phi_{r_{\leftarrow}^{ \pm}} \tag{6.70}
\end{align*}
$$

from which Eqs. (6.17) and (6.18) follow.

Now we proceed to derive Eq. (6.20) from the equation that results from imposing the single-valuedness condition, Eq. (6.8), in each sector:

$$
\begin{equation*}
e^{2 \pi i \Phi_{B} / \Phi_{0}}=\frac{1-r_{\leftarrow}^{ \pm} r_{\rightarrow}^{ \pm}+t_{\leftarrow}^{ \pm} t_{\longrightarrow}^{ \pm} \pm \sqrt{\left(1-r_{\longleftarrow}^{ \pm} r_{\rightarrow}^{ \pm}+t_{\leftarrow}^{ \pm} t_{\rightarrow}^{ \pm}\right)^{2}-4 t_{\leftarrow}^{ \pm} t_{\longrightarrow}^{ \pm}}}{2 t_{\leftarrow}^{ \pm}} . \tag{6.71}
\end{equation*}
$$

Here, the $\pm$ in front of the square root is independent from the sector labels $\pm$ labeling the scattering amplitudes. Introducing the average amplitude phases as in Sec. 6.5, $\phi_{x}^{\circ}=$ $\left(\phi_{x \rightarrow}+\phi_{x \leftarrow}\right) / 2$ where $x=r^{ \pm}, t^{ \pm}$, we can rewrite the above expression using Eqs. (6.66),
(6.67), (6.70):

$$
\begin{equation*}
e^{2 \pi i \frac{\Phi_{B}}{\Phi_{0}}}=e^{i\left(\phi_{t \rightarrow-}-\phi_{t_{\leftarrow}^{ \pm}}\right) / 2}\left[\cos \phi_{t}^{\circ} /|t| \pm \sqrt{\left(\cos \phi_{t}^{\circ} /|t|\right)^{2}-1}\right], \tag{6.72}
\end{equation*}
$$

where $\phi_{t^{+}}^{\circ}=\phi_{t^{-}}^{\circ} \equiv \phi_{t}^{\circ}$. From Eq. (6.68) we see that the overall phase factor differs only by a sign between the two sectors, $\phi_{t_{\rightarrow}^{+}}-\phi_{t_{\longleftarrow}^{\ddagger}}=-\left(\phi_{t_{\hookrightarrow}^{-}}-\phi_{t \leftarrow}\right)$. Combined with the two possible branches of the square root in Eq. (6.72), this therefore produces four distinct values of $\Phi_{B}$ for each value of the energy.

Next, we show how to derive the ratio of spin and probability currents given in Eq. (6.25). First, we apply the single-valuedness condition, Eq. (6.8), to Eq. (6.18) to obtain a formula for the wavefunction coefficients:

$$
\begin{equation*}
\frac{B^{ \pm}}{A^{ \pm}}=\frac{e^{2 \pi i\left(\Phi_{B} / \Phi_{0}\right)^{ \pm}} r_{\rightarrow}^{ \pm}}{1-e^{2 \pi i\left(\Phi_{B} / \Phi_{0}\right)^{ \pm}} t_{\leftarrow}^{ \pm}} \tag{6.73}
\end{equation*}
$$

Here, we have included the superscript $\pm$ on $\left(\Phi_{B} / \Phi_{0}\right)$ as a reminder that we must use the appropriate version of Eq. (6.72) corresponding to each sector. The spin current evaluates to

$$
\begin{equation*}
\left(2 / v_{0}\right) j_{s}=\psi(y)^{\dagger} \psi(y)=\left|A^{+}\right|^{2}+\left|r_{\rightarrow}^{+} A^{+}+t_{\leftarrow}^{+} B^{+}\right|^{2}+\left|A^{-}\right|^{2}+\left|r_{\rightarrow}^{-} A^{-}+t_{\leftarrow}^{-} B^{-}\right|^{2} \tag{6.74}
\end{equation*}
$$

Using Eq. (6.73) to eliminate $B$ 's, this expression becomes

$$
\begin{equation*}
\left(2 / v_{0}\right) j_{s}=\left|A^{+}\right|^{2}\left(1+\left|\frac{r_{\rightarrow}^{+}}{1-e^{2 \pi i\left(\Phi_{B} / \Phi_{0}\right)^{+}} t_{\leftarrow}^{+}}\right|^{2}\right)+\left|A^{-}\right|^{2}\left(1+\left|\frac{r_{\rightarrow}^{-}}{1-e^{2 \pi i\left(\Phi_{B} / \Phi_{0}\right)^{-}} t_{\leftarrow}^{-}}\right|^{2}\right) . \tag{6.75}
\end{equation*}
$$

From the constraint that we found on the magnitude of reflection coefficients we see that $\left|r_{\rightarrow}^{+}\right|^{2}=\left|r_{\rightarrow}^{-}\right|^{2}$. Furthermore, from Eq. (6.72) we observe that $e^{2 \pi i\left(\Phi_{B} / \Phi_{0}\right)^{+}} t_{\leftarrow}^{+}=e^{2 \pi i\left(\Phi_{B} / \Phi_{0}\right)^{-}} t_{\leftarrow}^{-}$ (again, since $\phi_{t^{+}}^{\circ}=\phi_{t^{-}}^{\circ}$ and that $|t|$ 's are the same). As a result of this, the two quantities
in parentheses in Eq. (6.76) are equal and can be factored out:

$$
\begin{equation*}
\left(2 / v_{0}\right) j_{s}=\left(\left|A^{+}\right|^{2}+\left|A^{-}\right|^{2}\right)\left(1+\left|\frac{r_{\rightarrow}^{+}}{1-e^{2 \pi 2 \pi i\left(\Phi_{B} / \Phi_{0}\right)^{+}} t_{\leftarrow}^{+}}\right|^{2}\right) . \tag{6.76}
\end{equation*}
$$

We can write a very similar expression for $j_{p}$ which differs from this expression only in a minus sign and also contains the same factor $\left|A^{+}\right|^{2}+\left|A^{-}\right|^{2}$. Therefore in writing $j_{s} / j_{p}$ those terms cancel out and we arrive at Eq. (6.25).

## Chapter 7

## Conclusion and outlook

The physical realization of a quantum computer with well-characterized qubits is a task that requires considerable effort; overcoming decoherence and achieving high fidelity control of the qubit are some of the main obstacles ahead of us. This thesis presents techniques and approaches to obtain a better understanding and battling these problems in self-assembled QDs and TIs. In the following I will summarize the work presented and discuss their potential future directions.

In Chapter 3 we looked at optical control methods of of QDs and their potential as sources of single photon emission. In Chapter 4, we developed a novel version of DRAG-assisted control for battling unwanted off-resonant couplings in $\Lambda$-systems with unwanted levels. We also showed how achieving such high-fidelity control allows for a protocol for generating of photonic cluster states from a single QDM. In this thesis we focused on a single QDM, however, as it was presented in Section 2.3, QDMs allow us for QD-based scalable structures. A potential future direction to pursue, is developing similar DRAG methods, for a multiqubit system of QDMs. Furthermore, it is shown in Ref. [96], that by having a photon emitter and an ancilla qubit, the photonic repeater graph states of Ref. [39], suitable for quantum communication, can be produced. This requires designing of a $C Z$ gate between the ancilla and the qubit. A schematic of two QDMs in a cavity is shown in Fig. 7.1. Developing a DRAG method for this system not only facilitates a two-qubit gate for a scalable QDM system, but also enables generation of photonic repeater graph states as well.


Figure 7.1: Two QDMs in cavity connected to the same cavity mode $\omega_{c}$. A future outlook is to design a two-qubit gate between the two qubits.

In Chapter 5 we studied the problem of HF interaction between confined electrons with their nuclear spin environment in a QD. HF interactions are the main source of decoherence in QDs and the mode-locking technique of Ref. [132] is one of the established methods for nuclei-induced frequency focusing of a QD ensemble. We studied the effects of higher nuclear spin, relevant to actual experiments in these systems and showed the effects of quadrupolar interactions on mode-locking experiments. In particular, we showed that mode-locking can be used as a tool to diagnose the effects of quadrupolar interactions in QDs. As we showed, HF interactions are the main driver behind mode-locking, while quadrupolar interactions are destructive to mode-locking. A potential future direction is to find the limit at which the HF flip-flop can be safely neglected, and moreover, can we go beyond the independent nuclear spin assumption to include inter-nuclear interactions? Furthermore, one could develop a similar formalism while including several species of nuclear spins.

In Chapter 6 we studied the effects of magnetic impurities in TIs. Most materials used for TIs include spinful nuclei that lower the conductivity of the edge states by breaking the TR symmetry. Our model of a TI ring with a magnetic impurity provides a novel tool for
studying the role of impurities in TIs. Specifically, using the magnetic flux threading the ring, the band structures of the system can be tuned to desirable schemes. The tunability of the system paves a potential path for several TI ring-based spintronics applications.

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[^0]:    ${ }^{1}$ The terms 'self-assembled QDs' and 'QDs' are used interchangeably throughout the thesis. Unless it is noted, QD will refer to a self-assembled quantum dot.

[^1]:    ${ }^{1}$ Notice that this happens regardless of presence or absence of the corrective drives.

[^2]:    ${ }^{1}$ This whole concept is similar to the BHZ model of HgTe quantum wells in which the two blocks of the Hamiltonian are related through time-reversal [8]. Furthermore, similar spin conservation arguments arise when we include spin-orbit interactions in this model [10, 261, 264], firstly obtained in Ref. [256].

[^3]:    ${ }^{2}$ At the band edges (from Eq. (6.15)), the current vanishes continuously as well.

[^4]:    ${ }^{3}$ Note that $t_{\rightarrow}=t_{\leftarrow}$ holds regardless.

