



UPPSALA
UNIVERSITET

BACHELOR'S THESIS IN PHYSICS

1FA599, 15 CREDITS

JUNE 5, 2023

Creating Artificial Quantum Chiral States

Time Evolving Open Spin Chains

AUTHOR:

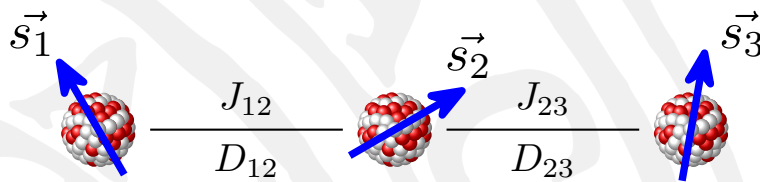
Emil Bror Carl Johansson Beiersdorf[†]

SUPERVISOR:

Erik Sjöqvist

SUBJECT READER:

Jonas Fransson



UPPSALA UNIVERSITY

DIVISION OF MATERIALS THEORY

DEPARTMENT OF PHYSICS AND ASTRONOMY

[†]E-mail: emil.beiersdorf@outlook.com

Abstract

The discoveries in applications of chirality in various areas of science seem to never cease to emerge. Chirality, being the property that some objects are geometrically distinguishable from their mirror image, is a tiny difference of vast importance. The fact that multiple biological structures are chiral is what permits life on Earth and its discovery had a severe impact on medical development. When the concept of *quantum chirality* was introduced, the connection between the chiral symmetry and the quantum states and operators that characterize quantum chirality was not particularly clear. It was shown that closed spin chains of an odd number of spins naturally had chiral states as eigenstates of a Hamiltonian describing Heisenberg and Dzyaloshinsky-Moriya (DM) interactions, and the symmetry of the system in direct relation to the chiral symmetry of the eigenstates quickly became of interest. The aim of this thesis is therefore to explore how quantum chirality is a chiral symmetry and to develop a scheme to create chiral states from systems that lack the required symmetry. The investigation showed that discretized probability current gives a good explanation to why the chiral states follow a chiral nature, but further examination is required in order to generalize a deeper connection between the probability current and the chiral states of spin chains. The results also indicated that it was possible to force open spin chains into purely chiral states, and into superpositions thereof, by time evolution. The scheme is still in its early stage and physical implementation and applications are yet to be explored.

Sammanfattning

Upptäckterna av tillämpningar av kiralitet inom ett flertal områden verkar ständigt öka i omfattning. Kiralitet är fenomenet att vissa objekt geometriskt kan särskiljas från sin spegelbild, vilket är en ringa skillnad men med väsentlig innebörd. Det faktum att flertalet biologiska strukturer är kirala är en förutsättning för liv på jorden och upptäckten av detta har haft en omfattande betydelse för medicinsk utveckling. När konceptet *kvantkiralitet* introducerades, var kopplingen mellan den kirala symmetrin och de kvantmekaniska tillstånden och operatorerna som utgör kvantkiralitet, inte trivial. Tidigare studier har visat att stängda spinnkedjor av ett udda antal spinn naturligt har kirala tillstånd som egentillstånd till en Hamiltonian beskrivande Heisenberg- och Dzyaloshinsky-Moriya-interaktioner. Att systemets symmetri stod i direkt relation till den kirala symmetrin av egentillstånden blev tidigt av intresse att undersöka. Syftet med denna kandidatuppsats är således att utforska en djupare förståelse till hur kvantkiralitet är en kiral symmetri samt utveckla en metod för hur kirala tillstånd kan drivas till att uppstå ur system som saknar den nödvändiga symmetrin. Resultaten visade att den diskretiserade sannolikhetsströmmen ger en god förklaring till varför de kirala tillstånden följer en kiral natur, men vidare efterforskning behövs för att kunna generalisera en djupare koppling mellan sannolikhetsströmmen och de kirala tillstånden hos spinnkedjor. Undersökningen indikerade också att det var möjligt att forcera en öppen spinnkedja till ett kiralt tillstånd, och till superpositioner därav, genom tidsutveckling. Metoden är fortfarande i sin tidiga utveckling och fysisk implementering samt tillämpningar väntar ännu på att upptäckas.

*I have not failed. I've just found 10,000
ways that won't work.*

THOMAS A. EDISON

Contents

1	Introduction	1
2	Background	3
2.1	Adding Spin in Quantum Mechanics	3
2.2	The Pauli Operators and Spin Convention	4
2.3	The Chiral Operators and States	6
2.4	A Λ -System	9
2.4.1	The Λ -System Hamiltonian	10
2.4.2	Time Evolution of the Λ -System	11
2.5	The Atomic Ramsay Interferometer	14
3	Approach and Results	15
3.1	How is Quantum Chirality Related to Chirality?	15
3.2	A Spin-1/2 Triangle	17
3.3	An Open Spin Chain	18
3.3.1	The Spin Chain Λ -System	18
3.3.2	The Spin Chain Hamiltonian	19
3.3.3	The Time Evolution Operator	21
3.3.4	Time Evolution Scheme	22
3.3.5	Time Evolution Into One of the Purely Chiral States	26
3.3.6	Restrictions for Ending Up in a Purely Chiral State	27
3.3.7	Time Evolution Into a Superposition of Nonzero Purely Chiral States	29
3.3.8	Speculation of Creating a Chiral Gate	30
4	Conclusions	31
5	Ethical Aspects	33
	References	35
	Appendix A Alternative Form of the Chiral Operators	38

1 Introduction

The concept of *chirality* has for a long time been a cornerstone in a variety of areas of science. Its versatility lies within the symmetry properties it characterizes, and can most generally be described by the geometrical distinction that occurs upon mirroring an object. This naturally occurring subtle distinction is a prerequisite for imperative processes in nature, such as chemical reactions including biological structures that make life on Earth possible. Amino acids [10], DNA and carbohydrates, all of which are chiral [21, 24], are a few of these vital structures. Chirality is also crucial in many pharmaceuticals since the biological effects of a molecule depend on its chiral geometry [14]. This is a cause of that chiral molecules are optically active, which means that they rotate plane polarized light in different directions [12]. In particle physics, chirality (for a massless particle also known as helicity [9]) is the distinction of a particle's spin and momentum being aligned (right chirality, positive helicity) or disaligned (left, negative) and is a central phenomenon to why the particle symmetry of the standard model is broken. This is due to that weak interactions depend on chirality [3, 26].

Moreover, chirality has a further reach in physics than just particle physics. It has been established that quantum effects such as the anomalous Hall effect can be used to control magnetism in thin layers of Cobalt, through enantiomeres (chiral molecules) [1, 25]. Further, this gave rise to a method of enantiomer separation through magnetic measurements [6]. It has also been discovered that specific spins are preferred when electrons move through chiral materials, giving rise to temperature-dependent [4] spin-dependent currents called Chiral-Induced Spin Selectivity (CISS) [29]. It is thereby evident that chirality gives rise to characteristic behaviour also on the quantum level.

A more subtle use of the concept is *quantum chirality*¹, which is characterized by quantum states and operators. The analogy to chirality is, however, quite subtle and it is not entirely clear what quantum chirality has to do with chiral symmetries. Trif et al. [27, 28] have previously shown that by considering an odd number of spin-1/2 particles in a closed spin chain, the eigenstates of the Hamiltonian describing Heisenberg and Dzyaloshinsky-Moriya interactions turned out to be the *chiral states*, obeying a spin-1 algebra together with the *chiral operators*. This shows that the chiral states and operators also have effects of importance and seem

¹The word quantum here is only used to describe that this description of chirality is realized through states and operators in accordance with the theory of quantum mechanics. The classical description of chirality still has applications on the quantum level, as was previously stated, and one should not confuse the term quantum chirality with the quantum mechanical effects that also arise from classical chirality.

to inherit the utility that the classical concept of chirality brings.

The aim for this thesis is to:

- A1.** Reach a deeper level of understanding for quantum chirality and its connection to the common knowledge of chiral symmetry,
- A2.** Examine the possibility to obtain quantum chirality through spin-interactions in open systems that lack the required symmetry in an attempt to develop a scheme that describes this.

The outline of the study is as follows. In section 2 we provide the background theory necessary to understand the line of work. Section 3 presents the scheme itself, carries out computations that are central in order to answer the aims of the thesis, and states the results. In section 4 the conclusions of the investigation is given, as well as some suggestions for applications of this work. Section 5 concludes the thesis with some ethical aspects and societal impacts.

2 Background

This section is devoted to providing the reader with the relevant background theory and terminology that this thesis is structured around.

2.1 Adding Spin in Quantum Mechanics

Spin-1/2 particles have spin that can be represented by a spin state that is an eigenstate of the spin projection operator S_z with eigenvalues $\pm\hbar/2$:

$$S_z |s, m\rangle = \hbar m |s, m\rangle, \quad m = \pm \frac{1}{2},$$

where $s = 1/2$ is the total spin and m the spin z -projection quantum numbers [20]. The states $|s, \pm 1/2\rangle$ represent the base kets that span an abstract complex vector space called the *Hilbert space*, commonly denoted \mathcal{H} , and is the state space of quantum mechanical systems. The operator S_z is said to act on this Hilbert space. When adding angular momentum quantities in the quantum mechanics formalism, one adds their operators as follows:

$$\begin{aligned} \vec{S}_j^{(j,N)} &\equiv \left(\bigotimes_{l=1}^{j-1} \mathbb{1}_l \right) \otimes \vec{S}_j \otimes \left(\bigotimes_{l=j+1}^N \mathbb{1}_l \right) \\ &= \mathbb{1}_1 \otimes \cdots \otimes \mathbb{1}_{j-1} \otimes \underbrace{\vec{S}_j}_{j^{th} \text{ place}} \otimes \mathbb{1}_{j+1} \otimes \cdots \otimes \mathbb{1}_N, \\ \vec{S} &= \sum_{j=1}^N \vec{S}_j^{(j,N)}, \end{aligned}$$

where $\mathbb{1}_i$ is interpreted as the unit operator (identity matrix) in Hilbert space \mathcal{H}_i . The resulting operator then operates on a Hilbert space corresponding to the tensor product of the respective Hilbert spaces that each operator operates on. Since the dimension of a vector space is the number of linearly independent basis vectors spanning the space, it becomes evident that the dimension of the composite Hilbert space is the product of the dimensions of each spin's Hilbert space:

$$\begin{aligned} \mathcal{H} &= \bigotimes_{j=1}^N \mathcal{H}_j, \\ \text{Dim}\{\mathcal{H}\} &= \prod_{j=1}^N \text{Dim}\{\mathcal{H}_j\}. \end{aligned}$$

For Hilbert spaces of spin-1/2 particles, this simplifies to $\text{Dim}\{\mathcal{H}\} = 2^N$. Similarly, states combine in the same manner:

$$\begin{aligned} |s_j, m_j\rangle &\in \mathcal{H}_j, \quad j = 1, \dots, N, \\ \implies |s, m\rangle &\in \text{Span}\left\{ \bigotimes_{j=1}^N |s_j, m_j\rangle \right\} = \mathcal{H}, \end{aligned} \quad (2.1)$$

where s and m add through the Clebsch-Gordan coefficients [20], and the notation $|s_j, m_j\rangle$ contains $2s + 1$ states. For a spin-1/2 particle, $s = 1/2$ and $m = \pm 1/2$ which has two states.

2.2 The Pauli Operators and Spin Convention

The convention is to denote the spin up state as $|0\rangle$ and the spin down state as $|1\rangle$, which using spinor formalism² translates to $\langle 0| \doteq (1 \ 0)$ and $\langle 1| \doteq (0 \ 1)$. The *Pauli operators* are 2×2 unitary spin-1/2 operators that, using the spinor formalism, look like [20]:

$$\begin{aligned} \sigma_1 = \sigma_x &\equiv |0\rangle\langle 1| + |1\rangle\langle 0| \doteq \begin{bmatrix} 0 & 1 \\ 1 & 0 \end{bmatrix}, \\ \sigma_2 = \sigma_y &\equiv -i|0\rangle\langle 1| + i|1\rangle\langle 0| \doteq \begin{bmatrix} 0 & -i \\ i & 0 \end{bmatrix}, \quad \vec{\sigma} \equiv \begin{pmatrix} \sigma_x \\ \sigma_y \\ \sigma_z \end{pmatrix}, \\ \sigma_3 = \sigma_z &\equiv |0\rangle\langle 0| - |1\rangle\langle 1| \doteq \begin{bmatrix} 1 & 0 \\ 0 & -1 \end{bmatrix}. \end{aligned} \quad (2.2)$$

The Pauli operators satisfy the *commutation* and *anti-commutation relations* [15]:

$$\begin{aligned} [\sigma_k, \sigma_l] &= 2i \sum_m \epsilon_{klm} \sigma_m, \quad k, l, m = x, y, z, \\ \{\sigma_k, \sigma_l\} &= 0, \quad k \neq l, \end{aligned}$$

where ϵ_{klm} is the *Levi-Civita (Permutation) symbol*:

$$\epsilon_{klm} = \begin{cases} +1, & \text{if } (k, l, m) \text{ is } (1, 2, 3), (3, 1, 2) \text{ or } (2, 3, 1), \\ 0, & \text{if } k = l, l = m \text{ or } m = k, \\ -1, & \text{if } (k, l, m) \text{ is } (1, 3, 2), (3, 2, 1) \text{ or } (2, 1, 3). \end{cases}$$

²The symbol \doteq is used as the conversion sign between operators on Dirac form and matrix form that constitutes the spinor formalism. The symbol should be interpreted as the matrix representation for the operator.

A *Hermitian matrix* H satisfies:

$$H^\dagger = H ,$$

where \dagger denotes the *Hermitian conjugate* which is the *transposed complex conjugate*. A *unitary matrix* U satisfies:

$$U^\dagger = U^{-1} ,$$

and a unitary operator preserves scalar products and therefore also norms [20]. This can be seen using the definition of unitarity by considering a scalar product between two states $|\alpha\rangle, |\beta\rangle$ before and after a unitary transformation $|k\rangle \rightarrow U|k\rangle$, $k = \alpha, \beta$:

$$\text{Before: } \langle\alpha|\beta\rangle ,$$

$$\text{After: } \langle\alpha|U^\dagger U|\beta\rangle = \langle\alpha|U^{-1}U|\beta\rangle = \langle\alpha|\beta\rangle ,$$

since $U^{-1}U = UU^{-1} = \mathbb{1}$ by definition of the *inverse* U^{-1} . The Pauli operators also satisfy:

$$\sigma_k^2 = \mathbb{1} , \quad k = x, y, z ,$$

which can be seen from that they are both Hermitian and unitary. With the definition of the matrix representation of the Pauli matrices, one can look at what they do when they act on the spin up and down states:

$$\begin{aligned} |0\rangle &\doteq \begin{pmatrix} 1 \\ 0 \end{pmatrix} , & |1\rangle &\doteq \begin{pmatrix} 0 \\ 1 \end{pmatrix} , \\ \sigma_x |0\rangle &= |1\rangle , & \sigma_x |1\rangle &= |0\rangle , \\ \sigma_y |0\rangle &= i |1\rangle , & \sigma_y |1\rangle &= -i |0\rangle , \\ \sigma_z |0\rangle &= |0\rangle , & \sigma_z |1\rangle &= -|1\rangle , \end{aligned} \tag{2.3}$$

where the matrix representations of the Pauli operators given by equation (2.2) were used.

2.3 The Chiral Operators and States

The *chiral operators* C_x , C_y and C_z are, for a three spin-1/2 interaction, defined³ as [27]:

$$\begin{aligned} C_x &\equiv -\frac{2}{3} \left(\vec{S}_1 \cdot \vec{S}_2 - 2\vec{S}_2 \cdot \vec{S}_3 + \vec{S}_3 \cdot \vec{S}_1 \right) , \\ C_y &\equiv \frac{2}{\sqrt{3}} \left(\vec{S}_1 \cdot \vec{S}_2 - \vec{S}_3 \cdot \vec{S}_1 \right) , \quad \vec{S} = \frac{\hbar}{2} \vec{\sigma} , \\ C_z &\equiv \frac{4}{\sqrt{3}} \vec{S}_1 \cdot \left(\vec{S}_2 \times \vec{S}_3 \right) , \end{aligned} \quad (2.4)$$

where the spin projection operators \vec{S}_i , $i = 1, 2, 3$ act on the respective Hilbert subspaces 1, 2, and 3. The chiral operators satisfy the commutation relations:

$$\begin{aligned} [C_k, C_l] &= 2i \sum_m \epsilon_{klm} C_m , \quad k, l, m = x, y, z , \\ [C_k, S_l] &= 0 , \quad k, l, m = x, y, z . \end{aligned}$$

The composite Hilbert space of a system of three spin-1/2 particles is, from equation (2.1), spanned by eight spin states represented by⁴ $|m_1, m_2, m_3\rangle$ with all possible combinations of m_1 , m_2 and m_3 :

$$\begin{cases} |000\rangle , & m = +\frac{3}{2} , \\ |100\rangle , |010\rangle , |001\rangle , & m = +\frac{1}{2} , \\ |110\rangle , |101\rangle , |011\rangle , & m = -\frac{1}{2} , \\ |111\rangle , & m = -\frac{3}{2} . \end{cases} \quad (2.5)$$

Looking at a normalized superposition of, say the $m = +1/2$ states:

$$\frac{1}{\sqrt{3}} \left[|100\rangle + |010\rangle + |001\rangle \right] ,$$

and applying the chiral operator [28] (setting $\hbar = 1$ during calculations):

$$C_z \left(\frac{1}{\sqrt{3}} \left[|100\rangle + |010\rangle + |001\rangle \right] \right) = 0 ,$$

³Here the inner products and vector products are only notation for a tensor product-like composition of the operators, see appendix A for explicit notation.

⁴From now on, the total spin denoted by s is omitted and only the spin projection quantum number m enters the spin state notation.

reveals that this state is a *chiral eigenstate* with eigenvalue zero. Creating superpositions with phases $\eta_{\pm} \equiv e^{\pm i \frac{2\pi}{3}}$:

$$\begin{aligned} & \frac{1}{\sqrt{3}} \left[|100\rangle + \eta_+ |010\rangle + \eta_- |001\rangle \right] , \\ & \frac{1}{\sqrt{3}} \left[|100\rangle + \eta_- |010\rangle + \eta_+ |001\rangle \right] , \end{aligned}$$

also turns out to be chiral eigenstates, with eigenvalues ± 1 [28]:

$$C_z \left(\frac{1}{\sqrt{3}} \left[|100\rangle + \eta_{\pm} |010\rangle + \eta_{\mp} |001\rangle \right] \right) = \pm \frac{1}{\sqrt{3}} \left[|100\rangle + \eta_{\pm} |010\rangle + \eta_{\mp} |001\rangle \right] .$$

Adapting the notation $|C_z, M\rangle$, C_z being the chiral eigenvalue to the C_z operator and M being the spin projection quantum number, and realizing that the same formalism applies to $M = -1/2$, we have arrived at the *symmetry-adapted basis* [28]:

$$\begin{aligned} \left| 0, +\frac{1}{2} \right\rangle &\equiv |W\rangle \equiv \frac{1}{\sqrt{3}} \left[|100\rangle + |010\rangle + |001\rangle \right] , \\ \left| \pm 1, +\frac{1}{2} \right\rangle &\equiv \frac{1}{\sqrt{3}} \left[|100\rangle + \eta_{\pm} |010\rangle + \eta_{\mp} |001\rangle \right] , \\ \left| 0, -\frac{1}{2} \right\rangle &\equiv |\overline{W}\rangle \equiv \frac{1}{\sqrt{3}} \left[|011\rangle + |101\rangle + |110\rangle \right] , \\ \left| \pm 1, -\frac{1}{2} \right\rangle &\equiv \frac{1}{\sqrt{3}} \left[|011\rangle + \eta_{\pm} |101\rangle + \eta_{\mp} |110\rangle \right] . \end{aligned} \tag{2.6}$$

The *chiral ladder operators* are defined as:

$$\begin{aligned} C_+ &\equiv C_x + iC_y = -\frac{2}{3} \left[(1 - i\sqrt{3}) \vec{S}_1 \cdot \vec{S}_2 - 2\vec{S}_2 \cdot \vec{S}_3 + (1 + i\sqrt{3}) \vec{S}_3 \cdot \vec{S}_1 \right] , \\ C_- &\equiv C_x - iC_y = -\frac{2}{3} \left[(1 + i\sqrt{3}) \vec{S}_1 \cdot \vec{S}_2 - 2\vec{S}_2 \cdot \vec{S}_3 + (1 - i\sqrt{3}) \vec{S}_3 \cdot \vec{S}_1 \right] , \end{aligned} \tag{2.7}$$

and it can be seen that they obey the commutation relations:

$$\begin{aligned} [C_z, C_{\pm}] &= \pm \hbar C_{\pm} , \\ [C^2, C_{\pm}] &= 0 , \end{aligned}$$

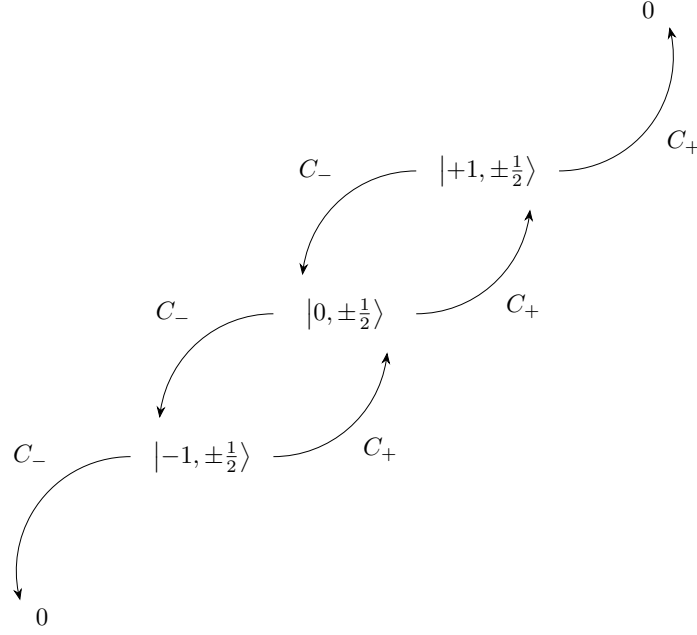


Fig. 1. The chiral ladder operators transitioning between states with different chirality. Note that the coefficients have been omitted.

where $C^2 \equiv C_x^2 + C_y^2 + C_z^2$ is to be interpreted as the total chiral operator. This means that the chiral ladder operators act on the chiral states similarly to how the angular momentum ladder operators act as raising and lowering operators [20]:

$$\begin{aligned} C_{\pm} |C_z, M\rangle &= \sqrt{(C \mp C_z)(C \pm C_z + 1)} \hbar |C_z \pm 1, M\rangle \\ &= \sqrt{2 - C_z(C_z \pm 1)} |C_z \pm 1, M\rangle , \end{aligned}$$

with $C = 1$, $s = 1/2$ being omitted in the state notation. This implies that the chiral operators obey a spin-1 algebra, and the ladder formalism is visualised in figure 1. The rest of the spin states in equation (2.5), $|000\rangle$ and $|111\rangle$ with spin projections $m = \pm 3/2$, are however not chiral. Although these states disappear when acted upon by the C_z operator, they are not considered to be chiral states since they do not obey the spin-1 algebra. These states actually also disappear when acted upon by the ladder operators:

$$\begin{aligned} C_{\pm} |000\rangle &= 0 , \\ C_{\pm} |111\rangle &= 0 . \end{aligned}$$

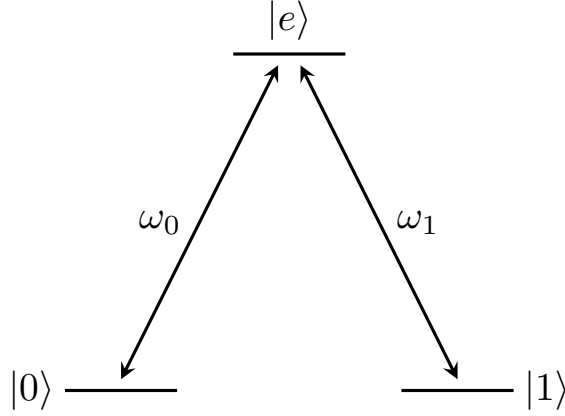


Fig. 2. The Λ -system where transitions to the excited state of each ground state is driven by polarized lasers of a characteristic parameter ω .

2.4 A Λ -System

A three level system is a system with three possible states. One can construct a so called Λ -*system* out of a three level system driving transitions between the internal states. Consider a general Λ -system, with two orthonormal *ground states*, denoted $|0\rangle$ and $|1\rangle$, and an *excited state*, denoted $|e\rangle$. The transitions $|0\rangle \leftrightarrow |e\rangle$ and $|1\rangle \leftrightarrow |e\rangle$ are driven⁵ respectively using polarized laser pulses characterized by the laser parameters $\omega_0(t)$ and $\omega_1(t)$. A specific case is when the laser parameters have the same time-dependence, which then can be factored out as a *pulse envelope* $\Omega(t)$, $t \in [0, \tau]$ [23], see figure 2. The laser parameters ω_0 and ω_1 are normalized as [11]:

$$|\omega_0|^2 + |\omega_1|^2 = 1, \quad (2.8)$$

and can therefore be parametrized as [23]:

$$\begin{aligned} \omega_0 &\stackrel{\text{def}}{=} \cos \frac{\theta}{2}, \\ \omega_1 &\stackrel{\text{def}}{=} -\sin \frac{\theta}{2} e^{i\phi}. \end{aligned}$$

⁵This is the reason why the configuration is called Λ -system; since there is no $|0\rangle \leftrightarrow |1\rangle$ transition the schematic picture of the configuration looks like the symbol Λ , see figure 2.

2.4.1 The Λ -System Hamiltonian

The Hamiltonian for the above Λ -system can be tuned (by laser tuning) to look like [23]:

$$H(t) = \Omega(t) \left(\omega_0 |e\rangle\langle 0| + \omega_1 |e\rangle\langle 1| + \text{H.c.} \right), \quad (2.9)$$

where H.c. denotes the Hermitian conjugate of the entire Hamiltonian. This particular Λ -system where the laser parameters ω_0 and ω_1 have the same time-dependence $\Omega(t)$, which is a real-valued function, assures that each part of the Hamiltonian is equally weighted during the laser exposure time. This coupled with the laser tuning assures that a *universal holonomic one-qubit gate* can be realized from the above Hamiltonian [23]. The Hamiltonian has a simpler expression in the basis constructed from the *bright* ($|b\rangle$) and *dark* ($|d\rangle$) ground states:

$$\begin{cases} |b\rangle \equiv \omega_0^* |0\rangle + \omega_1^* |1\rangle, \\ |d\rangle \equiv -\omega_1 |0\rangle + \omega_0 |1\rangle. \end{cases}$$

The basis vectors in the basis $\{|e\rangle, |b\rangle, |d\rangle\} \equiv \mathbb{E}$ are orthonormal, i.e. they satisfy $\langle k|l\rangle = \delta_{kl}$, $k, l = e, b, d$ which follows from the orthonormality of the states $|0\rangle, |1\rangle, |e\rangle$ and the normalization condition given by equation (2.8). In this basis, the Hamiltonian becomes:

$$\begin{aligned} H(t) &= \Omega(t) \left(\omega_0 |e\rangle\langle 0| + \omega_1 |e\rangle\langle 1| + \omega_0^* |0\rangle\langle e| + \omega_1^* |1\rangle\langle e| \right) \\ &= \Omega(t) \left[|e\rangle \left(\underbrace{\omega_0 \langle 0| + \omega_1 \langle 1|}_{\langle b|} \right) + \left(\underbrace{\omega_0^* |0\rangle + \omega_1^* |1\rangle}_{|b\rangle} \right) \langle e| \right] \\ &= \Omega(t) \left(|e\rangle\langle b| + |b\rangle\langle e| \right) \equiv \Omega(t) \mathcal{H}_\Lambda, \end{aligned}$$

where \mathcal{H}_Λ is the time-independent Λ -system Hamiltonian in the basis \mathbb{E} . It turns out that this Hamiltonian acts as a Pauli-X "bit flip" operator on the subspace $\mathbb{E}' \equiv \{|e\rangle, |b\rangle\}$ and acts trivially on the dark state, meaning:

$$\begin{aligned} \mathcal{H}_\Lambda |e\rangle &= \left(|e\rangle\langle b| + |b\rangle\langle e| \right) |e\rangle = |b\rangle, \\ \mathcal{H}_\Lambda |b\rangle &= \left(|e\rangle\langle b| + |b\rangle\langle e| \right) |b\rangle = |e\rangle, \end{aligned}$$

and

$$\mathcal{H}_\Lambda |d\rangle = 0 ,$$

where in the derivation, the orthonormality of the bright, dark and excited states have been used.

2.4.2 Time Evolution of the Λ -System

The time evolution of a system can, in the Schrödinger picture, be represented by the time evolution operator [20]:

$$\mathcal{U}(t, 0) = \exp\left(-\frac{i}{\hbar} \int_0^t H(t') dt'\right) ,$$

where $H(t)$ is the time-dependent Hamiltonian of the system. By design, this form assures that any time evolution operator is unitary. For a type of system where the time-dependence factors, as it does in the above Λ -system driven by the pulse envelope $\Omega(t)$, the Hamiltonian can be written as $H(t) = \Omega(t)\mathcal{H}_\Lambda$, and the time evolution operator becomes:

$$\begin{aligned} \int_0^t H(t') dt' &= \mathcal{H}_\Lambda \underbrace{\int_0^t \Omega(t') dt'}_{\equiv \mathcal{A}} = \mathcal{A}\mathcal{H}_\Lambda , \\ \implies \mathcal{U}(t, 0) &= \exp\left(-\frac{i}{\hbar} \mathcal{A}\mathcal{H}_\Lambda\right) , \end{aligned}$$

where \mathcal{A} usually is called the *pulse area* since for a specific choice of t the integral over the pulse envelope represents how much of the full pulse period that is used to drive the system. To see how the Λ -system evolves in time we therefore need to compute⁶ the time evolution operator for the Λ -system Hamiltonian. Let us consider the Hamiltonian expressed in the space $\mathbb{E} = \{|e\rangle, |b\rangle, |d\rangle\}$, setting $\hbar = 1$ during computations:

$$\begin{aligned} \mathcal{U}_\Lambda(t, 0) &= e^{-i\mathcal{A}\mathcal{H}_\Lambda} \\ &\stackrel{\text{Tay.}}{=} \sum_{n=0}^{\infty} \frac{1}{n!} \left(-i\mathcal{A}\mathcal{H}_\Lambda\right)^n . \end{aligned} \tag{2.10}$$

⁶Functions of operators in quantum mechanics are interpreted as the operators one get through Taylor expansion of said function [20].

Now, realizing that:

$$\begin{aligned}
 \mathcal{H}_\Lambda &= |e\rangle\langle b| + |b\rangle\langle e| \doteq \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \\
 \mathcal{H}_\Lambda^2 &\doteq \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{bmatrix}, \\
 \mathcal{H}_\Lambda^3 &\doteq \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{bmatrix} = \begin{bmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{bmatrix} \doteq \mathcal{H}_\Lambda, \\
 \implies \mathcal{H}_\Lambda^n &= \begin{cases} \mathbb{1}, & n = 0, \\ \mathcal{H}_\Lambda^2, & n \text{ even}, \\ \mathcal{H}_\Lambda, & n \text{ odd}, \end{cases}
 \end{aligned} \tag{2.11}$$

and grouping even and odd terms in the sum (2.10) we get:

$$\begin{aligned}
 \mathcal{U}_\Lambda(t, 0) &= \underset{3 \times 3}{\mathbb{1}} + \underbrace{\left(-\frac{\mathcal{A}^2}{2!} + \frac{\mathcal{A}^4}{4!} + \dots \right)}_{= \cos \mathcal{A} - 1} \mathcal{H}_\Lambda^2 - i \underbrace{\left(\mathcal{A} - \frac{\mathcal{A}^3}{3!} + \frac{\mathcal{A}^5}{5!} + \dots \right)}_{= \sin \mathcal{A}} \mathcal{H}_\Lambda \\
 &= \underset{3 \times 3}{\mathbb{1}} - \left(1 - \cos \mathcal{A} \right) \mathcal{H}_\Lambda^2 - i \sin \mathcal{A} \mathcal{H}_\Lambda,
 \end{aligned}$$

where the Taylor expansions for sine and cosine were used and the three by three unity is the unit operator of \mathbb{E} . Noting from the matrix representation of the Λ -Hamiltonian (2.11):

$$\mathcal{H}_\Lambda^2 \doteq \begin{bmatrix} 1 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 0 \end{bmatrix} \doteq |e\rangle\langle e| + |b\rangle\langle b| \doteq \underset{3 \times 3}{\mathbb{1}} - |d\rangle\langle d|,$$

the time evolution operator can be written as:

$$\mathcal{U}_\Lambda(t, 0) = |d\rangle\langle d| + \cos \mathcal{A} \left(|e\rangle\langle e| + |b\rangle\langle b| \right) - i \sin \mathcal{A} \left(|e\rangle\langle b| + |b\rangle\langle e| \right). \tag{2.12}$$

Let us now consider the time evolution of each of the ground states $|b\rangle$, $|d\rangle$ of the Λ -system:

$$\begin{aligned}
 \mathcal{U}_\Lambda(t, 0) |d\rangle &= |d\rangle, \\
 \mathcal{U}_\Lambda(t, 0) |b\rangle &= \cos \mathcal{A} |b\rangle - i \sin \mathcal{A} |e\rangle,
 \end{aligned}$$

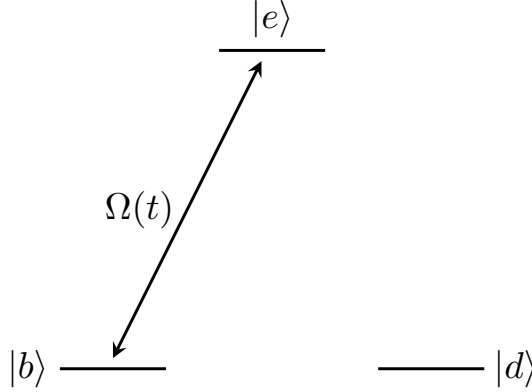


Fig. 3. Time evolution of the Λ -system with the bright and dark states, $|b\rangle$, $|d\rangle$ as ground states and $|e\rangle$ as excited state. The population of the dark state stays in the dark state with time and that of the bright state oscillates, with the π -periodic Rabi frequency $\Omega(t)$, between the bright state and the excited state.

which tells us that if the system initially populates the dark state $|d\rangle$ then it stays there and does not populate the excited state $|e\rangle$ at all, and that the population of the bright state $|b\rangle$ oscillates to the excited state $|e\rangle$ and back with some period time given by the pulse area \mathcal{A} . These results were derived without constraints on the pulse shape so they hold for any $\Omega(t)$ and then also for any exposure time t under which the system is being evolved. We can see that when choosing the exposure time τ such that⁷ $\mathcal{A} = \pi/2$ and again such that $\mathcal{A} = \pi$, we get:

$$\begin{aligned}\mathcal{U}_\Lambda(t_{\frac{\pi}{2}}, 0) |b\rangle &= \cos \frac{\pi}{2} |b\rangle - i \sin \frac{\pi}{2} |e\rangle = -i |e\rangle, \\ \mathcal{U}_\Lambda(t_\pi, 0) |b\rangle &= \cos \pi |b\rangle - i \sin \pi |e\rangle = -|b\rangle,\end{aligned}$$

which means that by driving the $|b\rangle \leftrightarrow |e\rangle$ transition through a $\frac{\pi}{2}$ -pulse the system populates only the excited state if it initially populated the bright state. Further, transitioning through a π -pulse, the system only populates the bright state again, (where the only difference is a global phase which does not matter) if it initially populated the bright state. The oscillation frequency for the transition $|b\rangle \leftrightarrow |e\rangle$ is often called the *Rabi frequency* which is π -periodic and characterized by the pulse envelope $\Omega(t)$ [23]. The time evolution of the Λ -system in the space \mathbb{E} is shown in figure 3.

⁷The terminology for evolving a system in time through a pulse, dependent of the exposure time, t , such that $\mathcal{A} = \int_0^t \Omega(t') dt' = \text{const}$, is "To apply the Hamiltonian as a π -pulse, $\frac{\pi}{2}$ -pulse" etc, depending on the constant.

2.5 The Atomic Ramsay Interferometer

In an article about quantum entanglement, Serge Haroche et al. [17] showed that it is possible to force a system, starting in a determinate state, to end up in a superposition of states using cavities. A two level atom, having the energy level states $|g\rangle$ (ground state) and $|e\rangle$ (excited state), was considered. By applying a microwave laser pulse with a specific pulse area and frequency, it could be seen that for the system starting in one of the two states, it would end up in [17]:

$$\begin{aligned} |g\rangle &\rightarrow \frac{1}{\sqrt{2}}(|e\rangle + |g\rangle) , \\ |e\rangle &\rightarrow \frac{1}{\sqrt{2}}(|e\rangle - |g\rangle) . \end{aligned}$$

This means that by starting in one of the two energy levels and applying laser pulses of specific parameters, the system then ends up in a superposition of the possible energy states, and the Ramsay interferometer can then be used to create superpositions of states. This was shown for atoms prepared in circular Rydberg states.

3 Approach and Results

In this section, the approach of the thesis is carried out and the results are presented as we advance.

3.1 How is Quantum Chirality Related to Chirality?

We would like to see a connection between the geometrical description that classically is referred to as chirality and the states that describe quantum chirality. The probability current could possibly provide us with precisely that. The probability current, \vec{j} , is to be interpreted as how a system's probability to appear in different states varies with time and or spatially. It is given by ($\hbar = 1$ and ignoring mass):

$$\vec{j} = \frac{1}{2i} \left(\Psi^* \nabla \Psi - \Psi \nabla \Psi^* \right) , \quad (3.1)$$

for a system's wavefunction Ψ . Any wavefunction can be expressed as a spatially dependent positive part $R(\vec{x})$ and complex phase:

$$\Psi(\vec{x}) = R(\vec{x}) e^{iS(\vec{x})} , \quad (3.2)$$

where $R^2(\vec{x})$ is the probability density of the system and $S(\vec{x})$ is real. Computing the probability current for the wavefunction (3.2), one finds:

$$\vec{j} = R^2(\vec{x}) \nabla S(\vec{x}) ,$$

which implies:

$$\frac{\vec{j}}{|\Psi|^2} = \frac{R^2(\vec{x}) \nabla S(\vec{x})}{R^2(\vec{x})} = \nabla S(\vec{x}) .$$

We then see that the probability current, up to a real constant, solely is governed by the phase of the wave function! In the case of our chiral states, which is a discrete set of basis kets, we need to look at the discretization of equation (3.1). The gradient of a discrete wavefunction can be approximated as:

$$\nabla \Psi \approx \frac{\Psi(x + \Delta x) - \Psi(x)}{\Delta x} \stackrel{\text{Discr.}}{=} \frac{\Psi_{l+1} - \Psi_l}{l+1-l} = \Psi_{l+1} - \Psi_l ,$$

for the wave function at state l . For this discretized gradient, the discrete probability current becomes:

$$\begin{aligned} \vec{j}_l &= \frac{1}{2i} \left[\Psi_l^* (\Psi_{l+1} - \Psi_l) - \Psi_l (\Psi_{l+1}^* - \Psi_l^*) \right] \\ &= \frac{1}{2i} \left(\Psi_l^* \Psi_{l+1} - \Psi_l \Psi_{l+1}^* \right) . \end{aligned} \quad (3.3)$$

The wavefunction is supposed to be interpreted as a probability amplitude for some corresponding state, such that its absolute value squared is the probability density, in accordance with the Dirac formalism through the completeness relation:

$$|\Psi\rangle = \mathbb{1} |\Psi\rangle = \int_a da |a\rangle \underbrace{\langle a|\Psi\rangle}_{\equiv \Psi(a)} = \int_a da \Psi(a) |a\rangle , \quad \text{Continuous Spectrum ,}$$

$$|\Psi\rangle = \mathbb{1} |\Psi\rangle = \sum_a |a\rangle \underbrace{\langle a|\Psi\rangle}_{\equiv \Psi(a)} = \sum_a \Psi(a) |a\rangle , \quad \text{Discrete Spectrum ,}$$

for some (complete) basis vectors⁸ $|a\rangle$. For our chiral states $|\pm 1\rangle$ we see that:

$$|+1\rangle = \underbrace{1}_{\Psi_1^+} |100\rangle + \underbrace{\eta}_{\Psi_2^+} |010\rangle + \underbrace{\eta^2}_{\Psi_3^+} |001\rangle ,$$

$$|-1\rangle = \underbrace{1}_{\Psi_1^-} |100\rangle + \underbrace{\eta^2}_{\Psi_2^-} |010\rangle + \underbrace{\eta}_{\Psi_3^-} |001\rangle ,$$

So let us compute the probability current at each of the states $|100\rangle, |010\rangle, |001\rangle$ for both $|\pm 1\rangle$ using equation (3.3):

$$|+1\rangle; \quad \Psi_1^+ = 1, \Psi_2^+ = \eta, \Psi_3^+ = \eta^2 , \quad |-1\rangle; \quad \Psi_1^- = 1, \Psi_2^- = \eta^2, \Psi_3^- = \eta ,$$

$$\text{at } |100\rangle: \quad j_1 = \frac{1}{2i} (\eta - \eta^2) = \frac{\sqrt{3}}{2} , \quad \text{at } |100\rangle: \quad j_1 = -\frac{1}{2i} (\eta - \eta^2) = -\frac{\sqrt{3}}{2} ,$$

$$\text{at } |010\rangle: \quad j_2 = \frac{1}{2i} (\eta - \eta^2) = \frac{\sqrt{3}}{2} , \quad \text{at } |010\rangle: \quad j_2 = -\frac{1}{2i} (\eta - \eta^2) = -\frac{\sqrt{3}}{2} ,$$

$$\text{at } |001\rangle: \quad j_3 = \frac{1}{2i} (\eta - \eta^2) = \frac{\sqrt{3}}{2} , \quad \text{at } |001\rangle: \quad j_3 = -\frac{1}{2i} (\eta - \eta^2) = -\frac{\sqrt{3}}{2} ,$$

where we have used:

$$\eta - \eta^2 = \eta - \eta^{-1} = e^{i\frac{2\pi}{3}} - e^{-i\frac{2\pi}{3}} = 2i \sin \frac{2\pi}{3} = i\sqrt{3} .$$

We then see that for the $|+1\rangle$ state with the positive chiral eigenvalue $C_z = +1$, all spin states have positive and equal probability current, which corresponds to a right-handed chirality! Similarly, for the $|-1\rangle$ state with the negative chiral eigenvalue $C_z = -1$, all spin states have negative and equal probability current, with a left-handed chirality. This can be visualized by assigning a point in the complex plane to each of the states $|100\rangle, |010\rangle, |001\rangle$, and looking at the probability currents, see figure 4.

⁸The set of all eigenvalues to a set of basis states in a system is usually referred to as the system's spectrum.

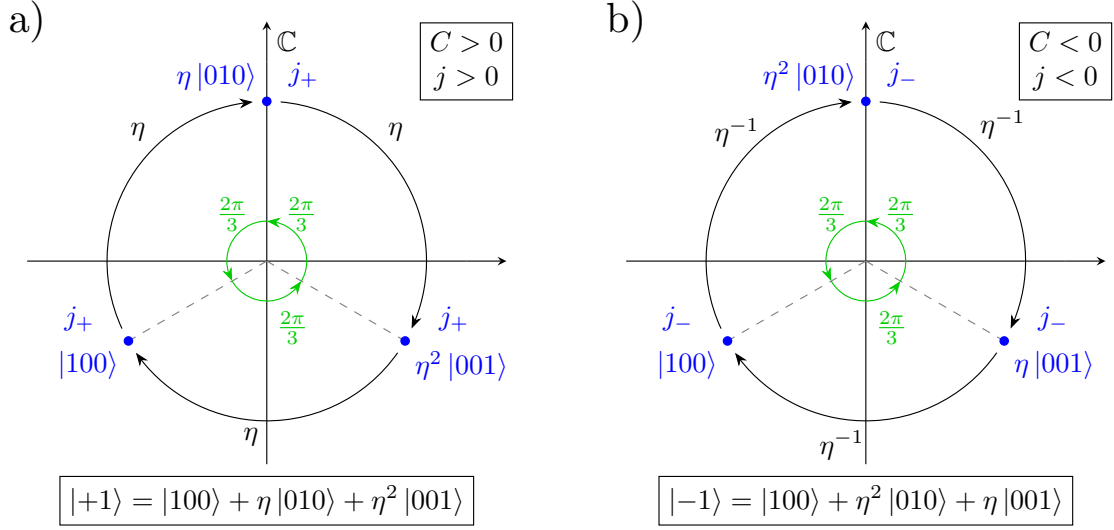


Fig. 4. Visualization of the a) right-handed and b) left-handed chirality of the quantum chiral states $|\pm 1\rangle$. Notice that for the right-handed chirality the phase increases with η in each step ($\eta^3 = 1$) and for the left-handed chirality the phase decreases with η for each step in the same direction ($\eta^{-1} = \eta^2$, $\eta^{-2} = \eta$)

3.2 A Spin-1/2 Triangle

A system of molecules that at low energies behave as one or a few interacting spins is called a *molecular nanomagnet* [28]. An example is three interacting spin-1/2-particles in a closed configuration, see figure 5. Without external applied fields, the Hamiltonian for such a system, considering *isotropic Heisenberg interaction* (J) and *spin orbital interaction* (SOI) called *Dzyaloshinsky-Moriya interaction* (D), looks like [28]:

$$H = \underbrace{\sum_{i=1}^{N=3} J_{i i+1} \vec{S}_i \cdot \vec{S}_{i+1}}_{\text{HI}} + \underbrace{\sum_{i=1}^{N=3} \vec{D}_{i i+1} \cdot (\vec{S}_i \times \vec{S}_{i+1})}_{\text{DM}}, \quad (3.4)$$

where HI stands for Heisenberg Interaction and DM for Dzyaloshinsky-Moriya interaction [5, 13]. The eigenstates of Hamiltonian (3.4) are with some degeneration the eight states $|000\rangle$, $|111\rangle$, $|0, \pm \frac{1}{2}\rangle$, $|\pm 1, \pm \frac{1}{2}\rangle$, all of which except the $m = \pm 3/2$ states are chiral states, see section 2.3. Trif et al. [27, 28] showed that it is possible to split the degeneration and drive transitions between the chiral states, using an electric field to transition between the different chiral eigenvalues and a magnetic field to transition between the different spin projections [27], indicating good control over both spin and chirality in such a system.

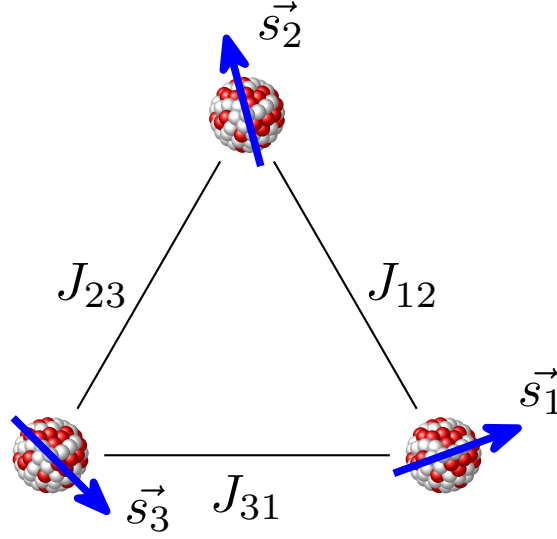


Fig. 5. Three spin-1/2-particles interacting with coupling strengths J_{i+1} . The interaction strengths are to be interpreted as stronger for closer particles.

3.3 An Open Spin Chain

It would then be interesting to see if it is possible to create chirality in an open spin chain, see figure 6, already known to have a physical Hamiltonian when considering *isotropic XY* and DM interactions [16, 31]. The open spin chain lack the triangular symmetry needed for chirality to occur, but the idea is to force the system into a chiral state using some manipulation.

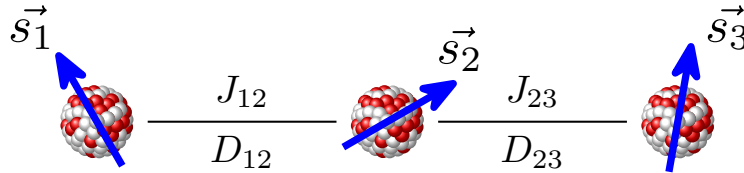


Fig. 6. A chain of three spin-1/2-particles with coupling constants J_{i+1} (XY) and D_{i+1} (DM).

3.3.1 The Spin Chain Λ -System

A Λ -system can be created by looking at the subspace with two spin up and one spin down of the spin chain, $\{|010\rangle, |100\rangle, |001\rangle\} \equiv S_{2\uparrow 1\downarrow}$ where $|010\rangle$ acts as the excited state and $|100\rangle, |001\rangle$ act as ground states, see figure 7. Each of the ground states can be coupled to the excited state through polarized laser pulses

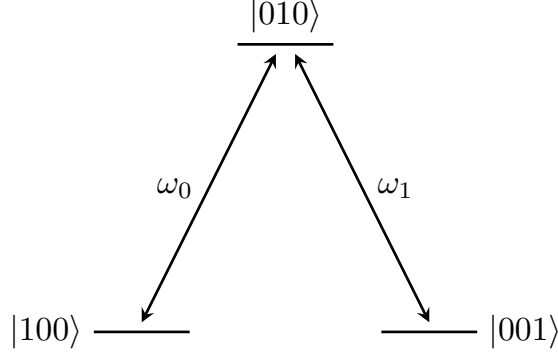


Fig. 7. The Λ -system for the open spin chain.

of different laser parameters ω_0 and ω_1 , see section 2.4. We see from figure 7 that the first laser parameter ω_0 drives the transition $|10\rangle \leftrightarrow |01\rangle$ of the first two qubits (particle one and two), and the second laser parameter ω_1 drives the transition $|01\rangle \leftrightarrow |10\rangle$ of the last two qubits (particles two and three). The Hamiltonian for this Λ -system (see equation (2.9)), is in the basis $\mathbb{S}_{2\uparrow 1\downarrow}$, given by:

$$\begin{aligned}
 H_{\Lambda} &= \Omega(t) \underbrace{\left\{ \omega_0 |010\rangle\langle 100| + \omega_1 |010\rangle\langle 001| + \text{H.c.} \right\}}_{\mathcal{H}_{\Lambda}^{\mathbb{S}_{2\uparrow 1\downarrow}}} \\
 &\doteq \Omega(t) \begin{bmatrix} 0 & \omega_0 & \omega_1 \\ \omega_0^* & 0 & 0 \\ \omega_1^* & 0 & 0 \end{bmatrix}, \tag{3.5}
 \end{aligned}$$

where $\Omega(t)$ is the time-dependent pulse envelope of the laser switched on at a time $t = 0$ and off at a time t .

3.3.2 The Spin Chain Hamiltonian

The Hamiltonian for the spin chain system, considering isotropic XY and DM interactions, takes the form [31]:

$$\begin{aligned}
 \mathcal{H}_{JD} = \frac{1}{2} &\left[J_{12} \left(\sigma_x \otimes \sigma_x \otimes \mathbb{1} + \sigma_y \otimes \sigma_y \otimes \mathbb{1} \right) + D_{12} \left(\sigma_x \otimes \sigma_y \otimes \mathbb{1} - \sigma_y \otimes \sigma_x \otimes \mathbb{1} \right) + \right. \\
 &\left. + J_{23} \left(\mathbb{1} \otimes \sigma_x \otimes \sigma_x + \mathbb{1} \otimes \sigma_y \otimes \sigma_y \right) + D_{23} \left(\mathbb{1} \otimes \sigma_x \otimes \sigma_y - \mathbb{1} \otimes \sigma_y \otimes \sigma_x \right) \right]. \tag{3.6}
 \end{aligned}$$

We are, however, interested in this Hamiltonian expressed in the spin basis of which the Λ -system Hamiltonian 3.5 is constructed, and therefore express \mathcal{H}_{JD} in

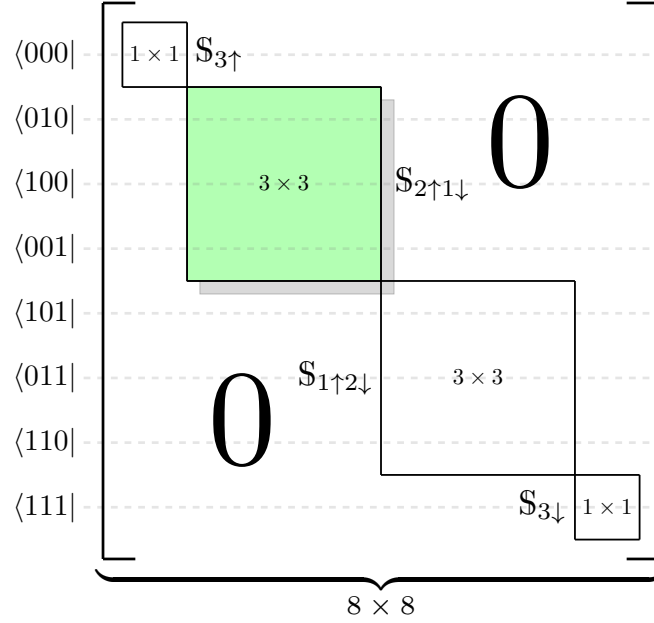


Fig. 8. Visual representation of the 8×8 spin Hamiltonian in the basis spanned by the eight spin states given by equation (2.5). Projecting on the subspace $\mathbb{S}_{2\uparrow 1\downarrow}$ (of two spin up and one spin down, shown as green), we get the Λ -Hamiltonian shown in equation (3.5), which is the subspace of interest to project the JD-Hamiltonian on. The block diagonal form is a result of the commutation relation $[\mathcal{H}_{JD}, \mathbf{Z}] = 0$, where $\mathbf{Z} \equiv \sigma_z \otimes \mathbb{1} \otimes \mathbb{1} + \mathbb{1} \otimes \sigma_z \otimes \mathbb{1} + \mathbb{1} \otimes \mathbb{1} \otimes \sigma_z$. This is in accordance with the $(2j+1)$ -dimensional irreducible representation of the rotation operator for angular momentum j , where the matrix representation of an arbitrary rotation operator in ket space can be brought to a block diagonal form for a suitable choice of basis as a result of the commutation relations $[J^2, J_k]$, $k = x, y, z$. [20].

the $\mathbb{S}_{2\uparrow 1\downarrow}$ basis by applying the projection operator $\mathcal{P}_{\mathbb{S}_{2\uparrow 1\downarrow}}$:

$$\begin{aligned}
 \mathcal{H}_{JD} &= \mathcal{P}_{\mathbb{S}_{2\uparrow 1\downarrow}} \mathcal{H}_{JD} \mathcal{P}_{\mathbb{S}_{2\uparrow 1\downarrow}} , \\
 \mathcal{P}_{\mathbb{S}_{2\uparrow 1\downarrow}} &= |010\rangle\langle 010| + |100\rangle\langle 100| + |001\rangle\langle 001| , \\
 \begin{cases} \mathcal{H}_{JD} |010\rangle = (J_{12} - iD_{12}) |100\rangle + (J_{23} + iD_{23}) |001\rangle , \\ \mathcal{H}_{JD} |100\rangle = (J_{12} + iD_{12}) |010\rangle , \\ \mathcal{H}_{JD} |001\rangle = (J_{23} - iD_{23}) |010\rangle , \end{cases} \\
 \Rightarrow \mathcal{H}_{JD} &= \begin{bmatrix} 0 & J_{12} + iD_{12} & J_{23} - iD_{23} \\ J_{12} - iD_{12} & 0 & 0 \\ J_{23} + iD_{23} & 0 & 0 \end{bmatrix} ,
 \end{aligned}$$

where in the derivation of \mathcal{H}_{JD} , equations (2.3) were used. Now comparing the JD-Hamiltonian expressed in the $\mathbb{S}_{2\uparrow 1\downarrow}$ basis with the Λ -Hamiltonian (equation (3.5), which is expressed in the same spin basis, see figure 8), we then see that for the choices:

$$\begin{cases} \omega_0 = J_{12} + iD_{12} , \\ \omega_1 = J_{23} - iD_{23} , \end{cases} \quad (3.7)$$

the Λ -Hamiltonian \mathcal{H}_{Λ} resembles the spin chain interaction Hamiltonian \mathcal{H}_{JD} as desired.

3.3.3 The Time Evolution Operator

The spin chain system, starting in some state $|\psi\rangle$, is by the laser pulses evolved in time to a new state $|\psi(t)\rangle$. As described in section 2.4, the bright state, $|b\rangle$, is completely driven up to the excited state, $|e\rangle$, when the pulse area is precisely equal to $\pi/2$. If the time of exposure of the laser pulse instead, for some starting state $|\psi\rangle$, is chosen such that the pulse area is in the interval $(0, \pi/2)$, the system can be "frozen" in a state $|f\rangle$ that lies between the excited and the starting state. Using the Hamiltonians derived in the previous chapter we can, using the theory developed in 2.4.2, define the time evolution operator for the spin chain Λ -system. First we need to consider our spin chain Λ -Hamiltonian (3.5) expressed in the basis $\mathbb{E} \equiv \{|e\rangle, |b\rangle, |d\rangle\}$. In this system the ground states $|0\rangle, |1\rangle$ are $|100\rangle, |001\rangle$, and the excited state is $|010\rangle$ so the $|b\rangle, |d\rangle, |e\rangle$ states are for our spin chain given by:

$$\begin{cases} |e\rangle = |010\rangle , \\ |b\rangle = \omega_0^* |100\rangle + \omega_1^* |001\rangle , \\ |d\rangle = -\omega_1 |100\rangle + \omega_0 |001\rangle , \end{cases} \quad \Leftrightarrow \quad \begin{cases} |010\rangle = |e\rangle , \\ |100\rangle = \omega_0 |b\rangle - \omega_1^* |d\rangle , \\ |001\rangle = \omega_1 |b\rangle + \omega_0^* |d\rangle . \end{cases} \quad (3.8)$$

We can then rewrite our Hamiltonian (3.5) as:

$$\begin{aligned} H_{\Lambda} &= \Omega(t) \left\{ \omega_0 |010\rangle\langle 100| + \omega_1 |010\rangle\langle 001| + \omega_0^* |100\rangle\langle 010| + \omega_1^* |001\rangle\langle 010| \right\} \\ &= \Omega(t) \left\{ \underbrace{|010\rangle}_{|e\rangle} \left(\underbrace{\omega_0 \langle 100| + \omega_1 \langle 001|}_{|b\rangle} \right) + \left(\underbrace{\omega_0^* |100\rangle + \omega_1^* |001\rangle}_{|b\rangle} \right) \underbrace{\langle 010|}_{|e\rangle} \right\} \\ \Rightarrow H_{\Lambda} &= \Omega(t) \left\{ |e\rangle\langle b| + |b\rangle\langle e| \right\} = \Omega(t) \mathcal{H}_{\Lambda} , \end{aligned}$$

which by design is precisely the Λ -Hamiltonian derived in equation (2.11), meaning that the theory in section 2.4 applies to our spin chain system. Now, acting with

the time evolution operator on basis \mathbb{E} (equation (2.12)) on a superposition of the bright and dark states yields:

$$\begin{aligned} \mathcal{U}_\Lambda(\alpha|b\rangle + \beta|d\rangle) &= \left[|d\rangle\langle d| + \cos \mathcal{A}(|e\rangle\langle e| + |b\rangle\langle b|) - i \sin \mathcal{A}(|e\rangle\langle b| + |b\rangle\langle e|) \right] \\ &\quad \times (\alpha|b\rangle + \beta|d\rangle) \\ &= \alpha(\cos \mathcal{A}|b\rangle - i \sin \mathcal{A}|e\rangle) + \beta|d\rangle. \end{aligned} \quad (3.9)$$

Equation (3.9) describes the time evolution of the starting state $|\psi\rangle$ which then looks like an oscillation between the bright and excited states whereas the dark state stays constant in time, just as expected (section 2.4.2).

3.3.4 Time Evolution Scheme

With the time evolution of the spin chain Λ -system known, we now want to find the exposure time τ (pulse area \mathcal{A}) and the constants in the JD-Hamiltonian such that the laser pulse evolves the starting state $|\psi\rangle$ into a chiral state. This would then propose a scheme on how to create *synthetic chirality* from a system that completely lacks the necessary symmetry by controlling its time evolution. Let us apply the inverse time evolution operator to a general superposition of the chiral states⁹:

$$\mathcal{U}^\dagger(t, 0)(\gamma|0\rangle + \delta|+1\rangle + \epsilon|-1\rangle). \quad (3.10)$$

The idea is then to find coefficients such that equation (3.10) lives in the subspace of the bright and dark states. To carry out the computation, we need to express the chiral states in the \mathbb{E} basis. To do this, we simply need to switch basis from $\mathbb{W} \equiv \{|0\rangle, |\pm 1\rangle\}$ to $\mathbb{E} \equiv \{|e\rangle, |b\rangle, |d\rangle\}$, the simplest way being through $\mathbb{S}_{2\uparrow 1\downarrow} \equiv \{|010\rangle, |100\rangle, |001\rangle\}$ since we know how to switch from \mathbb{E} to $\mathbb{S}_{2\uparrow 1\downarrow}$ and from $\mathbb{S}_{2\uparrow 1\downarrow}$ to \mathbb{W} . From equation (3.8) and the construction of the symmetry-adapted basis (equation (2.6)) we see:

$$\begin{pmatrix} |010\rangle \\ |100\rangle \\ |001\rangle \end{pmatrix} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \omega_0 & -\omega_1^* \\ 0 & \omega_1 & \omega_0^* \end{bmatrix} \begin{pmatrix} |e\rangle \\ |b\rangle \\ |d\rangle \end{pmatrix}, \quad \begin{pmatrix} |0\rangle \\ |+1\rangle \\ |-1\rangle \end{pmatrix} = \frac{1}{\sqrt{3}} \begin{bmatrix} 1 & 1 & 1 \\ \eta & 1 & \eta^2 \\ \eta^2 & 1 & \eta \end{bmatrix} \begin{pmatrix} |010\rangle \\ |100\rangle \\ |001\rangle \end{pmatrix}.$$

⁹The $+\frac{1}{2}$ will from now on be omitted, the state $|0\rangle$ is understood to be an abbreviation for the chiral state $|0, +1/2\rangle$ and not the regular spin up projection state or the first ground state in the Λ -system..

This allows us to express the base kets of the \mathbb{E} basis in the base kets of the \mathbb{W} basis as:

$$\begin{pmatrix} |0\rangle \\ | +1\rangle \\ | -1\rangle \end{pmatrix} = \underbrace{\frac{1}{\sqrt{3}} \begin{bmatrix} 1 & 1 & 1 \\ \eta & 1 & \eta^2 \\ \eta^2 & 1 & \eta \end{bmatrix} \begin{bmatrix} 1 & 0 & 0 \\ 0 & \omega_0 & -\omega_1^* \\ 0 & \omega_1 & \omega_0^* \end{bmatrix}}_{\equiv \mathcal{M}} \begin{pmatrix} |e\rangle \\ |b\rangle \\ |d\rangle \end{pmatrix},$$

$$\Rightarrow \begin{pmatrix} |e\rangle \\ |b\rangle \\ |d\rangle \end{pmatrix} = \mathcal{M}^{-1} \begin{pmatrix} |0\rangle \\ | +1\rangle \\ | -1\rangle \end{pmatrix},$$

where:

$$\mathcal{M} = \frac{1}{\sqrt{3}} \begin{bmatrix} 1 & \omega_0 + \omega_1 & \omega_0^* - \omega_1^* \\ \eta & \omega_0 + \eta^2 \omega_1 & \eta^2 \omega_0^* - \omega_1^* \\ \eta^2 & \omega_0 + \eta \omega_1 & \eta \omega_0^* - \omega_1^* \end{bmatrix},$$

$$\mathcal{M}^{-1} = \frac{1}{\sqrt{3}} \begin{bmatrix} 1 & \frac{1-\eta}{1+2\eta} & -\frac{2+\eta}{1+2\eta} \\ \omega_0^* + \omega_1^* & \omega_0^* - \omega_1^* \frac{2+\eta}{1+2\eta} & \omega_0^* + \omega_1^* \frac{1-\eta}{1+2\eta} \\ \omega_0 - \omega_1 & -\omega_0 \frac{2+\eta}{1+2\eta} - \omega_1 & \omega_0 \frac{1-\eta}{1+2\eta} - \omega_1 \end{bmatrix}.$$

The basis switch then yields:

$$\gamma |0\rangle + \delta | +1\rangle + \epsilon | -1\rangle = p_e |e\rangle + p_b |b\rangle + p_d |d\rangle, \quad (3.11)$$

$$\begin{cases} |0\rangle = \frac{1}{\sqrt{3}} \left[|e\rangle + (\omega_0 + \omega_1) |b\rangle + (\omega_0^* - \omega_1^*) |d\rangle \right], \\ | +1\rangle = \frac{1}{\sqrt{3}} \left[\eta |e\rangle + (\omega_0 + \eta^2 \omega_1) |b\rangle + (\omega_0^* \eta^2 - \omega_1^*) |d\rangle \right], \\ | -1\rangle = \frac{1}{\sqrt{3}} \left[\eta^2 |e\rangle + (\omega_0 + \eta \omega_1) |b\rangle + (\omega_0^* \eta - \omega_1^*) |d\rangle \right], \end{cases}$$

$$\Rightarrow \begin{cases} p_e &= \frac{1}{\sqrt{3}} \left[\gamma + \eta \delta + \eta^2 \epsilon \right], \\ p_b &= \frac{1}{\sqrt{3}} \left[(\gamma + \delta + \epsilon) \omega_0 + (\gamma + \eta^2 \delta + \eta \epsilon) \omega_1 \right], \\ p_d &= \frac{1}{\sqrt{3}} \left[(\gamma + \eta^2 \delta + \eta \epsilon) \omega_0^* - (\gamma + \delta + \epsilon) \omega_1^* \right]. \end{cases} \quad (3.12)$$

Equation (3.10) now becomes:

$$\mathcal{U}^\dagger(t, 0) \left(\gamma |0\rangle + \delta | +1\rangle + \epsilon | -1\rangle \right) = \mathcal{U}_\Lambda^\dagger(t, 0) \left(p_e |e\rangle + p_b |b\rangle + p_d |d\rangle \right),$$

and the reason for the basis switch is that we then can use the time evolution operator (2.12). We want the created superposition of the chiral states to be normalized after time evolution, so we require that p_e, p_b, p_d are normalized. We check if they are, for any choice of γ, δ, ϵ such that $|\gamma|^2 + |\delta|^2 + |\epsilon|^2 = 1$:

$$|p_e|^2 + |p_b|^2 + |p_d|^2 = 1, \quad (3.13)$$

which we see they are, where we plugged in the p's from relations (3.12) and simplified the expression using equation (2.8), the normalization of γ, δ, ϵ and the fact that:

$$\begin{aligned} 1 + \eta_+ + \eta_- &= 1 + e^{i\frac{2\pi}{3}} + e^{-i\frac{2\pi}{3}} \\ &= 1 + \cos \frac{2\pi}{3} + i \sin \frac{2\pi}{3} + \cos \frac{2\pi}{3} - i \sin \frac{2\pi}{3} \\ &= 1 + 2 \underbrace{\cos \frac{2\pi}{3}}_{=-\frac{1}{2}} = 1 - 1 = 0. \end{aligned}$$

Let us now act with the inverse time evolution operator:

$$\begin{aligned} \mathcal{U}^\dagger(t, 0) \left(\gamma |0\rangle + \delta |1\rangle + \epsilon |-1\rangle \right) &= \mathcal{U}_\Lambda^\dagger(t, 0) \left(p_e |e\rangle + p_b |b\rangle + p_d |d\rangle \right) \\ &= \left(p_e \cos \mathcal{A} + ip_b \sin \mathcal{A} \right) |e\rangle + \left(p_b \cos \mathcal{A} + ip_e \sin \mathcal{A} \right) |b\rangle + p_d |d\rangle. \end{aligned} \quad (3.14)$$

We then require the coefficient in front of $|e\rangle$ to be zero to assure that equation (3.14) ends up in the bright and dark subspace:

$$p_e \cos \mathcal{A} + ip_b \sin \mathcal{A} = 0 \implies p_e = -ip_b \tan \mathcal{A}. \quad (3.15)$$

Inserting this in the evolved state (equation (3.14)) then yields:

$$\mathcal{U}_\Lambda^\dagger(t, 0) \left(\gamma |0\rangle + \delta |1\rangle + \epsilon |-1\rangle \right) = \frac{p_b}{\cos \mathcal{A}} |b\rangle + p_d |d\rangle. \quad (3.16)$$

We also want the superposition of bright and dark states to be normalized:

$$\begin{aligned} \left| \frac{p_b}{\cos \mathcal{A}} \right|^2 + |p_d|^2 &= \frac{|p_b|^2}{\cos^2 \mathcal{A}} + \frac{2}{3} - |p_b|^2 = |p_b|^2 \left(\frac{1}{\cos^2 \mathcal{A}} - 1 \right) + \frac{2}{3} = \\ &= \frac{\cos^2 \mathcal{A}}{3 \sin^2 \mathcal{A}} \left(\frac{1}{\cos^2 \mathcal{A}} - 1 \right) + \frac{2}{3} = \frac{1}{3} \left(\frac{1 - \cos^2 \mathcal{A}}{\sin^2 \mathcal{A}} + 2 \right) \\ &= \frac{1}{3} \left(\frac{\sin^2 \mathcal{A}}{\sin^2 \mathcal{A}} + 2 \right) = 1, \end{aligned}$$

where in the derivation equations (3.13), (3.15) and $|p_e|^2 = \left| \frac{\delta}{\sqrt{3}} \eta \right|^2 = \frac{1}{3}$ were used. We then see that the superposition of bright and dark states is normalized regardless the choice of \mathcal{A} which is a result of that γ, δ, ϵ are normalized. The final step is then to act with the time evolution operator from left of both sides of equation (3.16) using the fact that the time evolution is unitary (see section 2.2 and 2.4.2):

$$\begin{aligned} \underbrace{\mathcal{U}_\Lambda \left[\mathcal{U}_\Lambda^\dagger \left(\gamma |0\rangle + \delta | +1\rangle + \epsilon | -1\rangle \right) \right]}_{= \mathbb{1}} &= \mathcal{U}_\Lambda(t, 0) \left(\frac{p_b}{\cos \mathcal{A}} |b\rangle + p_d |d\rangle \right) , \\ \implies \mathcal{U}_\Lambda(t, 0) \left(\frac{p_b}{\cos \mathcal{A}} |b\rangle + p_d |d\rangle \right) &= \gamma |0\rangle + \delta | +1\rangle + \epsilon | -1\rangle . \end{aligned}$$

Now, let us actually check if this holds:

$$\begin{aligned} \mathcal{U}_\Lambda(t, 0) \left(\frac{p_b}{\cos \mathcal{A}} |b\rangle + p_d |d\rangle \right) &= \\ &= \left[|d\rangle\langle d| + \cos \mathcal{A} \left(|e\rangle\langle e| + |b\rangle\langle b| \right) - i \sin \mathcal{A} \left(|e\rangle\langle b| + |b\rangle\langle e| \right) \right] \\ &\times \left(\frac{p_b}{\cos \mathcal{A}} |b\rangle + p_d |d\rangle \right) = p_d |d\rangle + p_b |b\rangle - \underbrace{ip_b \tan \mathcal{A}}_{-p_e \text{ Eq. (3.15)}} |e\rangle . \end{aligned}$$

Using equation (3.15) we obtain:

$$\begin{aligned} \mathcal{U}_\Lambda(t, 0) \left(\frac{p_b}{\cos \mathcal{A}} |b\rangle + p_d |d\rangle \right) &= p_d |d\rangle + p_b |b\rangle + p_e |e\rangle \\ &= \gamma |0\rangle + \delta | +1\rangle + \epsilon | -1\rangle , \end{aligned} \tag{3.17}$$

where in the last line we used equation (3.11). Note here that for $\mathcal{A} = \pi/2$; $\tan \mathcal{A} = 1/0$, but this is fine if we (using equation (3.15)) impose that $p_b = 0$ for $\mathcal{A} = \pi/2$ such that:

$$\lim_{\substack{p_b \rightarrow 0 \\ \mathcal{A} \rightarrow \frac{\pi}{2}}} \frac{p_b}{\tan \mathcal{A}} = \text{Well-defined} \neq 0 .$$

This means that the coefficient $p_b / \cos \mathcal{A}$ in equation (3.16) becomes well-defined, still leaving us in a superposition of the bright and dark states. We can continue to see what restrictions $p_b = 0$ imposes on ω_0 and ω_1 , but remember that for a $\pi/2$ -pulse, the bright state is already driven up into the excited state (see section 2.4.2) which is further than the time evolution of interest since we are interested in finding a chiral state by freezing the time evolved system in between the oscillation of the

bright and the excited states. For the scope of this investigation this is therefore unnecessary.

We then see that the scheme assures that the starting state $\alpha |b\rangle + \beta |d\rangle$ time evolves into a general superposition of the chiral states, $\gamma |0\rangle + \delta |+1\rangle + \epsilon |-1\rangle$, for all parameters such that:

$$\left\{ \begin{array}{l} \alpha = \frac{p_b}{\cos \mathcal{A}} = \frac{1}{\sqrt{3} \cos \mathcal{A}} \left[(\gamma + \delta + \epsilon) \omega_0 + (\gamma + \eta^2 \delta + \eta \epsilon) \omega_1 \right] , \\ \beta = p_d = \frac{1}{\sqrt{3}} \left[(\gamma + \eta^2 \delta + \eta \epsilon) \omega_0^* - (\gamma + \delta + \epsilon) \omega_1^* \right] , \\ \omega_0 = J_{12} + i D_{12} , \\ \omega_1 = J_{23} - i D_{23} , \\ 1 = |\gamma|^2 + |\delta|^2 + |\epsilon|^2 , \\ \tan \mathcal{A} = i \frac{p_e}{p_b} = i \frac{\gamma + \eta \delta + \eta^2 \epsilon}{(\gamma + \delta + \epsilon) \omega_0 + (\gamma + \eta^2 \delta + \eta \epsilon) \omega_1} , \quad \mathcal{A} \in \left(-\frac{\pi}{2}, \frac{\pi}{2} \right) . \end{array} \right. \quad (3.18)$$

The following subsections will be devoted to finding solutions for different combinations of γ , δ , ϵ .

3.3.5 Time Evolution Into One of the Purely Chiral States

We are interested in the evolution to a purely chiral state, say $|+1\rangle$, so we set $\gamma = \epsilon = 0 \implies \delta = e^{i\varphi}$:

$$\left\{ \begin{array}{l} p_e = \frac{\delta}{\sqrt{3}} \eta , \\ p_b = \frac{\delta}{\sqrt{3}} (\omega_0 + \eta^2 \omega_1) , \\ p_d = \frac{\delta}{\sqrt{3}} (\eta^2 \omega_0^* - \omega_1^*) . \end{array} \right. \quad (3.19)$$

For this choice of parameters, equation (3.17) becomes:

$$\mathcal{U}_\Lambda(t, 0) \frac{\delta}{\sqrt{3}} \left[\frac{\omega_0 + \eta^2 \omega_1}{\cos \mathcal{A}} |b\rangle + (\eta^2 \omega_0^* - \omega_1^*) |d\rangle \right] = \delta |+1\rangle .$$

This shows that $\delta = e^{i\phi}$ becomes a global phase and can then be set to $\delta = 1$ since global phases do not change the physics. Thus,

$$\mathcal{U}_\Lambda(t, 0) \frac{1}{\sqrt{3}} \left[\frac{\omega_0 + \eta^2 \omega_1}{\cos \mathcal{A}} |b\rangle + (\eta^2 \omega_0^* - \omega_1^*) |d\rangle \right] = |+1\rangle . \quad (3.20)$$

3.3.6 Restrictions for Ending Up in a Purely Chiral State

The restrictions for ending up in the purely chiral state $|+1\rangle$ is given by equation (3.18) with p_e , p_b , p_d given by equation (3.19):

$$\frac{i\eta}{\omega_0 + \eta^2\omega_1} = \tan \mathcal{A} .$$

We can rewrite $i\eta$ as $e^{i\pi/2}e^{i2\pi/3} = e^{i7\pi/6}$, and use the definition of $\omega_0 = \cos \frac{\theta}{2}$, $\omega_1 = -\sin \frac{\theta}{2}e^{i\phi}$:

$$\frac{e^{i\frac{7\pi}{6}}}{\cos \frac{\theta}{2} - \sin \frac{\theta}{2}e^{i\left(\frac{4\pi}{3}+\phi\right)}} = \tan \mathcal{A} .$$

Since $\tan \mathcal{A}$ is purely real, that means the numerator is required to have the same complex phase as the denominator, only then can the left hand side be real:

$$\frac{e^{i\frac{7\pi}{6}}}{\cos \frac{\theta}{2} - \sin \frac{\theta}{2}e^{i\left(\frac{4\pi}{3}+\phi\right)}} \stackrel{!}{=} \frac{\cancel{\pm}e^{i\frac{7\pi}{6}}}{\left| \cos \frac{\theta}{2} - \sin \frac{\theta}{2}e^{i\left(\frac{4\pi}{3}+\phi\right)} \right| e^{i\frac{7\pi}{6}}},$$

where the \pm comes from that $\tan \mathcal{A}$ is allowed to be negative (remembering that $\mathcal{A} \in (-\pi/2, \pi/2)$) meaning that the phase of the denominator is either $7\pi/6$ (+) or $\pi/6$ (-). This imposes the relations:

$$\begin{cases} \frac{\pm 1}{\left| \cos \frac{\theta}{2} - \sin \frac{\theta}{2}e^{i\left(\frac{4\pi}{3}+\phi\right)} \right|} = \tan \mathcal{A} , \\ \arg \left\{ \cos \frac{\theta}{2} - \sin \frac{\theta}{2}e^{i\left(\frac{4\pi}{3}+\phi\right)} \right\} = \frac{7\pi}{6} + k\pi, \quad k \in \mathbb{Z} . \end{cases}$$

So let us compute what restrictions these equations imply:

$$\begin{aligned} \cos \frac{\theta}{2} - \sin \frac{\theta}{2}e^{i\left(\frac{4\pi}{3}+\phi\right)} &= \cos \frac{\theta}{2} - \sin \frac{\theta}{2} \cos \left(\frac{4\pi}{3} + \phi \right) - i \sin \frac{\theta}{2} \sin \left(\frac{4\pi}{3} + \phi \right) , \\ \arg(z) &\stackrel{\text{def}}{=} \arctan \left(\frac{\text{Im}\{z\}}{\text{Re}\{z\}} \right) , \end{aligned}$$

$$\begin{aligned}
\Rightarrow \arg \left\{ \cos \frac{\theta}{2} - \sin \frac{\theta}{2} e^{i(\frac{4\pi}{3} + \phi)} \right\} &= \arctan \left(\frac{-\sin \frac{\theta}{2} \sin \left(\frac{4\pi}{3} + \phi \right)}{\cos \frac{\theta}{2} - \sin \frac{\theta}{2} \cos \left(\frac{4\pi}{3} + \phi \right)} \right) = \\
&= \arctan \left(\frac{\sin \left(\frac{4\pi}{3} + \phi \right) \tan \frac{\theta}{2}}{\cos \left(\frac{4\pi}{3} + \phi \right) \tan \frac{\theta}{2} - 1} \right) = \begin{cases} \frac{7\pi}{6} & \text{if } \tan \mathcal{A} > 0, \\ \frac{\pi}{6} & \text{if } \tan \mathcal{A} < 0. \end{cases}
\end{aligned}$$

Taking the tangent function of both sides of the equation and using its π -periodicity¹⁰ we get:

$$\begin{aligned}
\frac{\sin \left(\frac{4\pi}{3} + \phi \right) \tan \frac{\theta}{2}}{\cos \left(\frac{4\pi}{3} + \phi \right) \tan \frac{\theta}{2} - 1} &= \begin{cases} \tan \frac{7\pi}{6} = \frac{1}{\sqrt{3}} & \text{if } \tan \mathcal{A} > 0, \\ \tan \frac{\pi}{6} = \frac{1}{\sqrt{3}} & \text{if } \tan \mathcal{A} < 0, \end{cases} \\
\Rightarrow \tan \frac{\theta}{2} &= \frac{-1/\sqrt{3}}{\sin \left(\frac{4\pi}{3} + \phi \right) - \frac{1}{\sqrt{3}} \cos \left(\frac{4\pi}{3} + \phi \right)} \\
&= \frac{-1}{\sqrt{3} \left(\sin \frac{4\pi}{3} \cos \phi + \cos \frac{4\pi}{3} \sin \phi \right) - \left(\cos \frac{4\pi}{3} \cos \phi - \sin \frac{4\pi}{3} \sin \phi \right)} \\
&= \frac{1/2}{\frac{1}{2} \cos \phi + \frac{\sqrt{3}}{2} \sin \phi} = \frac{1/2}{\sin \frac{\pi}{6} \cos \phi + \cos \frac{\pi}{6} \sin \phi}, \\
\Rightarrow \tan \frac{\theta}{2} &= \frac{1}{2 \sin \left(\frac{\pi}{6} + \phi \right)}, \quad \mathcal{A} \in \left(-\frac{\pi}{2}, \frac{\pi}{2} \right).
\end{aligned}$$

Since θ and ϕ parameterize ω_0 and ω_1 it becomes evident that there is a solution θ for each chosen ϕ , meaning that we can obtain any coefficients in the JD-Hamiltonian (3.6) as long as θ depend on ϕ in accordance with the above relation. The second condition can be computed as:

$$\left| \cos \frac{\theta}{2} - \sin \frac{\theta}{2} e^{i(\frac{4\pi}{3} + \phi)} \right| = \sqrt{1 - \sin \theta \cos \left(\frac{4\pi}{3} + \phi \right)}.$$

Finally we have:

$$\begin{cases} \tan \frac{\theta}{2} = \frac{1}{2 \sin \left(\frac{\pi}{6} + \phi \right)}, \\ \tan \mathcal{A} = \frac{\pm 1}{\sqrt{1 - \sin \theta \cos \left(\frac{4\pi}{3} + \phi \right)}}, \\ \mathcal{A} \in \left(-\frac{\pi}{2}, \frac{\pi}{2} \right), \end{cases} \quad (3.21)$$

¹⁰i.e: $\tan(x + k\pi) = \tan(x)$ for $k \in \mathbb{Z}$.

where the first equation gives θ for a choice of ϕ , and the second equation gives us the pulse area \mathcal{A} for that particular combination of θ and ϕ . For any choice of \mathcal{A} , θ , ϕ that satisfies these relations, the result in equation (3.20) holds which concludes the hunt for parameters of the time evolution operator such that a superposition of bright and dark states evolve to a purely chiral state after a time exposure determined by \mathcal{A} . Note that by imposing relations (3.21), the normalization of both the starting state and the final state is assured and follows from the normalizations shown in section 3.3.4.

3.3.7 Time Evolution Into a Superposition of Nonzero Purely Chiral States

It might also be interesting to create superpositions of purely chiral states. This case would, for a superposition of the nonzero chiral states $|\pm 1\rangle$, correspond to $\gamma = 0$, $\delta, \epsilon \neq 0$ in equation (3.17). The coefficients p_e , p_b and p_d would then look like:

$$|\gamma|^2 + |\delta|^2 + |\epsilon|^2 = 1 \implies \epsilon = \sqrt{1 - |\delta|^2} e^{i\psi} ,$$

$$\begin{cases} p_e &= \frac{1}{\sqrt{3}} (\eta\delta + \eta^2\epsilon) , \\ p_b &= \frac{1}{\sqrt{3}} ([\delta + \epsilon]\omega_0 + [\eta^2\delta + \eta\epsilon]\omega_1) , \\ p_d &= \frac{1}{\sqrt{3}} ([\eta^2\delta + \eta\epsilon]\omega_0^* - [\delta + \epsilon]\omega_1^*) . \end{cases}$$

Plugging in the coefficients p_e and p_b into equation (3.17), we get:

$$\begin{aligned} \mathcal{U}_\Lambda(t, 0) \frac{1}{\sqrt{3}} \left[\frac{(\delta + \epsilon)\omega_0 + (\eta^2\delta + \eta\epsilon)\omega_1}{\cos \mathcal{A}} |b\rangle + ([\eta^2\delta + \eta\epsilon]\omega_0^* - [\delta + \epsilon]\omega_1^*) |d\rangle \right] = \\ = \delta | +1 \rangle + \epsilon | -1 \rangle . \end{aligned} \quad (3.22)$$

This time, the restrictions imposed by equation (3.18) look like:

$$\tan \mathcal{A} = i \frac{\eta\delta + \eta^2\epsilon}{(\delta + \epsilon)\omega_0 + (\eta^2\delta + \eta\epsilon)\omega_1} = i \frac{\delta + \eta\epsilon}{\eta^2(\delta + \epsilon)\omega_0 + (\eta\delta + \epsilon)\omega_1} , \quad (3.23)$$

where $\eta^{-1} = e^{-i2\pi/3} = e^{i4\pi/3} = \eta^2$ has been used. The method is to break up the numerator and denominator in their real and imaginary parts, and realize that $\tan \mathcal{A}$ is real, see section 3.3.6 for using this method to find the restrictions in the case of one purely chiral state. Any \mathcal{A} , ω_0 , ω_1 that fulfill the two relations given by the expression (3.23) assures that equation (3.22) holds.

3.3.8 Speculation of Creating a Chiral Gate

We saw in section 3.3.5 that it was possible to start in a superposition of the bright and dark states and time evolve the system into a purely chiral state. Later we saw in section 3.3.7 that we could start in a superposition of the bright and dark states and time evolve the system into a superposition of the purely chiral states. This raises the question if we could, by unitary operations, evolve a purely chiral state into a superposition of purely chiral states, i.e. creating a chiral quantum gate. Reminding the reader that the time evolution operator (that depends on the system Hamiltonian), is dependent on the parameters \mathcal{A} , ω_0 , ω_1 (and the bright and dark states also, by definition, depend on ω_0 , ω_1). From now on, we will distinguish the different time evolution operators from the previous sections by including what explicit parameters they depend on. To realize a gate, we could use:

$$\begin{aligned}
 \tilde{\mathcal{U}}^\dagger | +1 \rangle &= \frac{\tilde{p}_b}{\cos \tilde{\mathcal{A}}} \tilde{|b\rangle} + \tilde{p}_d \tilde{|d\rangle} \\
 \mathcal{U}_p^p \left(\frac{\tilde{p}_b}{\cos \tilde{\mathcal{A}}} \tilde{|b\rangle} + \tilde{p}_d \tilde{|d\rangle} \right) &= \frac{p_b}{\cos \mathcal{A}} |b\rangle + p_d |d\rangle , \\
 \mathcal{U} \left(\frac{p_b}{\cos \mathcal{A}} |b\rangle + p_d |d\rangle \right) &= \delta | +1 \rangle + \epsilon | -1 \rangle , \\
 \implies \underbrace{\mathcal{U} \mathcal{U}_p^p \tilde{\mathcal{U}}^\dagger}_{\equiv \mathcal{O}} | +1 \rangle &= \delta | +1 \rangle + \epsilon | -1 \rangle ,
 \end{aligned}$$

which, if it is possible to do, would create a superposition of the purely chiral states by starting in the purely chiral state $| +1 \rangle$.

4 Conclusions

In this thesis we saw that quantum chirality is described through chiral states and operators which obey a spin-1 algebra and has probability currents with the same sign as the chirality. This is one explanation why quantum chirality is called precisely chiral, since the probability currents suggest a handedness, and the answer to aim one. One should, however, generalize the idea of describing quantum chirality through its states' probability currents before it is possible to draw any conclusions. For example, it would be interesting to see if the description is consistent with higher odd numbers of spin, since Trif et al. [27, 28] showed that there exist chiral operators and states for a closed spin chain of five spin particles, or what happens to the probability current of an even number of spins, which Trif et al. claimed do not give rise to chiral states. It is also interesting to investigate how the probability current depends on not only the chiral states but also the chiral operators. Maybe there is an operator description of the probability current or some other connection that implies how the probability current handedness follows from the chiral operators themselves.

In sections 3.3.5 through 3.3.7 we saw that it is possible to prepare the open spin chain in a superposition of the bright and dark states and by time evolution force the system into the purely chiral states or in superpositions thereof, all made possible due to the XY and DM interactions that naturally occur between the spins. There were restrictions to these different cases so that the parameters could not be chosen arbitrarily, but still had solutions such that one or more of the parameters acted as a free parameter. This answers the second aim and verifies that quantum chirality can occur even in systems that are not in chiral states naturally, and the scheme to do so is realized through time evolution in a Λ -system configuration.

Further future work of interest could be to see if it is possible to create quantum chirality from open spin chains using merely Heisenberg interactions to confine the theory even more. It seems as though this is not possible using the Λ -based time evolution as was done in this scheme, since we saw that the solutions rely on that the laser parameters are complex valued, with the complex part given by the DM coupling constants, see equation (3.7). Regardless, it might be possible to realize chiral states by only considering Heisenberg interactions using some other scheme.

It should be addressed that the scheme to create chiral states in this thesis is presently merely an early stage speculation that waits for its physical implementation. There are a rich amount of problems that could occur when implementing the scheme in the laboratory. For example, it seems to be quite difficult to switch on and off spin interactions through lasers, which should be mentioned has been done in so called Paul traps (ion traps) [2, 7, 22, 30], and this is what the time evolution of the scheme depends on¹¹. The open spin chain could potentially be driven using a Paul trap whereas the spin triangle that Trif et al. [27, 28] considered is a molecular nanomagnet which allows for different kinds of experiments, and the good control of the system they presented might not be as easily translated to the open spin chain. The scheme does not address the robustness either, meaning how sensitive the chiral states are to decaying after being created, which is a problem that might occur. Innumeros of other complications with laser-tuning and the experimental setup is most likely to occur, in accordance with the history of physics experiments.

To propose some applications, Haroche comes to mind, where he showed that microwaves in a cavity can be used to create superposition of states and study entanglement and decoherence [8, 17]. In the future it will therefore maybe be possible to use chiral states for this, to prepare the open spin chain in a state living in the subspace of the bright and dark states, send it into a "cavity"¹² and measure the system to end up in a superposition of chiral states in accordance with the scheme developed in this thesis. If this would work then maybe chirality can be used to study entanglement and decoherence even further.

A second implementation of this thesis could be chiral gates, a vague scheme of this being described in section 3.3.8. Managing to create a chiral gate would allow us to study chirality as an information carrier in a quantum gate which might increase our knowledge about quantum information further. If we also incorporate what Trif et al. [27, 28] showed for closed spin chains, that transitions between spin and chirality can be controlled independently through magnetic and electric fields, we might then discover the potential existence of quantum bits that carry

¹¹Using Paul traps for the time evolution would look something like this (if possible): trap the spin particles that has latent spin interactions between them, and use laser pulses to activate the spin interactions during a time goverened by the pulse area. This would then drive the system, in accordance with the scheme developed in this thesis, for specific choices of the parameters.

¹²It should be noted that this is merely a speculation. The word cavity is here used as the "machine" used to create chiral superpositions. Haroche et al. showed that using cavities works to create superpositions for internal energy level transitions of circular Rydberg states [17] which assures nothing in the case of chirality. The speculation is about an application of a similar creator of superpositions for chiral states, which might or might not be possible using a cavity, and is perhaps not possible to construct at all.

both spin and chirality as information in a quantum circuit since the chirality then becomes an additional internal degree of freedom. This could possibly expand our knowledge and extent of quantum computers. Note again, however, that the good chiral control that was shown was on molecular nanomagnets whereas in this thesis we have looked at open spin chains which have different properties.

5 Ethical Aspects

The greatest societal impact of the eventual implementation of this thesis would probably be quantum computers if chirality could be used as an additional internal degree of freedom (but that is a big if). The theory of quantum computers is great for increasing computational power, advancing components and precision as well as improving the security since quantum keys are destroyed when interacted with [18, 19]. But with great power comes great responsibility; a problem with quantum computers is that they simplify hacking and violating personal integrity¹³, which puts the user in a powerful position while it puts governments and legislation in a tricky one.

A second aspect of how society can be affected by this thesis and theoretical work in general is the classic problem of why we should put resources into research with no direct applications (with other words, theoretical science). To this question there are multiple answers and variants of interpretation, none of which can be considered incorrect, but rather a subjective opinion. One point of view is that theoretical research contributes to distributing new knowledge to the society which enriches it (and whether this knowledge is valued equally to the amount of money and time put into the research I will leave for the reader to form an opinion about themselves). A second point of view is marketing, since any work can be used to distribute knowledge to, feed the curiosity of and inspire the reader, which also is of great value. A final point of view is that theoretical research can firstly "accidentally" find answers to questions or problems that previously was not believed to be connected (in this thesis: chiral gates, development of a greater unifying theory of chirality and maybe quantum chirality in medicine?), and secondly, areas of use can present themselves with time which reveals that the previous work was not in vain (in this thesis: quantum chirality as a potential additional degree of freedom

¹³The reader might rightfully find this comment peculiar since I just stated that quantum computers can be used to improve security and now speak of how they can violate it. Both statements, however, I claim to be true but the catch is this: quantum computers are more secure but it is easier to hack classical computers with a quantum computer, which makes cybersecurity an economical question. If you can afford a quantum computer you have bought yourself an advantage, both in defense and attack.

and development of Paul traps). Since this thesis is, indirectly, a study of XY and DM interactions and also of mathematical quantum mechanical schemes, maybe this work can be connected to these broader areas in the future.

References

- [1] BEN DOR, O., YOCHELIS, S., RADKO, A., VANKAYALA, K., CAPUA, E., CAPUA, A., YANG, S., BACZEWSKI, L. T., PARKIN, S. S. P., NAAMAN, R., and PALTIEL, Y. Magnetization switching in ferromagnets by adsorbed chiral molecules without current or external magnetic field. *Nat. Commun.* **8**, 14567 (2017).
- [2] CHANG, M., CHANG, Y., KIM, N.-Y., KIM, H., LEE, S.-H., CHOI, M.-S., KIM, Y.-H., and KAHNG, S.-J. Tuning and sensing spin interactions in Co-porphyrin/Au with NH₃ and NO₂ binding. *Phys. Rev. B* **100**, 245406 (2019).
- [3] DALLEY, S. and MCCARTOR, G. Spontaneously broken quark helicity symmetry. *Ann. Phys. (N.Y.)* **321**, 402 (2006).
- [4] DAS, T. K., TASSINARI, F., NAAMAN, R., and FRANSSON, J. Temperature-Dependent Chiral-Induced Spin Selectivity Effect: Experiments and Theory. *J. Phys. Chem. C* **126**, 3257 (2022).
- [5] DZYALOSHINSKY, I. A thermodynamic theory of “weak” ferromagnetism of antiferromagnetics. *J Phys Chem Solids* **4**, 241 (1958).
- [6] FRANSSON, J. Charge Redistribution and Spin Polarization Driven by Correlation Induced Electron Exchange in Chiral Molecules. *Nano Lett.* **21**, 3026 (2021).
- [7] GILSON, E. P., DAVIDSON, R. C., EFTHIMION, P. C., MAJESKI, R., and QIN, H. The Paul Trap Simulator Experiment. *Laser Part. Beams* **21**, 549 (2003).
- [8] HAROCHE, S. Controlling photons in a box and exploring the quantum to classical boundary. *Ann. Phys. (Berl.)* **525**, 753 (2013).
- [9] HARRIS, M. *Left-Right Symmetric Model*. MA thesis. DiVA-Portal: Uppsala University, 2017 (1).
- [10] HELMENSTINE, A. Amino Acid Chirality. Retrieved from <https://www.thoughtco.com/amino-acid-chirality-4009939> (2019).
- [11] HERTERICH, E. and SJÖQVIST, E. Single-loop multiple-pulse nonadiabatic holonomic quantum gates. *Phys. Rev. A* **94**, 052310 (2016).
- [12] LIU, X. *Organic Chemistry 1*. (Kwantlen Polytechnic University, 2021).
- [13] MORIYA, T. Anisotropic Superexchange Interaction and Weak Ferromagnetism. *Phys. Rev.* **120**, 91 (1960).

-
- [14] NGUYEN, L. A., HE, H., and PHAM-HUY, C. Chiral Drugs: An Overview. Retrieved from <https://www.ncbi.nlm.nih.gov/pmc/articles/PMC3614593/> (2006).
- [15] NIELSEN, M. A. and CHUANG, I. L. *Quantum computation and quantum information*. 10th anniversary ed. (Cambridge University Press, 2010).
- [16] OHSHIMA, T., EKERT, A., OI, D. K. L., KASLIZOWSKI, D., and KWEK, L. C. *Robust state transfer and rotation through a spin chain via dark passage*. Preprint at [arXiv:quant-ph/0702019](https://arxiv.org/abs/quant-ph/0702019) [quant-ph] (2008).
- [17] RAIMOND, J. M., BRUNE, M., and HAROCHE, S. Manipulating quantum entanglement with atoms and photons in a cavity. *Rev. Mod. Phys.* **73**, 565 (2001).
- [18] RATHNAYAKE, D. The impact of Quantum Computing on cybersecurity. Retrieved from <https://www.tripwire.com/state-of-security/impact-quantum-computing-cybersecurity> (2023).
- [19] RJAIBI, W., MUPPIDI, S., and O'BRIEN, M. Quantum computing and cybersecurity: How to capitalize on opportunities and sidestep risks. Retrieved from <https://www.ibm.com/thought-leadership/institute-business-value/en-us/report/quantumsecurity> (2018).
- [20] SAKURAI, J. J. *Modern Quantum Mechanics*. Third edition. (Cambridge University Press, 2021).
- [21] SAYINER, S. The Importance of Chirality in Biological Systems. Retrieved from <https://biyokimya.vet/en-gb/the-importance-of-chirality-in-biological-systems/> (2022).
- [22] SEMERIKOV, I. A., ZALIVAKO, I. V., BORISENKO, A. S., AKSENOV, M. D., KOLACHEVSKY, N. N., and KHABAROVA, K. Y. Linear Paul Trap for Quantum Logic Experiments. *Lebedev Phys. Inst.* **47**, 385 (2020).
- [23] SJÖQVIST, E., TONG, D. M., ANDERSSON, L. M., HESSMO, B., JOHANSSON, M., and SINGH, K. Non-adiabatic holonomic quantum computation. *New J. Phys.* **14**, 103035 (2012).
- [24] SQUIRES, G. The discovery of the structure of DNA. *Contemp Phys.* **44**, 289 (2003).
- [25] SUKENIK, N., TASSINARI, F., YOCHELIS, S., MILLO, O., BACZEWSKI, L. T., and PALTIEL, Y. Correlation between Ferromagnetic Layer Easy Axis and the Tilt Angle of Self Assembled Chiral Molecules. *Molecules* **25**, (2020).

- [26] TANEDO, F. Helicity, Chirality, Mass, and the Higgs. Retrieved from <https://www.quantumdiaries.org/2011/06/19/helicity-chirality-mass-and-the-higgs/> (2011).
- [27] TRIF, M., TROIANI, F., STEPANENKO, D., and LOSS, D. Spin-Electric Coupling in Molecular Magnets. *Phys. Rev. Lett.* **101**, 217201 (2008).
- [28] TRIF, M., TROIANI, F., STEPANENKO, D., and LOSS, D. Spin electric effects in molecular antiferromagnets. *Phys. Rev. B* **82**, 045429 (2010).
- [29] WALDECK, D. H., NAAMAN, R., and PALTIEL, Y. The spin selectivity effect in chiral materials. *APL Mater.* **9**, 040902 (2021).
- [30] WANG, J.-L., LIU, Q., MENG, Y.-S., ZHENG, H., ZHU, H.-L., SHI, Q., and LIU, T. Synergic on/off Photoswitching Spin State and Magnetic Coupling between Spin Crossover Centers. *Inorg. Chem.* **56**, 10674–10680 (2017).
- [31] XU, G. F., ZHANG, J., TONG, D. M., SJÖQVIST, E., and KWEK, L. C. Nonadiabatic Holonomic Quantum Computation in Decoherence-Free Subspaces. *Phys. Rev. Lett.* **109**, 170501 (2012).

Appendix A Alternative Form of the Chiral Operators

Computing the dot and vector products from the definition of the chiral operators (2.4) and the ladder operators (2.7), yields their more explicit form:

$$C_x = -\frac{\hbar^2}{6} \sum_{j=1}^3 \left(\sigma_j \otimes \sigma_j \otimes \mathbb{1} - 2\mathbb{1} \otimes \sigma_j \otimes \sigma_j + \sigma_j \otimes \mathbb{1} \otimes \sigma_j \right),$$

$$C_y = -\frac{\hbar^2}{2\sqrt{3}} \sum_{j=1}^3 \left(\sigma_j \otimes \sigma_j \otimes \mathbb{1} - \sigma_j \otimes \mathbb{1} \otimes \sigma_j \right),$$

$$C_z = \frac{\hbar^3}{2\sqrt{3}} \sum_{k=1}^3 \sum_{l=1}^3 \sum_{m=1}^3 \epsilon_{klm} \sigma_k \otimes \sigma_l \otimes \sigma_m,$$

$$C_+ = -\frac{\hbar^2}{6} \sum_{j=1}^3 \left[\left(1 - i\sqrt{3} \right) \sigma_j \otimes \sigma_j \otimes \mathbb{1} - 2\mathbb{1} \otimes \sigma_j \otimes \sigma_j + \left(1 + i\sqrt{3} \right) \sigma_j \otimes \mathbb{1} \otimes \sigma_j \right],$$

$$C_- = -\frac{\hbar^2}{6} \sum_{j=1}^3 \left[\left(1 + i\sqrt{3} \right) \sigma_j \otimes \sigma_j \otimes \mathbb{1} - 2\mathbb{1} \otimes \sigma_j \otimes \sigma_j + \left(1 - i\sqrt{3} \right) \sigma_j \otimes \mathbb{1} \otimes \sigma_j \right],$$