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Creating Rydberg crystals in ultra-cold gases using stimulated Raman adiabatic passage schemes

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Award date:
2012

Link to publication
Eindhoven University of Technology
Department of Applied Physics
Coherence and Quantum Technology group
CQT 2012-08

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July 2012

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Abstract

This report is the result of a bachelor internship of two applied physics students. In it, we study Rydberg physics. The goal is to create a strongly-coupled Rydberg crystal by exciting atoms in an ultra-cold atomic gas from their ground state to the Rydberg state. Due to the van der Waals blockade effect, the energy required to excite an atom to the Rydberg state is a function of the number of neighbouring Rydberg atoms. We therefore propose to use an excitation scheme called STIRAP. Such a scheme ensures a full population inversion for a single atom in a three-level system, by using adiabatic following. For our multi-particle system, we detune the lasers for every STIRAP pulse to account for an increasing number of nearest neighbours. This way, we aim to excite a full lattice with hexagonal symmetry after seven STIRAP pulses. After a general introduction, we do a theoretical investigation to see whether this scheme can work. We do this by looking for suitable eigenvectors of the Hamiltonian, which we can then follow adiabatically. It is concluded that for a system with no spontaneous emission from the intermediate state, the scheme can work perfectly. However, if we proceed to include spontaneous emission from the intermediate state by running Quantum Monte Carlo (QMC) simulations, the scheme fails for realistic experimental parameters. However, using laser intensities we cannot achieve in practice, it is possible to obtain a Rydberg crystal in theory.
Preface

This report is the product of a bachelor internship of 10 weeks within the capacity group Coherence and Quantum Technology (CQT) at the Applied Physics Department of Eindhoven University of Technology.

In the beginning of this year we did an internship with one of the few theorists of our faculty: prof. dr. ir. Paul van der Schoot, from the group Theory of Polymers and Soft matter (TPS). For this project we created a one-dimensional model for the distribution of single-stranded RNA in an icosahedral virus capsid. Before the beginning of this project we were applied physics students who were interested in theory, after this project we were theorist who just so happen to study applied physics.

For our minor we followed a course in elementary quantum optics from one of the other few theorists of our faculty: dr. ir. Servaas Kokkelmans. Having enjoyed his course and the theoretical project with Paul, we turned our hopes to him for a theoretical bachelor project. Although he normally does not supervise bachelor internships, he was willing to make an exception, for which we are very grateful. We certainly hope we have lived up to his expectations.

During our internship, Rick van Bijnen was our daily supervisor. Especially in the beginning, he was always there for all our questions and problems, which we really appreciate. He turned us from computer-fearing nerds into full-blown hackers in just 10 weeks! His legendary intuitive explanations were always much appreciated, like for instance: "In quantum mechanics, if things go up, they go down at the same pace." He also taught us quite some new theoretical quantum mechanics, which shall come in handy when we start our master in theoretical physics in Utrecht. We hope Rick has a great and productive time in Dresden and we wish him the best of luck with completing his PhD thesis next year.

Furthermore, we would like to thank Edgar Vredenbregt for his suggestions during the weekly meetings. He was not planned to be one of our supervisors orginally, but he most certainly become one.

Finally we want to thank all master students working on their experimental graduation project in the student zoo of CQT. Because of their extreme loudness, non-productivity and incredibly small knowledge of mathematics, we now know for sure that we never want to become experimental physicists and continue our career in the much more quiet area of theoretical physics.
Disclaimer: the physics in this report has been done under the influence of liters of coffee and would not have been possible without it.
## Contents

1 Introduction ................................................................................. 5
   1.1 Laser cooling ........................................................................ 5
   1.2 Rydberg atoms .................................................................... 7
   1.3 The van der Waals interaction .............................................. 9
   1.4 Rydberg crystals ................................................................. 11
   1.5 Rydberg research: why bother? ............................................. 13
   1.6 Summary and outline ........................................................... 14

2 STIRAP ......................................................................................... 15
   2.1 Rabi oscillations .................................................................. 15
   2.2 The STIRAP procedure ....................................................... 17
   2.3 Experimental setup and restrictions ...................................... 21
   2.4 Adiabaticity criterion ......................................................... 23
   2.5 Summary ............................................................................. 26

3 Multi-particle STIRAP without spontaneous emission ................. 27
   3.1 The N-particle Hamiltonian in a three-level system .............. 27
   3.2 Two-level system with three particles on a line ................. 28
   3.3 Perturbation approach ....................................................... 30
      3.3.1 Zeroth-order perturbation ............................................. 30
      3.3.2 First-order perturbation .............................................. 30
      3.3.3 Second-order perturbation .......................................... 32
   3.4 Eigenvectors of the three-particle Hamiltonian ................. 34
      3.4.1 Adiabaticity criterion .................................................. 41
   3.5 Eigenvectors for other systems ........................................... 42
      3.5.1 Two particles .............................................................. 42
      3.5.2 Four particles on a line .............................................. 43
      3.5.3 Three particles in a triangle configuration .................. 44
      3.5.4 General systems ....................................................... 45
   3.6 Summary ............................................................................. 47

4 Multi-particle STIRAP with spontaneous emission ..................... 48
   4.1 Rate of spontaneous emission ............................................ 48
   4.2 Review of density operator theory ..................................... 50
   4.3 Using density operator theory to describe spontaneous emission ........................................ 52
      4.3.1 Example: a dissipative two-level system .................. 53
   4.4 The quantum Monte Carlo approach .................................. 55
Chapter 1

Introduction

In this chapter we give both an experimental and a theoretical basis for the rest of this report about the formation of Rydberg crystals. We start by discussing the experimental techniques for obtaining an ultra-cold atomic gas, which is essential for creating a Rydberg crystal. We proceed by describing the properties of Rydberg atoms and how these properties can be used to form so-called Rydberg crystals. After explaining the relevance of Rydberg research, we present a general outline for the report.

1.1 Laser cooling

In 1960 the first functioning LASER (Light Amplification by Stimulated Emission of Radiation) was operated. It was the product of a long and fierce race between the Bell Labs led by Charles Townes - who had invented the MASER (Microwave Amplification by Stimulated Emission of Radiation) a few years beforehand - and the Technical Research Group (TRG) led by Gordon Gould. Eventually, the latter won the race by an inch. Paradoxically, the former received a Nobel Prize for his research into the laser [1], contrary to the latter.

After his defeat, Townes supposedly sighed: "Lasers are a solution looking for a problem," [2]. This was a misjudgement to say the least. At the moment the laser is both used in everyday life - for instance for scanning barcodes or playing a CD - and in the fundamental physical sciences. Almost all optical experiments use the laser as a light source, with as a main reason the fact that it is a coherent light source. These kinds of experiments were already performed before the rise of the laser. They only became a lot easier by the usage of the laser.

There are also types of experiments which are only possible because of the highly coherent and monochromatic character of the laser. In the early 1970s, the first proposals for laser cooling were published. A general definition of laser cooling is "the reduction of random thermal velocities in an atomic gas, by usage of electromagnetic radiative forces" [3]. In 1978 A. Ashkin et al. cooled an atomic beam of sodium down to $\sim \mu$K by using the radiation pressure of a laser beam tuned to an atomic resonance. Since then, an explosion of interest in laser cooling occurred. It is a highly active field of research and of grave importance. This accumulated in a Nobel Prize "for development of methods to cool and trap atoms with laser light" in 1997 for Steven Chu, Claude
Cohen-Tannoudji and William Daniel Phillips. Besides that, the technique was used to create the first Bose-Einstein Condensate in an ultra-cold gas in 1995, for which the Nobel Prize was also awarded a few years later.

So, how does laser cooling work? The main idea is to slow an atomic beam down by using the transfer of momentum that occurs when an atom absorbs a photon. This slowing down caused by the photon is also called the scattering force. How this fictive force emerges, is displayed in figure 1.1. An atomic beam with certain velocity \( v \) is irradiated by a laser beam from one side. Due to conservation of momentum, each ground-state atom which absorbs a photon, is slowed down by \( \frac{\hbar k}{m} \). After a certain time interval, the atom shall emit a photon when falling back to its ground state. This time interval depends on the life time of the excited state and can be obtained experimentally. The photons are emitted into random directions.

![Figure 1.1: (a) A photon with momentum \( \hbar k \) encounters an atom with velocity \( v \); (b) after absorption, the atom slows down to \( v - \frac{\hbar k}{m} \) because of conservation of momentum; (c) the atom emits a photon by spontaneous or stimulated emission into a random direction, causing the atom to slow down on average. Picture taken from [3].](image)

A small calculation provides some insight. Consider an ordinary sodium atom, which undergoes laser cooling. At room temperature, the Maxwell-Boltzmann velocity distribution predicts a speed of around 600 m/s for single sodium atoms. A laser of \( \sim 600 \text{ nm} \) (which lies close to the Sodium D-lines of 589 nm and 589.6 nm) can be used to perform laser cooling on the sodium beam. Per photon, this accounts for a velocity reduction of \( \frac{\hbar k}{m} \approx 3 \text{ cm/s} \). Slowing down a Sodium atom so that it reaches a corresponding hypothetical 0 K would mean that it should absorb and re-emit about 20,000 photons. Lasers can induce absorption rates of millions of photons per second, so cooling a Sodium atom takes only several milliseconds. This paragraph is only a very short and qualitative introduction into laser cooling. For further explanations on experimental techniques concerning laser cooling, see [4].

One may wonder: what are the advantages and purposes of ultra-cold atomic gases? Firstly, it enables experimentalists to create a reasonable observation time. Atoms and molecules in a gas at room temperature move at typical speeds of several hundred meters per second. This high speed makes measurements difficult: observation time and thus spectral resolution form a real problem. However, it is known that the so-called thermal velocity of the atoms in a gas is a function of temperature: \( v_{th} \propto \sqrt{T} \). This yields an immediate desire to cool gases down to extreme low temperatures, in order to be able to
measure more easily. Laser cooling can provide such a gas with an extremely low temperature. Secondly, it was expected that in ultra-cold gases, interesting phenomena would take place. This was inspired by the expression for the thermal wavelength known from thermodynamics: \( \Lambda \equiv (\beta h^2/2\pi m)^{1/2} \), with \( \beta \equiv 1/k_B T \) the reciprocal thermal energy. The point were \( \rho \Lambda^3 \sim 1 \), with \( \rho \) a characteristic density for the system, is called the transition point to quantum degeneracy. At this value, the thermal De Broglie wavelength of the system and the inter-particle separation \( \rho^{-1/3} \) are of the same order of magnitude.

At this point a transition takes place from classical to quantum statistics. The associated temperature to reach this point is about 0.02 K for liquid hydrogen. Going to a fully quantum mechanical regime requires even lower temperatures. This regime can be reached by usage of laser cooling.

By creating periodic excitations in an ultra-cold gas, one can make ordered crystal-like structures. In particular, when the atoms are excited to very high quantum numbers, the so-called Rydberg states, the ordered structures are called Rydberg crystals. We discuss Rydberg atoms in the next section and how to make Rydberg crystals from Rydberg atoms in the following paragraphs.

### 1.2 Rydberg atoms

A Rydberg atom is an excited atom of which one or more excited electrons occupy a state with a high principal quantum number \( n \), ranging from 50 to 100. This is most easily achieved for the valence electrons in alkali atoms (Cesium, Sodium, Rubidium). An example of a frequently used alkali atom is \( ^{87}\text{Rb} \) or \( ^{85}\text{Rb} \). In experiments the Rydberg state is most often chosen to be circular, in the form of an s-orbital. A schematic figure of a Rydberg atom is displayed in figure 1.2.

![Figure 1.2: A graphical interpretation of a Rydberg atom. Relatively close to the nucleus are the orbitals containing the valence electrons. The orbitals with a high principal quantum number \( n \) have a larger radius and are further away from the nucleus.](image)

Rydberg atoms have several key properties, which we shall discuss one by one:

- They have a relatively small binding energy for the excited electron, as the binding energy scales as \( n^{-2} \). The excited electron experiences a screened Coulomb potential because the core electrons effectively screen the electric field of the nucleus. Thus, the outer electron experiences a nucleus with only one proton and
will behave approximately like the electron of a hydrogen atom. The electronic binding energy is given by $E = -13.6/n^2 \text{eV}$, which is a well-known expression for the hydrogen atom. In this case $n^*$ represents an effective principle quantum number: $n^* \equiv n - \delta_l$. The parameter $\delta_l$ accounts for the deviation of the binding potential for a pure hydrogen-like situation.

- It can also be calculated that the energy difference between adjacent $n$-states is proportional to $n^3$ by considering the Rydberg atom wave functions [5]. Intuitively this is clear: subtracting two numbers of order $n^{-2}$ with $n$ large, yields a number of order at least $n^{-3}$.

- **Their radius is large.** The semi-classically derived radius goes proportional to $n^2a_0$, where $a_0$ is the the Bohr radius of about $5 \cdot 10^{-2}$ nm. As these Rydberg states have a large principle quantum number $n$, their radius is considerably large. For $n \sim 100$ the radius of the Rydberg atom is in the order of several hundred nanometers, which is larger than certain viruses.

- They have a large dipole moment. This is due to the fact that the dipole moment is linear in the radius of the state: $\vec{d} \sim qn^2a_0$. Because of this large dipole moment, Rydberg atoms have a large coupling to light. Furthermore, only transitions between circular states are allowed. These are the states with the highest allowed value for the angular momentum quantum number $l$: $l = n - 1$. In the classical limit they describe an electron in a circular orbit, hence the name.

- A Rydberg state has a long spontaneous emission life time. For atoms in a Rydberg state, the rate of spontaneous emission $\Gamma$ goes as: $\Gamma \sim n^{-3}$ [3], which means that the spontaneous emission life time goes as the inverse: $n^3$. As $n$ is large for Rydberg atoms, lifetimes of hundreds of $\mu$s can be reached. This stands in sharp contrast to spontaneous emission lifetimes of nanoseconds for lowly excited atoms.

- The polarizability of Rydberg atoms goes as $n^7$. In general, the polarizability is proportional to the sum of squares of the relevant electric dipole matrix elements divided by relevant energies, which are dominated by contributions from nearby levels. As the dipole matrix elements are proportional to $n^2$ and the energy differences to $n^{-3}$, the polarizability goes as $(n^2)^2/n^{-3} = n^7$.

A summary of the scaling properties of Rydberg atoms is displayed in table 1.1.

<table>
<thead>
<tr>
<th>Property</th>
<th>n-dependence</th>
</tr>
</thead>
<tbody>
<tr>
<td>Binding energy</td>
<td>$n^{-2}$</td>
</tr>
<tr>
<td>Energy between adjacent n-states</td>
<td>$n^{-3}$</td>
</tr>
<tr>
<td>Orbital radius</td>
<td>$n^2$</td>
</tr>
<tr>
<td>Dipole moment</td>
<td>$n^2$</td>
</tr>
<tr>
<td>Spontaneous emission life time</td>
<td>$n^3$</td>
</tr>
<tr>
<td>Polarizability</td>
<td>$n^7$</td>
</tr>
</tbody>
</table>

Table 1.1: Scaling properties of Rydberg atoms, taken from [3].

These properties make Rydberg atoms ideal for experimental purposes. If an atom is excited to a Rydberg state in an ultra-cold gas, it will stay in this state relatively long,
enabling experimentalists to manipulate the Rydberg atoms easily by making use of their large dipole moment. A large dipole moment effectively means that the Rydberg atoms can be manipulated by the usage of the electric field generated by, for example, a laser. In the next paragraph we consider long-range van der Waals interactions between Rydberg atoms and how these can be used to create ordered structures of single Rydberg atoms.

1.3 The van der Waals interaction

Rydberg crystals are highly ordered structures, formed by placing single Rydberg atoms in special patterns. The possibility of formation of these ordered structures in a disordered atomic gas is a consequence of the previously discussed special properties of the Rydberg atoms. It was earlier presented that the polarizability of Rydberg atoms scales as \( n^7 \). This causes a van der Waals type of interaction to arise between Rydberg atoms. Let us take a closer look at the origin of this.

From elementary electrostatics it is known that the coordinate free form for the electric field of a pure dipole is given by [7]:

\[
E_{dp}(r) = \frac{1}{4\pi\epsilon_0} \frac{1}{r^3} \left( (d \cdot \hat{r}) \hat{r} - d \right),
\]

where \( \hat{r} \) is a unit vector pointing from one dipole to another and \( r \) the distance between the two dipoles. Furthermore, \( d \equiv qr \) is the dipole moment for a specific dipole consisting of charge \( q \). Finally, \( r \) is the vector pointing from accumulated positive charge to accumulated negative charge inside the dipole. The situation is displayed in figure 1.3.

The interaction Hamiltonian of two dipoles is therefore given by:

\[
\hat{H}(r)_{int} = -\hat{d}_1 \cdot \hat{E}_2 = \frac{1}{4\pi\epsilon_0} \frac{1}{r^3} \left( \hat{d}_2 \cdot \hat{d}_1 - 3(\hat{d}_2 \cdot \hat{r})(\hat{r} \cdot \hat{d}_1) \right). \tag{1.2}
\]

Figure 1.3: A schematised view of the two dipoles interacting. The distance between the dipoles is \( r \).

The first atom has dipole moment \( d_1 \) and makes an angle \( \phi_1 \) with the line joining the centers of the two dipoles. The parameters for the second dipole are analogously defined.
This interaction Hamiltonian is a perturbation to the total Hamiltonian, which contains kinetic and potential energy terms for the separate atoms. We now choose the dipoles to be on a line. In Figure 1.3 this comes down to choosing \( \phi_1 = 0 \) and \( \phi_2 = 0 \), which can be done without loss of generality. This yields:

\[
\hat{H}(r)_{\text{int}} = -\frac{q^2 r_1 r_2}{2\pi\epsilon_0 r^3},
\]

where \( q \) is the amount of separated charge inside one dipole, which is assumed to be the same for both dipoles. The intramolecular distances \( r_{1,2} \) separate the dipole charge. We can now use perturbation theory to calculate the first order ground-state energy associated with the interaction Hamiltonian. For this we assume that the wave function of the system can be written as the product of two one-particle wave functions:

\[
\psi_n(r_1, r_2) = \psi_{n,1}(r_1)\psi_{n,2}(r_2)
\]

Using perturbation theory, the first order ground-state energy is given by:

\[
E^{(1)}_0 = \langle \psi_0(r_1, r_2) | \hat{H}(r)_{\text{int}} | \psi_0(r_1, r_2) \rangle
\]

where it was used that within an atom, the charge is randomly fluctuating. Thus, the average distance between negative and positive charge inside a dipole is zero. The second order ground-state energy is then given by:

\[
E^{(2)}_0 = \sum_{n=1}^{\infty} \frac{\left| \langle \psi_n(r_1, r_2) | \hat{H}(r)_{\text{int}} | \psi_0(r_1, r_2) \rangle \right|^2}{E_0 - E_n},
\]

which is always negative as \( E_n > E_0 \) \( \forall n \). For a general system, the higher order wave functions are not known, so that this expression cannot be evaluated. What we can do, is find a general scaling law by using the scaling laws of paragraph 1.2. We know that the dipole moments per atom scale with \( n^2 \), which yields a total scaling of \( n^8 \) for the denominator of equation (1.5). Furthermore, it was also argued that the difference in energy between adjacent \( n \)-states scales with \( n^3 \). This then yields a total scaling for the second-order ground-state energy of \( n^{11} \). Plugging in equation (1.3) into equation (1.5) yields a qualitative expression for the interaction energy:

\[
U_{VdW} = \frac{C_6}{r^6},
\]

which is a van der Waals type of interaction because of the characteristic \( r^{-6} \). The strength of the interaction is characterized by the \( C_6 \) parameter, for which we now know that it scales with \( n^{11} \). This is a qualitative expression, as (1.5) only holds for ground-state atoms, whereas Rydberg atoms are highly excited. Note that the van der Waals interaction energy can be positive or negative. However, we shall consider systems in which it is strictly positive. As the system wants to minimize its energy the atoms shall aspire to distance themselves from each other. This means that the force between two Rydberg atoms is repulsive. For further inquiries concerning this subject, see [8]. We shall now consider how these van der Waals interactions between Rydberg atoms can cause ordered structures to appear.
1.4 Rydberg crystals

In the previous section it was argued that Rydberg atoms experience a van der Waals interaction. This van der Waals interaction is repulsive. Let us consider an ultra-cold diluted atomic gas of Rubidium atoms. If we shine lasers with the correct frequency on it, we might excite some atoms to the Rydberg state. As the van der Waals interaction is repulsive, it is not energetically favorable for two Rydberg atoms to be close to one another.

The phenomenon that a Rydberg atom prevents atoms close to it to be excited to a Rydberg state is called the Rydberg blockade effect. This effect belongs to the larger group of general blockade interactions. These interactions all have in common that a single particle prevents the excitation of other particles. This provides a mechanism for the control of quantum states. The Rydberg blockade effect has been shown in a many-body regime, using ultra-cold atoms a decade ago. However, in order to use the Rydberg blockade effect for controlled quantum systems, a smaller number of particles should be used. Recently, it has been demonstrated that a single Rydberg-excited rubidium atom blocks excitation of a second atom located more than 10 µm away [10].

A visual interpretation of the Rydberg blockade effect is displayed in figure 1.4. Con-

![Diagram](image1)

**Figure 1.4:** In (a) a line of atoms in the ground state is displayed. Exciting one atom to a Rydberg state as in (b), causes the energy levels of the surrounding atoms to shift upwards due to the van der Waals interaction. This effect is largest for the nearest neighbors of the excited atom and weakens for second and third neighbors. The laser with frequency \( \omega \) which was used to excite the first atom, is now no longer resonant with the states of the other atoms.
sider a line of atoms all in their ground state. Imagine that one of these atoms is excited to a Rydberg state by usage of a resonant laser with frequency $\omega$. Due to the van der Waals interaction the energy levels of the Rydberg states of the surrounding atoms become higher. The initially used laser is therefore not resonant anymore with any of the atomic transitions. This is the Rydberg blockade effect in its essence.

As the van der Waals interaction scales with $n^{11}$, it is tunable by choosing a range of Rydberg states. In an ultra-cold gas, it should be possible to reach a regime in which the van der Waals interaction energy dominates the kinetic energy of the single atoms. The atoms are then called to be strongly coupled. In this case the range of the van der Waals interaction is much larger than the average separation of the particles. It is therefore called a long range interaction, although it falls off with $r^{-6}$.

If we manage to get more and more atoms excited to a Rydberg state, we create a strongly-coupled system due to the van der Waals interaction. Due to energy minimization we expect certain special configurations to emerge. Recently it has been predicted that many-body correlations can arise from the blockade effect [12]. These ordered structures are called Rydberg crystals and can be calculated theoretically. The 2D-case is displayed in figure 1.5 for several numbers of atoms [13].

![Figure 1.5: Two-dimensional Rydberg crystal structures as a function of the number of Rydberg atoms $m$. The circles indicate the excitation volume. For low $m$, the Rydberg atoms reside on the edge of the excitation volume. From $m = 7$ there are also Rydberg atoms located in the middle of the excitation volume.](image-url)
1.5 Rydberg research: why bother?

Interest in Rydberg atoms and crystals has increased in the scientific community over the past decade. One may wonder why this is the case. Several good reasons can be presented. Firstly, Rydberg systems are interesting to investigate many-body physics. As described before: an ultra-cold atomic gas can reach the regime where the kinetic energy is of less importance than the van der Waals interaction energy. It is then called a strongly-coupled system. For short timescales the system is in the frozen gas regime. This means that atoms need several tens, maybe one hundred $\mu$s to move over typical interatomic distances, whereas the Rydberg excitation dynamics take place on a smaller scale of $1 - 10 \, \mu$s \cite{13}. In such an ultra-cold gas it is a good approximation to assume that the atoms are frozen. It therefore gives the ideal circumstances to investigate many-body physics on an atomic scale. One might find interesting new physics in this regime.

A second reason to study Rydberg crystals is their relation to solid state physics. In the recent years, optical lattices have been used to create toy-model solid state systems. An optical lattice - see figure 1.6 - is just a periodic potential caused by a periodic spatial polarization pattern. It is generated by the interference of laser beams. It may be used to trap neutral ultra-cold atoms, so that one atom is located at each potential minimum. This periodic structure resembles the periodic potential in a lattice. The big difference is that the atoms do not really interact. They can tunnel from one potential minimum to another, but this is rather unlikely.

Rydberg crystals are a better model for dilute metallic solids, as is thoroughly described in \cite{14}. It turns out that collectively exciting atoms to Rydberg levels can yield a situation in which the charge-density distributions of neighboring atoms overlap. This causes a system which resembles a strongly-coupled solid state. It is even possible to compute classic solid-state parameters like the hopping rate of the electrons in such a system. The fact that the Rydberg systems are rather dilute and have a low Fermi temperature - compared to ’ordinary’ solid state systems - it makes them attractive for the study and simulation of strongly correlated electronic systems.

Figure 1.6: An artist impression of an optical lattice. Neutral atoms are trapped in periodic potential minima, created by interfering laser beams.

Finally, Rydberg systems are of particular interest for their possible usage in quantum
computers. The main point is that one atom can be used to manipulate the state of an other atom by using the Rydberg blockade effect. This gives one the ability to perform computational operations as it is possible to coherently control these interactions. Add this to the fact that Rydberg atoms have a long lifetime and one recognizes a suitable candidate for a quantum computer. Several interesting proposals have been made to use the Rydberg blockade effect for quantum computing. One example is the creation of multibit CNOT-gates as is described in [15]. Another example is the implementation of a search algorithm, called the Grover algorithm, as proposed in [16]. A thorough review of the usage of Rydberg system in order to establish quantum computing can be found in [17].

1.6 Summary and outline

In this chapter we began with a discussion of laser-cooling as a technique to create ultracold gases. We then discussed Rydberg atoms and their extreme properties. Following this discussion we derived the long-range van der Waals interaction, which causes the Rydberg blockade mechanism to occur. With this, we were able to describe the aspects of Rydberg crystals, formed from Rydberg atoms. We ended with a justification of the research into Rydberg crystals. Now we know all this, we continue our investigation into Rydberg atoms and crystals.

The general outline of this report is as follows:

- In chapter 2 we discuss a protocol to obtain Rydberg atoms, called STIRAP. In order to understand this protocol we review basic theory from quantum optics, including Rabi oscillations. We then consider the STIRAP protocol for one particle. Furthermore, we present realistic experimental parameters, which we use in our further discussion. Finally, we investigate the adiabaticity condition for the single-particle STIRAP protocol, both theoretically and experimentally.

- In chapter 3 we present a generalized STIRAP protocol for $N$ particles by investigating the $N$-particle Hamiltonian. We then present a analytical solution for three two-level particles on a line, using perturbation theory. We continue with a search for the eigenvectors of the Hamiltonian which yield a correct STIRAP procedure. Finally, we try to implement the STIRAP procedure for $N$ particles without spontaneous emission.

- In chapter 4 we start with a review of the derivation of the spontaneous emission rate in the Wigner-Weisskopf approximation. We continue with reviewing density operator theory and a possible way to implement this for $N$ particle STIRAP with spontaneous emission. We conclude that this master equation approach shall not be possible for a large number of particles, due to problems with the computation time. We therefore present a stochastic alternative, called the quantum Monte Carlo approach. We eventually apply this to three particles on a line.

- Finally, in chapter 5 we draw conclusions from the research we have done. We shall also give recommendations for further inquiries.
Chapter 2

STIRAP

In this chapter we introduce a procedure to get isolated ground-state atoms into a Rydberg state. This procedure is called STIRAP, which is short for Stimulated Raman Adiabatic Passage Scheme. The scheme makes use of a three-level system: besides the ground \( |g\rangle \) and Rydberg state \( |r\rangle \), there is also an intermediate excited state \( |e\rangle \), for a direct transition from \( |g\rangle \) to \( |r\rangle \) is not allowed by the selection rules. In order to understand the STIRAP scheme we must first review Rabi oscillations. This is done in the first section. After that, we describe the STIRAP scheme. This is followed by a description of the experimental setup which is used for STIRAP procedures. We end the chapter with a discussion of the adiabaticity of the STIRAP procedure.

2.1 Rabi oscillations

In this section we follow the discussion by Knight and Gerry [6]. Consider an atom with two relevant energy levels: a ground state \( |g\rangle \) with energy \( E_g \) and an excited state \( |e\rangle \) with energy \( E_e \). The system is characterized by a transition frequency: \( \omega_0 = (E_e - E_g)/\hbar \). We would like to investigate the occupation of the excited state when the atom is subjected to an external electric field. Let us assume that this electric field is of the form \( E(t) = E_0 \cos(\omega t) \). The typical frequency \( \omega \) of the electric field is assumed to be close, but not necessarily equal to the transition frequency \( \omega_0 \). We can therefore define a non-zero detuning \( \Delta \equiv \omega - \omega_0 \). The reader should note that the detuning can either be positive or negative, depending on the fact whether the laser frequency is higher or lower than the atomic transition frequency. A sketch of the system is given in figure 2.1.

One can show that the total Hamiltonian is the sum of a free-atom Hamiltonian \( \hat{H}_{\text{atom}} \), a free-field Hamiltonian \( \hat{H}_{\text{field}} \) and an interaction Hamiltonian \( \hat{H}_I \) of the atom-field system:

\[
\hat{H}(t) = \hat{H}_{\text{atom}} + \hat{H}_{\text{field}} + \hat{H}_I = \hat{H}_0 - \hat{d} \cdot \mathbf{E}_0 \cos(\omega t),
\] (2.1)

where \( \hat{d} \) is the dipole moment operator of the atom. In this time-dependent system we can write the state vector of the system as

\[
|\Psi(t)\rangle = C_g(t) \exp(-iE_g t/\hbar) |g\rangle + C_e(t) \exp(-iE_e t/\hbar) |e\rangle.
\] (2.2)

Plugging this wave vector into the Schrödinger equation yields, upon taking the inner product with the two states, a coupled set of equations for the probability amplitudes
Figure 2.1: A visual interpretation of the Rabi problem. An atom has two relevant levels: a ground state $|g\rangle$ and an excited state $|e\rangle$. The transition frequency is given by $\omega_0 = (E_e - E_g)/\hbar$. The offered light does not have the same frequency, so that a detuning $\Delta \equiv \omega - \omega_0$ can be defined. In this figure, $\Delta < 0$.

$C_e$ and $C_g$:

$$
\begin{align*}
\dot{C}_g &= \frac{i}{\hbar}(\mathbf{d} \cdot \mathbf{E}_0)_{eg} \cos(\omega t) \exp(-i\omega_0 t) C_e \\
\dot{C}_e &= \frac{i}{\hbar}(\mathbf{d} \cdot \mathbf{E}_0)_{eg} \cos(\omega t) \exp(+i\omega_0 t) C_g,
\end{align*}
$$

where $(\mathbf{d} \cdot \mathbf{E}_0)_{eg} \equiv \langle e | \mathbf{d} \cdot \mathbf{E}_0 | g \rangle$. In these expression Euler’s formula can be used to write the cosine as a complex exponential. This yields two exponentials: one oscillating at frequency $\omega_0 - \omega$ and one at frequency $\omega_0 + \omega$. A frequently used approximation consists of neglecting the larger frequency in order to obtain a simpler expression. This is called the Rotating Wave Approximation (RWA) and shall be used more often throughout this report. A visual interpretation of this approximation is displayed in figure 2.2.

Figure 2.2: A visual interpretation of the Rotating Wave Approximation. On the left a plot is made of the real part of $e^{ix} + Ae^{100ix}$, with $A$ a small amplitude. Applying the RWA consists of plotting the real part of $e^{ix}$.

This set of coupled first-order differential equations can be transformed into one second-order differential equation. Taking as an Ansatz a complex exponential $C_e(t) = e^{i\lambda t}$ yields two eigenvalues:

$$
\lambda = \frac{1}{2} \left( -\Delta \pm \sqrt{\Delta^2 + \frac{(\mathbf{d} \cdot \mathbf{E}_0)_{eg}^2}{\hbar^2}} \right).
$$

(2.4)
Using as an initial condition $C_g(0) = 1 \land C_e(0) = 0$, i.e. the atom begins in the ground state, yields a final solution

$$
\begin{cases}
  C_e(t) &= -i((d \cdot E_0)_{eg}/\Omega_R h)e^{-i\Delta t/2} \sin(\Omega_R t/2) \\
  C_g(t) &= e^{-i\Delta t} (\cos(\Omega_R t/2) + i(\Delta/\Omega_R) \sin(\Omega_R t/2)),
\end{cases}
\quad (2.5)
$$

in which the generalised Rabi frequency is defined as: $\Omega_R = \sqrt{\Delta^2 + ((d \cdot E_0)_{eg}/h)^2}$. For the case of $\Delta = 0$ we denote the ordinary Rabi frequency as $\Omega_0$. When we shall speak about the Rabi frequency in the remainder of this report, we imply $\Omega_0$, unless stated otherwise. Taking the square of the absolute value of $C_e(t)$ and $C_g(t)$ yields the chance to measure the atom in the excited or ground state:

$$
\begin{cases}
  P_e(t) \equiv |C_e(t)|^2 = \left(\frac{\Omega_0}{\Omega_R}\right)^2 \sin^2(\Omega_R t/2) \\
  P_g(t) \equiv |C_g(t)|^2 = 1 - \left(\frac{\Omega_0}{\Omega_R}\right)^2 \sin^2(\Omega_R t/2).
\end{cases}
\quad (2.6)
$$

In these expressions for $P_e(t)$ and $P_g(t)$ we immediately see that the atom oscillates between $|e\rangle$ and $|g\rangle$ as a function of time. Oscillations of this sort are called Rabi oscillations. A plot for several detunings $\Delta$ is displayed in figure 2.3.

### 2.2 The STIRAP procedure

In effect, STIRAP is a technique that permits control of the population transfer between two states in an atom. It allows one to completely transfer the population of one state to another state, if the states are chosen with care. The interesting point is that the STIRAP procedure does not use one laser resonantly coupling two states, but rather two lasers coupling three states in total. This has two major advantages. Firstly, one can establish population transfer between states for which single-photon transitions are forbidden by the selection rules for electromagnetic radiation. Secondly, the excitation efficiency is less sensitive to the shapes and specifications of the laser pulses [18].

The STIRAP procedure is essentially different from using standard Rabi oscillations. When using Rabi oscillations for population transfer, the used Rabi frequency is constant and therefore the eigenstates are constant as well. In this case, we do not start in an eigenstate and use the resulting oscillations to cause a population transfer by careful timing. In contrast to this, STIRAP uses time-dependent Rabi frequencies. These Rabi frequencies are chosen as to manipulate the Hamiltonian and thereby also the eigenstates in time. When using the STIRAP procedure, we do start in an eigenstate. Since this eigenstate evolves in time from the initial state to the desired state, the aim is to make the system follow this eigenstate in time.

Consider the system displayed in figure 2.4. A laser with Rabi frequency $\Omega_P$, called the Pump laser, couples a ground state $|g\rangle$ to an excited state $|e\rangle$. Another laser with Rabi frequency $\Omega_S$, called the Stokes laser, couples the excited state $|e\rangle$ to a Rydberg state $|r\rangle$. Note that the Rabi frequencies are proportional to the electric field of the
laser and therefore a measure for the intensity of the laser: $I \propto \sqrt{\Omega}$. The laser might be off-resonance, hence the detunings $\Delta_P$ and $\Delta_S$. These are defined to be positive when the laser frequency exceeds the transition frequency. Effectively, the ground state is coupled to the Rydberg state via the intermediate excited state.

As we have seen earlier, the Rydberg state has a lifetime proportional to $n^3$, which is in the order of hundreds of $\mu s$ for the systems we are concerned with. The pulses shall have a characteristic duration of tens of $\mu s$, so that decay from the Rydberg state can be ignored. However, the intermediate excited state might have a spontaneous emission lifetime in the order of a $\mu s$. It is therefore important that this state is only used to couple the ground and Rydberg state, it should attain no - or as little as possible - population during the excitation scheme.

The STIRAP procedure consists of using the pulse scheme in figure 2.5. The order of the pulses in the scheme is rather counter-intuitive. We start with a Stokes laser pulse, which couples the intermediate and the Rydberg state. Then we apply a pump

Figure 2.3: Plots of Rabi oscillations for an atom in a two-level system for three values of the detuning $\Delta$. In all plots, $\Omega_0 = 1$ Mrad/s. The chance for the atom to be in the excited state, i.e. $|C_e(t)|^2$, is displayed in purple. The chance for the atom to be in the excited state, i.e. $|C_g(t)|^2$, is displayed in blue. The general trend is that for a non-zero detuning there is no time at which there is a probability of 1 to detect the atom in $|e\rangle$ or $|g\rangle$. 

(a) $\Delta = 0$

(b) $\Delta = 0.5$ Mrad/s

(c) $\Delta = 1$ Mrad/s
Figure 2.4: A three-level system. A laser with Rabi frequency $\Omega_P$, called the Pump laser, couples states $|g\rangle$ and $|e\rangle$. Another laser with Rabi frequency $\Omega_S$, called the Stokes laser, couples states $|e\rangle$ and $|r\rangle$. The laser might be off-resonance, hence the detunings $\Delta_P$ and $\Delta_S$. These are negative in this figure. Spontaneous emission might occur from the second level.

pulse, which couples the ground and the intermediate state. This is done in such a way that two the pulses overlap. Using STIRAP, we can cause an atom in the ground state to be transferred to the Rydberg state, which is exactly what we seek to do. Furthermore, during the STIRAP procedure, we always acquire a probability of zero of being in the intermediate state. This is important to avoid spontaneous emission.

Figure 2.5: The STIRAP scheme.

To understand why the STIRAP procedure works, consider the Hamiltonian of the
three-level system with one particle. This Hamiltonian can be written as

\[
H(t) = \frac{\hbar}{2} \begin{pmatrix}
0 & \Omega_P & 0 \\
\Omega_P & -2\Delta_P & \Omega_S \\
0 & \Omega_S & -2(\Delta_P + \Delta_S)
\end{pmatrix}
\] (2.7)

where the Rabi frequencies can be time dependent. This is a Hamiltonian in the rotating frame, where the rotating wave approximation has been used. This Hamiltonian can be found from the general Hamiltonian (B.20) derived in appendix B.1. The used basis is \{\{g\},\{e\},\{r\}\} ≡ \{(1 0 0)^T, (0 1 0)^T, (0 0 1)^T\}. Moreover, we have assumed that the timescales we consider are small enough, so that we can ignore the motions of the atoms. This is called the frozen-gas limit, which we will use throughout this entire report. Thus, the Hamiltonians in this report never contain kinetic energy terms. For the STIRAP scheme to work, two-photon resonance is required, i.e. \(\Delta_P = -\Delta_S\), for reasons we will mention later. The reader should note that when \(\Omega_S = 0\), the third level remains unpopulated.

When using two-photon resonance, it is easy to derive the eigenvalues [18]:

\[
\begin{cases}
\omega^\pm = -\Delta_P \pm \sqrt{\Delta_P^2 + \Omega_P^2 + \Omega_S^2} \\
\omega^0 = 0.
\end{cases}
\] (2.8)

From these, the corresponding eigenvectors follow:

\[
\begin{align*}
|a^+\rangle &= \sin \Theta \sin \Phi |g\rangle + \cos \Theta \sin \Phi |e\rangle + \cos \Theta \cos \Phi |r\rangle \\
|a^-\rangle &= \sin \Theta \cos \Phi |g\rangle - \sin \Theta |e\rangle + \cos \Theta \cos \Phi |r\rangle \\
|a^0\rangle &= \cos \Theta |g\rangle - \sin \Theta |r\rangle,
\end{align*}
\] (2.9)

where \(\Theta = \Theta(t)\) is called the mixing angle, defined as

\[
\Theta \equiv \arctan \left( \frac{\Omega_P}{\Omega_S} \right). \] (2.10)

The expressions for \(\sin \Theta\) and \(\cos \Theta\) follow from this expression and normalisation of the eigenvectors. \(\Phi = \Phi(t)\) is defined as [19]

\[
\Phi \equiv \arctan \left( \frac{\sqrt{\Omega_P^2 + \Delta_P^2}}{\sqrt{\Omega_S^2 + \Delta_S^2 + \Delta_P^2 - \Delta_P}} \right). \] (2.11)

In order to explain the STIRAP procedure, we shall utilise the well-known adiabatic theorem from quantum mechanics. This theorem states the following. Suppose that the state vector is in one of the eigenstates of the Hamiltonian. If the Hamiltonian is subsequently changed in time, the eigenstate will also change in time. The state vector will then follow the eigenstate in time, provided the change of the Hamiltonian occurs slowly enough. Furthermore, the eigenvalue corresponding to the eigenstate which is to be followed must not be degenerate.
Let us now apply the STIRAP pulse scheme from figure 2.5. In the beginning, when both lasers are off, it follows from (2.8) that $\omega^0$ is degenerate with either $\omega^+$ or $\omega^-$, depending on the sign of $\Delta P$. We can lift this degeneracy by turning on either one of the lasers. If we turn on only the Stokes laser, which couples $|e\rangle$ and $|r\rangle$, then $\Omega_S \neq 0$ and $\Omega_P = 0$, so $\Theta = 0$. From the eigenvectors (2.9), we see that in this case $|a^0\rangle = |g\rangle$ and $|a^0\rangle$ does not change as long as $\Omega_P = 0$, whilst the other eigenstates have no components of $|g\rangle$. So, if an atom starts in the ground state, the state vector is in the eigenstate $|a^0\rangle$ of the system.

We then start turning on the pump laser and after a while start turning off the Stokes laser. From equation (2.10) it follows that this causes $\Theta$ to change from 0 to $\pi/2$. Furthermore, we see that the eigenvector $|a^0\rangle$ becomes a superposition of $|g\rangle$ and $|r\rangle$. Since the eigenvalue $\omega^0$ is non-degenerate, the state vector can follow $|a^0\rangle$ adiabatically. When the Stokes laser is off, $\Theta = \pi/2$ and $|a^0\rangle = |r\rangle$. If the state vector has followed $|a^0\rangle$ adiabatically, the atom will be in the Rydberg state. The adiabatic following is now completed, so we can turn off the pump laser and $\omega^0$ becomes degenerate again.

Thus, by applying the STIRAP process as is displayed in figure 2.5, the eigenstate $|a^0\rangle$ changes from $|g\rangle$ to $|r\rangle$ and the state vector can adiabatically follow this eigenvector. Furthermore, $|a^0\rangle$ never acquires a component of the intermediate state, so spontaneous emission is avoided. We can therefore bring an atom from the ground state to the Rydberg state with a probability of 1. Had we not used two-photon resonance, the eigenvector $|a^0\rangle$ would acquire a component of the intermediate state during the pulse scheme and the eigenvalues would be degenerate at some point during the STIRAP scheme [19]. Both these features would prevent the possibility of following the eigenvector. Moreover, the proceedings described above must be carried out slowly enough for adiabatic following to occur. We will discuss the meaning of 'slow enough' later. Before this, we shall consider an experimental situation which is often used in Rydberg physics.

### 2.3 Experimental setup and restrictions

For the creation of Rydberg crystals, one often uses Rubidium (Rb) atoms. If figure 2.4 depicts a Rb atom, the ground state is a 5s state, the intermediate state is a 5p state and the Rydberg state is a 65s state. The 5p state contains a spontaneous emission rate of 5.98 MHz [4]. Furthermore, the wavelength of the transition from 5s to 5p is 780 nm and the wavelength of the transition from the 5p to the 65s state is 480 nm. The transition dipole moments are $d_{ge} = 2.9\, ea_0$ and $d_{sr} = 5 \cdot 10^{-3}\, ea_0$ respectively [20]. Here, $e$ is the elementary charge and $a_0$ is the Bohr radius.

Using these parameters, we can calculate some realistic values for the Rabi frequencies $\Omega_P$ and $\Omega_S$. The intensity of the laser is given by

$$I = \frac{P}{A} = \frac{1}{2}\epsilon_0 c E_0^2$$

with $P$ the power of the laser, $A$ the area of the laser spot, $E_0$ the amplitude of the electric field, $\epsilon_0$ the vacuum permittivity and $c$ the speed of light. Using the previously defined Rabi frequency (see section 2.1) we immediately find the following expression.
for the Rabi frequency of a laser:

$$\Omega = \frac{d}{\hbar} \sqrt{\frac{2P}{\Delta v_0 c}}. \tag{2.13}$$

Substituting all constants and parameters then yields the Rabi frequencies $\Omega_S = 4.0 \cdot 10^2$ Mrad/s and $\Omega_P = 2.3 \cdot 10^5$ Mrad/s for a 100 mW laser with a 10 $\mu$m FWHM laser spot diameter.

For the creation of Rydberg crystals, one creates an array of laser spots at all positions where a Rydberg atom is desired. This can be achieved by shining a laser at a *Spatial Light Modulator* (SLM). The SLM, displayed in figure 2.7, can shape the laser beam into an arbitrary pattern, in this case an array of spots as displayed in figure 2.8. If we take into account some intensity loss caused by the SLM and the fact that the intensity of the laser must be divided over all spots, then we can assume that reasonable Rabi frequencies for creating Rydberg crystals are $\Omega_S = 10$ Mrad/s and $\Omega_P = 40$ Mrad/s. For more information on the SLM, see [21].
2.4 Adiabaticity criterion

In section 2.2 we mentioned that STIRAP depends on the state vector adiabatically following of eigenvector $\ket{a^0}$. For this to occur, the adiabatic theorem states that the STIRAP procedure must be applied slowly enough. To see what ‘slow enough’ means, we will run simulations of the state vector. The script used for the simulations can be found in appendix C.4. This script calculates the time evolution of the state vector by solving the Schrödinger equation with the Hamiltonian (2.7). We use Gaussian shaped laser pulses. Hence the Rabi frequencies can be written as

$$\begin{align*}
\Omega_P &= \Omega_{P,0} \exp\left(-\frac{(t - a_P)^2}{2\sigma_P^2}\right), \\
\Omega_S &= \Omega_{S,0} \exp\left(-\frac{(t - a_S)^2}{2\sigma_S^2}\right),
\end{align*}$$

in which $a_P$ and $a_S$ are translation coefficients in units of time and $\sigma_{P,S}$ the standard deviations of the Gaussian curves in units of time. We assume that the two laser pulses have the same shape and only differ a translation in time, i.e.: $\sigma_P = \sigma_S \equiv \sigma$. For the STIRAP process, we can take $a_P = 0$ and $a_S < 0$. To see that this script yields the correct results, we run the script with $\Omega_S = 0$, $\Delta_P = 1$ Mrad/s and $\sigma = \infty$ so that $\Omega_P(t) = 1$ Mrad/s. We then expect Rabi oscillations between the levels $|g\rangle$ and $|e\rangle$, with a period of $2\pi/\sqrt{\Omega_P^2 + \Delta_P^2} \approx 4.4\mu$s and an amplitude $\Omega_P^2/\left(\Omega_P^2 + \Delta_P^2\right) = 0.5$, where we used equation (2.6). The simulation yields figure 2.3 (c), which is the correct result.

We now run the script using a STIRAP pulse scheme, with the Rabi frequencies given by equation (2.14) with $\Omega_{P,0} = \Omega_{S,0} = 10$ Mrad/s and $\Delta_P = 0$. The result for different timescales is shown in figure 2.9, where the vertical axis contains the chances of being in a certain state. In figure 2.9 (a), the eigenvector $|a^0\rangle$ is followed adiabatically. The simulation in figure 2.9 (b) is done using a timescale which is approximately ten times as short. Clearly, the eigenvector $|a^0\rangle$ is not followed in this case. One can notice some Rabi oscillations in this simulation, which are typical for non-adiabatic processes.
From the non-adiabatic process in figure 2.9 (b), two things are worth noting. Firstly, the final probability of being in the Rydberg state is not 1. Secondly, the state vector acquires a component of the intermediate state |e⟩. We can therefore introduce two quantities to quantify the adiabaticity of a procedure: the final probability of being in the Rydberg state |⟨r|ψ⟩|_end and the maximum probability of being in the intermediate state |⟨e|ψ⟩|_max. Using these quantities, we study the effect of the laser parameters on the adiabaticity.

In figure 2.10 (a) we plotted the final probability of being in the Rydberg state as a function of the timescale, for different amplitudes of the Rabi frequencies. The horizontal axis shows the value of $\sigma = \sigma_P = \sigma_S$. Furthermore, we choose $a_P = 0$ and $a_S = -2\sigma$, so that the laser scheme always looks like figure 2.5 but with a varying time axis. From figure 2.10 (a) it follows that a certain timescale is required to get a perfect population transfer to the Rydberg state. This is in accordance with the adiabatic theorem. It also follows that for larger amplitudes of the Rabi frequencies, shorter timescales are possible for population transfer. Furthermore, from figure 2.10 (b) it follows we require a certain timescale to suppress the population of the intermediate state. For larger amplitudes of the Rabi frequency, the required timescales are shorter. Therefore, we can conclude from figures 2.10 (a) and (b) that a process is more adiabatic when large Rabi frequencies are used, which is equivalent to using stronger lasers.

In figure 2.11 we plotted both the final probability of being in the Rydberg state and the maximum probability of being in the intermediate state as a function of the detuning of the intermediate state. Each simulation is performed using identical amplitudes of the Rabi frequencies and identical timescales. One can see that the maximum population of the intermediate state decreases for a larger detuning $\Delta_P$. However, this is exactly what we would expect considering standard Rabi oscillation theory (see section 2.1). In figure 2.9 (b), one can see that in the non-adiabatic process Rabi oscillations between the ground and intermediate state arise. An increased detuning of the intermediate state
then suppresses the probability amplitude of this state, analogous to equation (2.6). In this case, a better indicator for adiabaticity is the final probability of the Rydberg state, which decreases when $\Delta P$ increases. So apparently, for better adiabaticity we wish to keep $\Delta P$ as low as possible.

To summarise, we conclude that to enhance adiabatic following, we must use:

- larger timescales
- larger Rabi frequencies
- smaller detunings of the intermediate state.

Figure 2.11: Testing adiabaticity as a function of $\Delta P$. The red plot depicts the final probability of being in the Rydberg state, the black plot depicts the maximum probability of being in the intermediate state. The used laser pulses are given by equation (2.14) with $a_P = 0$, $a_S = -4\mu s$, $\sigma = 2\mu s$ and $\Omega_{P,0} = \Omega_{S,0} = 5\text{ Mrad/s}$. 
To see the effect of the Rabi frequencies induced by the lasers and the detuning $\Delta P$ on the adiabaticity, we can also use the following criterion for adiabaticity of a process: 

$$|\langle \dot{a}^0 | a^\pm \rangle| \ll |\omega^\pm - \omega^0|.$$ 

(2.15)

The reader should note that these are in fact two criteria: one for $|a^+\rangle$ and $\omega^+$ and one for $|a^-\rangle$ and $\omega^-$. This condition is in accordance with the adiabatic theorem: if larger timescales are used, the time derivative of $|a^0\rangle$ decreases and the process is less adiabatic. Moreover, if the $\omega^0$ is degenerate, the criterion cannot be obeyed.

Upon substitution the expressions for the eigenvalues (2.8) and eigenvectors (2.9), we find

$$\begin{aligned}
|\dot{\Theta} \sin \Phi| &\ll \left| -\Delta P + \sqrt{\Delta P^2 + \Omega_P^2 + \Omega_S^2} \right| \\
|\dot{\Theta} \cos \Phi| &\ll \left| \Delta P + \sqrt{\Delta P^2 + \Omega_P^2 + \Omega_S^2} \right| ,
\end{aligned}$$

(2.16)

where both criteria should hold. To simplify the criteria, we replace both $\sin \Phi$ and $\cos \Phi$ by 1 and note that if the resulting criteria hold, then the criteria of equation (2.16) hold as well. By using equation (2.10) to determine $\dot{\Theta}$, we can then write the adiabaticity criteria as one criterion:

$$\left| \frac{\hat{\Omega}_P \Omega_S - \Omega_P \hat{\Omega}_S}{\Omega_P^2 + \Omega_S^2} \right| \ll \sqrt{\Delta P^2 + \Omega_P^2 + \Omega_S^2} - |\Delta P|. $$

(2.17)

The reader should note that the left-hand side of this criterion does not depend on the detuning of the intermediate state or the amplitude of the Rabi frequencies. Thus, upon inspection of the right-hand side it becomes clear that if amplitude of either Rabi frequency increases, the adiabaticity improves. Furthermore, from

$$\frac{d}{d|\Delta P|} \left( \sqrt{\Delta P^2 + \Omega_P^2 + \Omega_S^2} - |\Delta P| \right) = \frac{|\Delta P|}{\sqrt{\Delta P^2 + \Omega_P^2 + \Omega_S^2}} - 1 < 0$$

(2.18)

it becomes clear that when the absolute detuning of the intermediate state increases, the adiabaticity declines. These findings correspond with what we found earlier by running the simulations.

### 2.5 Summary

In this chapter we started with a review of Rabi oscillation theory. We then introduced the STIRAP procedure, which can be used to take an atom from the ground state to the Rydberg state, by coupling these states to an intermediate state. This state remains unpopulated during the procedure, which is important to avoid spontaneous emission. After this, we discussed an experimental setup that is often used for STIRAP procedures. Because STIRAP depends on the adiabatic following of an eigenvector, we subsequently discussed what conditions are most appropriate for adiabatic processes. We found that higher Rabi frequencies and a lower magnitude of the detuning of the intermediate state improve the adiabaticity.

We have now seen that STIRAP can bring one atom to the Rydberg state with a probability of 1. In the next chapter we shall investigate if we can also bring multiple particles to the Rydberg state by using STIRAP schemes.
Chapter 3

Multi-particle STIRAP without spontaneous emission

In this chapter we aim to use the STIRAP procedure, which we introduced in chapter 2, to excite multiple particles to the Rydberg state. We will start by generalising the Hamiltonian (2.7) for one particle to a system with \( N \) particles. In order to do so, one must take into account the van der Waals interactions between Rydberg states, as described in chapter 1. To investigate the blockade effect caused by these interactions, consider a two-level system and use perturbation theory to obtain analytical expressions for the components of the state vector. Afterwards, we suggest a method to create a lattice of Rydberg atoms, which we apply to a system of 3 particles on a line. This method consists of using variable detunings to find suitable eigenvectors of the Hamiltonian. We end this chapter by analysing simulations of the system, in which we attempt to adiabatically follow the suitable eigenvectors.

When applying STIRAP procedures, the aim is to leave the intermediate level unpopulated at any time. If this can be accomplished, we do not have to include spontaneous emission in our simulations. Therefore, in this entire chapter, spontaneous emission is not included in the models.

3.1 The N-particle Hamiltonian in a three-level system

In order to derive the Hamiltonian for a three-level system with \( N \) particles, we first reconsider the Hamiltonian for one particle in the three-level system. In equation (2.7), this Hamiltonian is written as an operator on a three-dimensional Hilbert space, in the basis \( \{ |g\rangle , |e\rangle , |r\rangle \} \). We now write the Hamiltonian for the \( i \)th particle as follows:

\[
\hat{H}_i = -\hbar \Delta_P |e_i\rangle \langle e_i| - \hbar (\Delta_P + \Delta_S) |r_i\rangle \langle r_i| \\
+ \frac{\hbar}{2} (\Omega_P (|e_i\rangle \langle g_i| + |g_i\rangle \langle e_i|) + \Omega_S (|e_i\rangle \langle r_i| + |r_i\rangle \langle e_i|)).
\]

This one-particle Hamiltonian can now be considered as an operator on a \( 3^N \)-dimensional Hilbert space. This space describes a system of \( N \) particles in a three-level system and the operator \( \hat{H}_i \) only operates on particle \( i \), leaving the other particles untouched. In that case \( |e_i\rangle \langle e_i| \) is a projection operator which projects the state on the space where
particle \(i\) is in the \(|e\rangle\) level. The operator \(|e_i\rangle \langle g_i|\) causes a transition for particle \(i\) from the ground state to the intermediate state.

If the particles do not interact, the Hamiltonian of the three-level system with \(N\) particles can be written as a sum of \(N\) one-particle Hamiltonians. However, as mentioned before, we must include an additional term for the van der Waals interactions between atoms in the Rydberg state. Hence, the Hamiltonian can be written as

\[
\hat{H}_N = \sum_{i=1}^{N} \hat{H}_i + \hat{H}_{vdW}
\]

\[
= -\hbar \sum_{i=1}^{N} \left( \Delta_P |e_i\rangle \langle e_i| + (\Delta_P + \Delta_S) |r_i\rangle \langle r_i| \right) + \hbar \sum_{i=1}^{N} \left( \Omega_P (|g_i\rangle \langle e_i| + |e_i\rangle \langle g_i|) + \Omega_S (|e_i\rangle \langle r_i| + |r_i\rangle \langle e_i|) \right) + \sum_{i=1}^{N} \sum_{j>i}^{N} \frac{C_6}{r_{ij}^6} |r_ir_j\rangle \langle r_ir_j| \quad (3.2)
\]

where \(r_{ij}\) is the distance between particles \(i\) and \(j\). The operator \(|r_ir_j\rangle \langle r_ir_j|\) ensures that only contributions of the van der Waals interactions between atoms in the Rydberg state are added. Note that this is a Hamiltonian in a rotating frame, and that the rotating wave approximation has been used. This is explained in appendix B.1. The Hamiltonian (3.2) can also be derived immediately from equation (B.20).

If we choose the detuning \(\Delta_P\) of the intermediate state high enough, we can effectively consider a two-level system and ignore the \(|e\rangle\) level. In the next sections we will study the effect of the van der Waals interactions by considering this two-level system.

### 3.2 Two-level system with three particles on a line

In this section we will consider a two-level system, with levels \(|g\rangle\) and \(|r\rangle\), with three particles. The atoms are placed on a line and have equal distance between adjacent atoms. The system and the corresponding parameters are displayed in figure 3.1. The van der Waals interaction between the particles in the Rydberg states are given by \(V_{12} = V_{23} = C_6/r^6\) and \(V_{13} = C_6/(2r)^6 = V_{12}/64\).

For a two-level system with three particles, the Hamiltonian is given by

\[
\hat{H} = -\hbar \Delta \sum_{k=1}^{3} |r_k\rangle \langle r_k| + \frac{\hbar \Omega}{2} \sum_{k=1}^{3} (|r_k\rangle \langle g_k| + |g_k\rangle \langle r_k|) + \sum_{k<l}^{3} V_{kl} |r_kr_l\rangle \langle r_kr_l| . \quad (3.3)
\]

This Hamiltonian holds for a rotating frame and follows from the general expression (B.20), which is derived in appendix B.1. Also, the rotating wave approximation has been used. The projection operator \(|r_k\rangle \langle r_k|\) is a sum of projections operators of all states where the \(k\)th atom is in the Rydberg state.

28
Figure 3.1: A two-level system with three particles. The two levels $|g\rangle$ and $|r\rangle$ are coupled by the Rabi frequency $\Omega$. The detuning of the laser is defined as the laser’s angular frequency minus the angular frequency of the transition, so in this figure $\Delta < 0$. The distance between adjacent particles is $r$.

The following complete orthonormal basis will be used:

$$
\begin{align*}
|1\rangle &= |ggg\rangle \\
|2\rangle &= |rgg\rangle \\
|3\rangle &= |grg\rangle \\
|4\rangle &= |rrg\rangle \\
|5\rangle &= |ggr\rangle \\
|6\rangle &= |rgr\rangle \\
|7\rangle &= |grr\rangle \\
|8\rangle &= |rrr\rangle,
\end{align*}
\tag{3.4}
$$

where the short-handed notation $|ijk\rangle \equiv |i\rangle \otimes |j\rangle \otimes |k\rangle$ is used to describe the three-particle state as the tensor product of one-particle states. Using this basis, the Hamiltonian matrix elements can be calculated. This yields:

$$
\hat{H}_1 = \frac{\hbar}{2} 
\begin{pmatrix}
0 & \Omega & \Omega & 0 & 0 & \Omega & 0 & 0 & 0 \\
\Omega & -2\Delta & 0 & \Omega & 0 & \Omega & 0 & 0 & 0 \\
0 & \Omega & \Omega & \tilde{V}_{12} - 4\Delta & 0 & 0 & 0 & \Omega & 0 \\
\Omega & 0 & 0 & 0 & \Omega & 0 & 0 & \Omega & 0 \\
0 & \Omega & 0 & \Omega & \tilde{V}_{13} - 4\Delta & 0 & \Omega & 0 & 0 \\
0 & 0 & \Omega & 0 & \Omega & \tilde{V}_{23} - 4\Delta & \Omega & 0 & \Omega \\
0 & 0 & 0 & \Omega & 0 & \Omega & \Omega & \tilde{V}_{123} - 6\Delta
\end{pmatrix}
$$

where $\tilde{V}_{ij} \equiv 2V_{ij}/\hbar$ and $\tilde{V}_{123} \equiv \tilde{V}_{12} + \tilde{V}_{13} + \tilde{V}_{23}$. Applying the Schrödinger equation to the Hamiltonian yields eight coupled differential equations, which we cannot solve analytically. Therefore we will use perturbation theory to obtain analytical expressions for the state vector of the system in the next section.
3.3 Perturbation approach

In the Hamiltonian we recognise the sum of two distinct matrices. The first is a diagonal matrix which contains the energies of the different states. The second is a matrix with only off-diagonal $\Omega$ terms, which couples three-particle states that have only one atom in a different one-particle state. If we treat the second matrix as a perturbation, then the basis states \( |3.4 \rangle \) are eigenstates of the unperturbed Hamiltonian. So in the unperturbed case, the state vector can be written as

$$|\psi(t)\rangle = \sum_{k=1}^{8} C_k |k\rangle \exp\left(-\frac{iE_k t}{\hbar}\right) \quad (3.5)$$

where the coefficients $C_k \in \mathbb{C}$ are time independent. When the perturbation is added, the basis states are no longer eigenstates of the Hamiltonian. Thus when expressing the state vector as a linear combination of the basis states as in \( (3.5) \), one must use time dependent coefficients $C_k$. Applying the Schrödinger equation to \( (3.5) \) and using the orthonormality of the basis states yields eight coupled differential equations for the probability amplitudes $C_k$:

$$
\begin{align*}
\dot{C}_1 &= -\frac{i}{2} \Omega (C_2 + C_3 + C_5) e^{i\Delta t} \\
\dot{C}_2 &= -\frac{i}{2} \Omega (C_1 e^{-i\Delta t} + C_4 e^{i(\Delta - \tilde{\Omega}/2)t} + C_6 e^{i(\Delta - \tilde{\Omega}/2)t}) \\
\dot{C}_3 &= -\frac{i}{2} \Omega (C_1 e^{-i\Delta t} + C_4 e^{i(\Delta - \tilde{\Omega}/2)t} + C_7 e^{i(\Delta - \tilde{\Omega}/2)t}) \\
\dot{C}_4 &= -\frac{i}{2} \Omega (C_2 + C_3) e^{-i(\Delta - \tilde{\Omega}/2)t} + C_8 e^{i(\Delta - (\tilde{\Omega}_{13} + \tilde{\Omega}_{23})/2)t} \\
\dot{C}_5 &= -\frac{i}{2} \Omega (C_1 e^{-i\Delta t} + C_6 e^{i(\Delta - \tilde{\Omega}/2)t} + C_7 e^{i(\Delta - \tilde{\Omega}/2)t}) \\
\dot{C}_6 &= -\frac{i}{2} \Omega (C_2 + C_3) e^{-i(\Delta - \tilde{\Omega}/2)t} + C_8 e^{i(\Delta - (\tilde{\Omega}_{23} + \tilde{\Omega}_{12})/2)t} \\
\dot{C}_7 &= -\frac{i}{2} \Omega (C_3 + C_5) e^{-i(\Delta - \tilde{\Omega}/2)t} + C_8 e^{i(\Delta - (\tilde{\Omega}_{12} + \tilde{\Omega}_{13})/2)t} \\
\dot{C}_8 &= -\frac{i}{2} \Omega (C_4 + C_6) e^{i(\tilde{\Omega}_{13} + \tilde{\Omega}_{23})/2} + C_7 e^{i(\tilde{\Omega}_{12} + \tilde{\Omega}_{13})/2} e^{-i\Delta t}.
\end{align*}
\quad (3.6)$$

3.3.1 Zeroth-order perturbation

In zeroth-order perturbation theory, there is no coupling at all. Therefore, when all atoms start in the ground state, all atoms will stay in the ground state, yielding the following expression for the zeroth order coefficients:

$$C_k^{(0)}(t) = \delta_{k1}, \quad (3.7)$$

where $\delta_{k1}$ is the Kronecker delta symbol.

3.3.2 First-order perturbation

We obtain the first-order coefficients by inserting the zeroth-order coefficients into the coupled differential equations for the probability amplitudes \( (3.6) \):

$$C_k^{(1)}(t) = \begin{cases} 
0 & \text{if } k = 1, 4, 6, 7, 8 \\
-\frac{i}{2} \Omega(t) e^{-i\Delta t} & \text{if } k = 2, 3, 5,
\end{cases}$$

30
Let us presume that the laser is switched on for \( t = 0 \) with a typical strength \( \Omega_0 \): \( \Omega(t > 0) = \Omega_0 \). The reader should note that the laser intensity is proportional to the square root of this Rabi frequency: \( I \propto \sqrt{\Omega(t)} \). We can now formally integrate the system of differential equations, which yields:

\[
C_k^{(1)}(t) = \begin{cases} 
1 & \text{if } k = 1 \\
\frac{(\Omega_0/2\Delta)(e^{-i\Delta t} - 1)}{if \ k = 2, 3, 5 \wedge \Delta \neq 0} \\
-i\Omega_0 t/2 & \text{if } k = 2, 3, 5 \wedge \Delta = 0 \\
0 & \text{if } k = 4, 6, 7, 8.
\end{cases} \tag{3.8}
\]

We observe that the amplitudes of the states \(|2\rangle\), \(|3\rangle\) and \(|5\rangle\) have obtained a non-zero value in the first-order perturbation. These states represent a system in which one of the three atoms is excited to the Rydberg state. We observe that the obtained expression for these amplitudes is zero for \( t = 0 \), as it should be, and oscillates for \( t > 0 \). These oscillations are in fact Rabi oscillations, given by equation (2.6). As we are working in a perturbation approach concerning the coupling between states, the following condition holds: \( \Omega_0/\Delta \ll 1 \). The equations for \( C_{2,3,5} \) in (3.8) are linear in this term. Therefore the amplitude of the Rabi oscillations is rather small: the atoms remain predominantly in the ground state.

In figure 3.2 the value of \(|C_2|^2\) obtained by use of perturbation theory is plotted and compared to the numerically calculated solution. It is clearly visible that the perturbation theory works best when \( \Omega \) is small compared to \( \Delta \). Since we are considering a first-order perturbation, the amplitudes of \(|C_2|^2\) are correct at least up to order \((\Omega/\Delta)^2\). Naturally, this plot is the same for \(|C_3|^2\) and \(|C_5|^2\).

![Figure 3.2: Plots of the value of |C2|^2 with (a) \( \Omega = 1 \) Mrad/s and (b) \( \Omega = 4 \) Mrad/s. In both plots \( \Delta = 10 \) Mrad/s. The wave function obtained with perturbation theory is displayed in blue and the numerically calculated solution is displayed in red.](image)

Note that since there is no population of states with more than one Rydberg atom, the interaction has no influence on the result, up to order \((\Omega_0/\Delta)^2\). Therefore the numerically calculated solutions are similar to the Rabi oscillations given by equation (2.6), but in this case between the levels \(|g\rangle\) and \(|r\rangle\). Using the expressions for Rabi oscilla-
tions (2.6) and perturbation theory respectively, we find the following expressions for the probability of the state $|rgg\rangle$:

$$P_{|rgg\rangle,\text{Rabi}} = P, P_g^2 = \frac{\Omega^2}{\Omega_R} \sin^2 \left( \frac{\Omega R t}{2} \right) \left( 1 - \frac{\Omega^2}{\Omega_R} \sin^2 \left( \frac{\Omega R t}{2} \right) \right)^2,$$  (3.9)

$$P_{|rgg\rangle,\text{pert}} = \frac{\Omega^2}{\Delta^2} \sin^2 \left( \frac{\Delta t}{2} \right).$$  (3.10)

Here, $P_{|rgg\rangle,\text{pert}}$ is just the squared absolute value of $C^{(1)}_2$ in equation (3.8) and $\Omega_R \equiv \sqrt{(\Omega^2 + \Delta^2)}$ is the generalised Rabi frequency. It follows that up to order $(\Omega/\Delta)^2$, both the amplitude and the angular frequency are different: instead of $\Omega_R$, the perturbation theory yields $\Delta$. This gives a quantitative argument for the fact that if $\Omega \ll |\Delta|$, so that $\Omega_R \approx |\Delta|$, the solution from perturbation theory is indeed comparable to the analytic solution. (3.9).

From (3.8) we see that the solution which follows from perturbation theory diverges if at resonance, i.e. $\Delta = 0$. This demonstrates once again importance of the condition $\Omega_0/|\Delta| \ll 1$ for the perturbation approach.

Equation (3.8) also shows that to first order the amplitudes of states with two atoms in the excited state (i.e. $|4\rangle$, $|6\rangle$ and $|7\rangle$) are identically zero. Therefore, we cannot see the blockade effect of the van der Waals interaction. In order to study the blockade effect, we must proceed with second-order perturbation theory.

### 3.3.3 Second-order perturbation

In order to obtain the amplitudes up to second order, we plug the first-order expressions from (3.8) into the right side of the equations for the amplitudes (3.6). We distinguish two cases: $\Delta \neq 0$ and $\Delta = 0$. In the first case, the following expressions can be obtained:

$$C^{(2)}_k(t) = \begin{cases} 
1 - \frac{3\Omega_0^2}{2\Delta} (1 - e^{i\Delta t}) - \frac{3\Omega_0^2 t}{\Delta^2} & \text{if } k = 1 \\
\frac{\Omega_0^2}{2\Delta^2} (e^{-i\Delta t} - 1) & \text{if } k = 2, 3, 5 \\
\frac{\Omega_0^2}{2\Delta^2} \left( e^{-i(2\Delta - \frac{\Omega_0^2}{2}\Delta) t} - 1 \right) - \frac{1}{\Delta^2} \left( e^{-i(2\Delta - \frac{\Omega_0^2}{2}\Delta) t} - 1 \right) & \text{if } k = 4, 6, 7 \\
0 & \text{if } k = 8 
\end{cases}$$

with $\tilde{V}_{ij} = \tilde{V}_{12}$ for $k = 4$, $\tilde{V}_{13}$ for $k = 5$ and $\tilde{V}_{23}$ for $k = 7$. In the case of $\Delta = 0$ the expressions for the probability amplitudes are:

$$C^{(2)}_k(t) = \begin{cases} 
1 - \frac{3\Omega_0^2}{8} t^2 & \text{if } k = 1 \\
-\frac{\Omega_0^2 t}{8} & \text{if } k = 2, 3, 5 \\
\frac{4\Omega_0^2}{\tilde{V}_{ij} \Delta^2} \left( 1 - e^{-i\tilde{V}_{ij} t} + \frac{i}{2} \tilde{V}_{ij} t e^{i\tilde{V}_{ij} t} \right) & \text{if } k = 4, 6, 7 \\
0 & \text{if } k = 8 
\end{cases}$$

where $\tilde{V}_{ij} = \tilde{V}_{12}$ for $k = 4$, $\tilde{V}_{13}$ for $k = 5$ and $\tilde{V}_{23}$ for $k = 7$. We see that the solutions for the states with two Rydberg atoms diverge if $\Delta = 0$, $\Delta = \tilde{V}_{ij}/2$ or $\Delta = \tilde{V}_{ij}/4$. Examining the Hamiltonian’s matrix elements, it becomes clear that a necessary condition for
the use of perturbation theory is $\Omega \ll |\Delta|$ and $\Omega \ll |\hat{V}_{ij} - 4\Delta|$. These conditions explain the divergence of probability amplitudes for $\Delta = 0$ and $\Delta = \hat{V}_{ij}/4$. A value $\Delta = \hat{V}_{ij}/2$ does not necessarily imply a violation of the mentioned conditions. However, this value of $\Delta$ results in a resonant transition to the Rydberg state in the presence of another Rydberg atom.

We find that up to second order the states with two Rydberg atoms, i.e. $|4\rangle$, $|6\rangle$ and $|7\rangle$, obtain non-zero probability amplitudes. These amplitudes depend on the strength of the van der Waals interactions. As expected, if there are no interactions, then these amplitudes are equal to $C_k^2$, with $k \in \{2, 3, 5\}$.

To study the effect of the van der Waals interactions on the amplitudes $C_{4,6,7}$, we compare them to the amplitudes of two non-interacting Rydberg atoms. The latter can be calculated using (2.6). This yields for the probabilities of these states:

$$P_{|rgr\rangle,\mathrm{Rabi}} = P_g^2 P_r = \frac{\Omega^4}{\Omega_R^4} \sin^4 \left( \frac{\Omega_R t}{2} \right) \left( 1 - \frac{\Omega^2}{\Omega_R^2} \sin^2 \left( \frac{\Omega_R t}{2} \right) \right),$$  

(3.13)

and the same expression for $P_{|rrg\rangle}$ and $P_{|grr\rangle}$.

In figure 3.3, the value of $|C_6|^2$ given by equation (3.11) is plotted in blue for two different interaction strengths. Equation (3.13) is plotted in red, to emphasise the effects of the van der Waals interaction. With respect to the state $|rgr\rangle$, the van der Waals interaction

![Figure 3.3: Plot of the value of $|C_6|^2$ with $\hat{V}_{12} = 1 \text{ Mrad/s}$, $\Delta = 10 \text{ Mrad/s}$ and $\Omega = 1 \text{ Mrad/s}$. The wave function obtained with perturbation theory is displayed in blue and the analytical solution for non-interacting atoms is displayed in red.](image-url)

In figure 3.4, the value of $|C_4|^2$ given by (3.11) is plotted in blue for two values of the interaction strength. Equation (3.13) is plotted in red, to emphasise the effects of the van der Waals interaction. With respect to the state $|rgr\rangle$, the van der Waals interaction

![Figure 3.4: Plot of the value of $|C_4|^2$ with $\hat{V}_{12} = 1 \text{ Mrad/s}$, $\Delta = 10 \text{ Mrad/s}$ and $\Omega = 1 \text{ Mrad/s}$. The wave function obtained with perturbation theory is displayed in blue and the analytical solution for non-interacting atoms is displayed in red.](image-url)
is now stronger by a factor 64. It can be seen that the van der Waals interaction greatly influences the probability of the state $|rrg\rangle$. Also, comparing both figures in 3.4, it follows that higher maximum probabilities are obtained when the interaction increases. This is due to the choice of a positive detuning. By looking at equation (3.11), one can see that the probabilities of obtaining two Rydberg atoms increase when the detuning is chosen closer to the value $\tilde{V}_{ij}/2 = V_{ij}/\hbar$. Since we are considering the case where three atoms are placed on a line and $r_{12} = r_{23}$, the graph for $|C_7|^2$ is also represented by figure 3.4.

![Figure 3.4: Plots of the value of $|C_4|^2$, corresponding to state $|rrg\rangle$, with (a) $\tilde{V}_{12} = 1$ Mrad/s and (b) $\tilde{V}_{12} = 2$ Mrad/s. In both plots $\Delta = 10$ Mrad/s and $\Omega = 1$ Mrad/s. The wave function obtained with perturbation theory is displayed in blue and the analytical solution for non-interacting atoms is displayed in red.](image)

By comparing figures 3.3 and 3.4 one can see that the van der Waals interactions have a big influence on the probability amplitudes of states with adjacent Rydberg atoms. It also follows that the blockade effect caused by the van der Waals interactions can be countered by using suitable detunings. Using this knowledge, we will propose a scheme to create a lattice of Rydberg atoms in the next section.

### 3.4 Eigenvectors of the three-particle Hamiltonian

The method that we now propose to create a lattice of Rydberg atoms consists of several STIRAP schemes. Recall from chapter 2 that a STIRAP scheme can bring a particle from the ground state to the Rydberg state, without ever populating the intermediate state. In the previous section, we found that the van der Waals interactions have great effect on nearest-neighbours atoms. Therefore, when applying one STIRAP scheme, we expect that we can never excite two adjacent atoms at the same time. However, an atom in the ground state that has a nearest neighbour in the Rydberg state, can be excited by applying another STIRAP scheme with a total detuning $\hbar\Delta = C_6/r_6^6$. This detuning opposes the effect of the van der Waals interaction, causing a resonant transition. Thus, a method to create a lattice of Rydberg atoms could be as follows. Applying the first
STIRAP scheme, we excite a random set of atoms, but never two adjacent atoms. Then, another STIRAP scheme with a detuning $\hbar \Delta = C_6/r^6$ is applied, exciting only atoms with one nearest-neighbour atom in the Rydberg state. This is followed by another scheme, exciting atoms with two nearest-neighbour Rydberg atoms. By continuing this process, we wish to create a Rydberg crystal. In a hexagonal lattice this would require seven schemes, for each atom has six nearest neighbours. In figure 3.5 we illustrate this method for a small array of atoms.

Let us now return to the system where three atoms are placed on a line, with an equal distance $r = 5 \mu$m between subsequent atoms. The initial state consists of three atoms in the ground state: $|ggg\rangle$. The first STIRAP scheme should then cause a transition to either $|grg\rangle$ or $|rgr\rangle$. In case $|grg\rangle$ is obtained, a STIRAP scheme with a total detuning $\hbar \Delta = C_6/r^6$ should cause a transition to $|rrr\rangle$. In case $|rgr\rangle$ is obtained, a scheme with total detuning $\hbar \Delta = 2C_6/r^6$ should cause a transition to $|rrr\rangle$.

In chapter 2 we described that one particle could be transferred from the ground state to the Rydberg state, with a probability of 1 and without ever acquiring any probability of being in the intermediate level. This was done by adiabatically following the required eigenvector. We will now attempt to excite the three particles on a line by using the same approach. Thus, we start by looking for suitable eigenvectors of the Hamiltonian (3.2) of the three-level system with three particles. All eigenvectors are calculated with the script in appendix C.6. If the particles do not interact, a suitable eigenvector is $|a^0\rangle \otimes |a^0\rangle \otimes |a^0\rangle$. As shown in figure 3.6 this would bring all particles to the Rydberg state without populating the intermediate level at any time.
When interactions are included, we do not expect to excite adjacent atoms when applying one STIRAP scheme. As mentioned above, the first scheme should yield either $|rgr\rangle$ or $|grg\rangle$. Thus, we wish to find an eigenvector of the Hamiltonian (3.2), which causes a population transfer from $|ggg\rangle$ to either one of those states. Moreover, to prevent spontaneous emission the eigenvector must never allow population of the $|e\rangle$ level.

We will use the following pulse scheme:

$$\begin{align*}
\Omega_P(t) &= \Omega_{P,0} \exp\left(-\frac{t^2}{2\sigma^2}\right) \\
\Omega_S(t) &= \Omega_{S,0} \exp\left(-\frac{(t-t_{diff})^2}{2\sigma^2}\right)
\end{align*}$$

(3.14)

with $\sigma = 1$ a.u. and $t_{diff} = 2$ a.u. Note that since we are considering the instantaneous eigenvectors of $H(t)$, the timescale is irrelevant here. However, the timescale will be of relevance later, when the suitable eigenvectors are to be followed adiabatically. The eigenvectors are now defined by the parameters $\Delta_P$, $\Delta_S$, $\Omega_{P,0}$, $\Omega_{S,0}$ and $r$. The total detuning $\Delta_P + \Delta_S$ should be chosen such that a resonant transition occurs. Furthermore, since the eigenvectors do not change when the Hamiltonian is multiplied by a constant, we can eliminate another parameter. We use this degree of freedom to choose $r = 5 \mu m$. However, note that if we had chosen the interaction to be twice as strong, we would have obtained the same eigenvectors by also choosing the other parameters to be twice as large. The eigenvectors are therefore defined by the three parameters $\Omega_P$, $\Omega_S$ and $\Delta_P$.

Using $\Omega_P = \Omega_S = -\Delta_P = 10$ Mrad/s, we can obtain the eigenvector displayed in figure 3.7. This eigenvector is suitable insofar as it causes a population transfer from the ground state $|ggg\rangle$ to the state $|rgr\rangle$. However, the eigenvector contains a component which allows population of the intermediate state. As this state has a high spontaneous emission rate, it will probably be hard to follow. We will show this in chapter 4. Hence,
this eigenvector is not adequate.

Figure 3.7: An eigenvector of the Hamiltonian of a three-level system, with three interacting particles. Here, $\Delta S + \Delta P = 0$, $\Omega_S = \Omega_P = 10 \text{ Mrad/s}$, $\Delta P = -1 \text{ Mrad/s}$, $r = 5 \mu m$.

Apparently, the van der Waals interactions cause the eigenvector to acquire components containing the intermediate level $|e\rangle$. Therefore, the eigenvector is impossible to follow and one could conclude that applying STIRAP schemes to create Rydberg crystals is impossible. It is however possible to suppress the population of the intermediate state by increasing the detuning of the intermediate state and the coupling of the levels $|e\rangle$ and $|r\rangle$. Intuitively this is clear: from Rabi oscillations between two levels we know that an increased detuning yields a decreased probability of being in the excited state, whilst an increased Rabi frequency yields a higher probability.

Figure 3.8: An eigenvector of the Hamiltonian for a three-level system with three atoms. In both figures, $r = 5 \mu m$, $\Omega_S = 5 \Omega_P$ and $\Delta P = -10\Omega_P$. Furthermore, in (a) $\Omega_P = 10 \text{ Mrad/s}$, in (b) $\Omega_P = 100 \text{ Mrad/s}$. The other visible components are states with one Rydberg atom and two ground-state atoms.

In figure 3.8 (a), the Rabi frequency $\Omega_S$ is increased by a factor 5 and the detuning $\Delta P$ is increased by a factor 10, with respect to figure 3.7. This eigenvector seems to be suitable: it causes a population transfer from $|ggg\rangle$ to $|rgr\rangle$ and the population of the intermediate state is negligibly small. However, as we have seen in chapter 2, a larger
detuning $\Delta_P$ has a negative effect on the adiabaticity of the process. It appears that
to follow the eigenvector adiabatically, very large timescales are required. This fact is
displayed in figure 3.9, where we have used the script in appendix C.4 for the simulation.
In order to follow the eigenvector adiabatically, we require timescales of the order
of a millisecond. The spontaneous emission rate of the Rydberg state is then no longer
negligible, neither can we expect the frozen gas limit to hold.

Figure 3.9: Simulations of the three-level system with three atoms. The used parameters are the
same as in figure 3.8 (a). In (a), the state vector is not following the eigenvector from figure 3.8 (a)
adiabatically. Here we used the pulses from equation (2.14), with $a_P = 0$, $a_S = 10\mu s$ and $\sigma_P = \sigma_S = 5\mu s$.
In (b), the eigenvector is followed adiabatically. The timescale used is 20 times as slow as in (a).

In chapter 2 we found that increasing the Rabi frequencies yields better adiabaticity.
Therefore, in figure 3.8 (b), we increased both Rabi frequencies by a factor 10 with re-
spect to figure 3.8 (a). The detuning $\Delta_P$ is increased by a factor 10 as well, for otherwise
it fails to suppress population of the intermediate state. In figure 3.10 it follows that we
are able to follow the eigenvector adiabatically using reasonable timescales. However, the used Rabi frequencies greatly transcend the experimentally obtainable values, which were discussed in chapter 2.

\[
\begin{align*}
&[g,r,g] \\
&[r,g,g], [g,g,r]
\end{align*}
\]

Figure 3.10: Simulations of the three-level system with three atoms. The used parameters are the same as in figure 3.8 (b). Here we used the pulses from equation (2.14), with \( a_P = 0 \), \( a_S = 10 \mu s \) and \( \sigma_P = \sigma_S = 5 \mu s \). The eigenvector from figure 3.8 (b) is followed adiabatically.

Suppose we succeed at causing the population transfer to \(|rgr\rangle\). We then apply a second STIRAP scheme to bring the middle atom to the Rydberg state. A total detuning \( \hbar(\Delta_P + \Delta_S) = 2C_6/r^6 \) is required to account for the two nearest-neighbour atoms in the Rydberg state. Using this detuning, we can find the eigenvector displayed in figure 3.11 (a). This eigenvector contains no components with the intermediate level \(|e\rangle\) and is therefore a suitable eigenvector. It appears that this eigenvector can easily be followed adiabatically, as is shown in figure 3.11 (b). Note that the middle atom is the only atom which has two nearest neighbours. Therefore, by choosing the total detuning to account for two nearest-neighbour interactions, the process becomes basically equivalent to a STIRAP procedure for one particle.
Figure 3.11: (a) The eigenvector of the second STIRAP scheme. (b) The simulation of the second STIRAP scheme. The used parameters are $\Omega_P = \Omega_S = 10 \text{ Mrad/s}$, $\hbar(\Delta_P + \Delta_S) = C_6/r_6$ with $r = 5 \mu\text{m}$ and $\Delta_P = 0$. In the simulation in (b) we used the pulses from equation (2.14), with $\sigma_P = 0$, $\sigma_S = 10 \mu\text{s}$ and $\sigma_P = \sigma_S = 5 \mu\text{s}$. Clearly, the eigenvector from (a) is being followed adiabatically in (b).
3.4.1 Adiabaticity criterion

From the previous discussion, it follows that the eigenvector in figure 3.8 (a) requires much larger timescales to follow adiabatically than figure 3.8 (b). We can understand this by looking at the adiabaticity criterion for the case of one particle, which is given by (2.15). We now assume that a similar adiabaticity condition holds for the three-level system with three particles. For this system, the adiabaticity criterion can then be written as follows:

\[ \left| \left\langle \dot{a}^k | a^i \right\rangle \right| \ll \left| \omega^i - \omega^k \right| \quad \forall i \in \{1, 2, ..., 27\} \setminus \{k\} \]  (3.15)

where \( |a^i \rangle \) are the eigenvectors of the Hamiltonian with corresponding eigenfrequencies \( \omega^i \) and \( |\dot{a}^k \rangle \) is the time derivative of the eigenvector that we wish to follow adiabatically, with \( k \in \{1, 2, ..., 27\} \). Note that this criterion can be evaluated at any time during the STIRAP procedure. However, the criterion is most important when the population transfer takes place and \( |\dot{a}^k \rangle \) is large.

Since we do not know analytical expressions for the eigenvectors and eigenvalues, we cannot derive an adiabaticity criterion like equation (2.18). However, upon inspection of the eigenvectors in figure 3.8 one notices that for the left eigenvector, the population transfer occurs faster than for the right eigenvector, if the same timescales are used. This corresponds to a larger time-derivative for the eigenvector in figure 3.8 (a) than for the eigenvector in figure 3.8 (b).

Furthermore, we can also consider the eigenvalues of corresponding to the eigenvectors in figure 3.8. These eigenvalues are displayed in red in figure 3.12 together with the eigenvalues of all other eigenvectors in blue. Notice the different scalings of the vertical axes. By inspecting figure 3.12 (b) and (d), one can clearly see that for the parameters used in figure 3.8 (a), the quantities \( |\omega^i - \omega^k| \) are smaller than for the parameters used in figure 3.8 (b).

From these two facts, it follows that in order to obey the condition (3.15), the eigenvector in figure 3.8 (a) requires much larger timescales than the eigenvector in figure 3.8 (b). This is exactly what we found in the previous discussion.

We have seen that the eigenvector in figure 3.8 (b) can be followed adiabatically by using reasonable timescales, but the required Rabi frequencies are far greater than experimentally achievable. One could try to choose another value for \( r \), say \( r = 10 \mu \text{m} \). This decreases the van der Waals interactions by a factor 64. If we now divide all Rabi frequencies and detunings by 64 as well, the complete Hamiltonian is now divided by 64 with respect to the original case. From the standard expression for an eigenvalue problem, it follows that

\[ A x = \lambda x \iff (cA) x = (c \lambda) x, \quad c \in \mathbb{C}. \]  (3.16)

Therefore, the eigenvectors of the Hamiltonian have not changed after division by 64, but all the eigenvalues have decreased by a factor 64 as well. Hence, in the adiabaticity criterion (3.15), the left-hand side does not change, but the right-hand side decreases by a factor 64. It has thus become harder to follow the eigenvector adiabatically. To be
Figure 3.12: Eigenvalues of the Hamiltonian. For all figures, $r = 5 \mu m$, $\Omega_s = 5\Omega_P$ and $\Delta_P = -10\Omega_P$. The eigenvalues corresponding to the eigenvectors from figure 3.8 are plotted in red. (a) $\Omega_P = 10$ Mrad/s. (b) Zoomed view of (a). (c) $\Omega_P = 100$ Mrad/s. (d) Zoomed view of (c).

more precise, making the timescale 64 times as large should yield the same adiabaticity. This decreases the time derivative of the eigenvector by a factor 64, restoring the ratio of the left-hand side and the right-hand side of the condition (3.15).

3.5 Eigenvectors for other systems

In the previous sections we mainly focussed on a system which consists of 3 particles on a line. In this section we will shortly consider several other systems, using the same method as described in the previous section. In all cases, we first find a possible sequence of STIRAP schemes, then look for appropriate eigenvectors. We shall not broadly discuss adiabaticity here. This section will be ended with a general discussion concerning the possibility of creating Rydberg crystals.

3.5.1 Two particles

Using the method as described in the previous section, we need two STIRAP schemes to excite both atoms. It appears that, similar to the case with three particles, the van der Waals interactions induce components of the intermediate state. This eigenvector is visible in figure 3.13(a). This is also what Møller, Madsen and Mølmer found [23]. As
with the three-particle case, the population of the intermediate state can be suppressed by increasing the detuning of the intermediate state. This is visible in figure 3.13 (b). The required detuning to suppress the population of the $|e\rangle$ level is lower than for the three-particle system. We therefore expect adiabatic following of this eigenvector to be easier than for the eigenvector for the three-particle case, in figure 3.8 (a). Because of the symmetry of the system, a symmetric superposition of the states $|rg\rangle$ and $|gr\rangle$ is obtained. After the first STIRAP scheme, a second one with a total detuning $\hbar(\Delta_P + \Delta_S) = C_6/r^6$ can cause a transition to the $|rr\rangle$ state. This is visible in figure 3.13 (c).

![Graphs showing the eigenvectors of the three-level Hamiltonian with two particles.](image)

Figure 3.13: Eigenvectors of the three-level Hamiltonian with two particles. The other used parameters are $\Omega_P = \Omega_S = 10$ Mrad/s and $r = 5\mu$m in all plots. The eigenvector (a) is not suitable because of the population of the level $|e\rangle$. The eigenvector in (b) is suitable for the first STIRAP scheme, the eigenvector (c) is suitable for the second scheme.

### 3.5.2 Four particles on a line

The case of four particles on a line, with a distance $r$ between subsequent atoms, is very similar to the case with two or three particles on a line. Using a detuning $\Delta_P$ large enough to suppress population of the intermediate state, the first STIRAP scheme causes a transition to $|rggr\rangle$. The second STIRAP scheme again needs a detuning $\Delta_P$ large enough to suppress population of the $|e\rangle$ level. Furthermore, a total detuning $\hbar(\Delta_S + \Delta_S) = C_6/r^6$ is needed to account for one nearest-neighbour interaction, if we ignore the next-nearest neighbours. A transition is then caused to a state which is a superposition of the states $|rgrr\rangle$ and $|rrgr\rangle$. For the third STIRAP scheme, a total...
detuning $h(\Delta S + \Delta_S) = 2C_6/r^6$ is applied to cause a transition to $|rrrr\rangle$. The three eigenvectors which result from these schemes are displayed in figure 3.14 including the used parameters. Note that these eigenvectors may be hard to follow adiabatically.

Figure 3.14: Suitable eigenvectors of the three-level Hamiltonian with four particles. The used parameters are $\Omega_P = \Omega_S = 10$ Mrad/s and $r = 5\mu$m in all plots. Furthermore, in (a) $\Delta_P = -10\Omega_P, \Delta_S = 0$. In (b) $\Delta_P = -10\Omega_P, \Delta_S = -\Delta_P + C_6/r^6$. In (c) $\Delta_P = 0, \Delta_S = 2C_6/r^6$.

### 3.5.3 Three particles in a triangle configuration

Consider a system of three particles, where there is a distance $r$ between any pair of particles. The system is displayed in figure 3.15. This is, in contrast to the previous situations, a two-dimensional configuration. However, the used method is the same. Using the appropriate total detuning for each STIRAP scheme, the first scheme yields a
superposition of all states with one Rydberg atom, the second yields a superposition of all states with two Rydberg atoms and the third yields the state $|rrr\rangle$. This is displayed in figure 3.16. For the first two schemes, a large detuning $\Delta_P$ was used to suppress population of the intermediate state.

![First STIRAP scheme](image1)

![Second STIRAP scheme](image2)

![Third STIRAP scheme](image3)

Figure 3.16: Suitable eigenvectors of the three-level Hamiltonian with three particles in a triangle configuration. The used parameters are $\Omega_P = \Omega_S = 10$ Mrad/s and $r = 5 \mu m$ in all plots. Furthermore, in (a) $\Delta_P = -10\Omega_P$, $\Delta_S = 0$. In (b) $\Delta_P = -10\Omega_P$, $\Delta_S = -\Delta_P + \frac{C_6}{r^6}$. In (c) $\Delta_P = 0$, $\Delta_S = 2\theta_6/r^6$.

### 3.5.4 General systems

From the configurations we have considered, one can draw several conclusions.

Firstly, the proposed method to create Rydberg crystals seems to yield some satisfying results. Although adiabatic following sometimes seems to require impractically high laser intensities, the suitable eigenvectors do appear to exist. However, it is important that we know the specific outcome of the applied STIRAP schemes. Consider for example the case of three particles on a line. If the standard method as described in the beginning of this section were applied, we would use three STIRAP schemes, where the second and third scheme account for one and two nearest-neighbour interactions respectively. The first scheme would excite atoms which are not nearest neighbours. If this yields the state $|rr\rangle$, the second scheme would do nothing and the third would yield $|rrr\rangle$, as required. However, what if the first scheme yielded the $|grg\rangle$ state? The second scheme would then cause a transition to $|rrr\rangle$. However, the third pulse which
accounts for two nearest-neighbour interactions, would then cause a transition back to $|rgr\rangle$. This means that the method has failed, since it should yield $|rrr\rangle$ as the final result. It is therefore of utmost importance to know in advance which state we acquire after the first scheme. The eigenvector we found show that a transition to $|rgr\rangle$ occurs and not to $|grg\rangle$, so in this case, this is not a problem.

For more complex lattices, it could prove difficult to calculate the eigenvectors of the system in advance. One could think of a lattice in which the proposed method does not work. Consider for example a hexagonal configuration of 7 atoms, displayed in figure 3.17. Since the first STIRAP scheme will not excite two adjacent atoms, a possible outcome could be to excite the middle atom. Suppose the second scheme then yields the result from figure 3.17. The next scheme could then excite two non-adjacent ground-state atoms. However, since the middle atom has two nearest-neighbour atoms in the Rydberg state, the scheme could also bring the middle atom back to the ground state. If this happens, the sequence of STIRAP schemes has failed. From the case of three particles on a line, it is known that the first scheme does not cause an excitation of the middle atom. If this holds for the hexagonal configuration, the problem from figure 3.17 does not occur. It would be interesting to see whether the eigenvectors for more general complex systems can always prevent this problem.

Another conclusion, which we can draw from for example the case with two particles, is that the first scheme yields a superposition of states. This is of course obvious if one considers the symmetry of the configuration. The important thing to note here, in the case of two particles, is that it is not true that the first STIRAP scheme yields a transition to either $|rg\rangle$ or $|gr\rangle$. The first scheme yields a transition to a state which is a superposition of $|rg\rangle$ and $|gr\rangle$. The relevant eigenvector for the second pulse scheme then starts in the superposition of states and causes a transition to $|rr\rangle$. However, no eigenvectors exist which cause a transition from the state $|rg\rangle$ or $|gr\rangle$ to $|rr\rangle$. Consequently, running a simulation which starts in $|rg\rangle$ does not yield a transition to $|rr\rangle$. An interesting topic of further research would be the prediction of the eigenvectors by exploiting the high symmetries of lattices. For example, do all obtainable components of a suitable eigenvectors only differ from each other by one symmetry operation? In the cases we have considered, this is true. For instance, $|rg\rangle$ and $|gr\rangle$ differ by a rotation of $\pi$ around the center of mass. Moreover, in the case of three particles, we obtained $|rgr\rangle$ but not $|grg\rangle$. It would be interesting to investigate this further.

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Figure 3.17: Possible outcome of a sequence of STIRAP schemes on a hexagonal configuration. The blue atoms are in the ground state, the red atoms are in the Rydberg state.
3.6 Summary

In this chapter we started by generalising the Hamiltonian of a three-level system with one particle to the case with \( N \) particles. We saw that the main difference with the one-particle Hamiltonian is the additional term describing the van der Waals interactions. To study the effect of these interactions, we used perturbation methods on a two-level system containing three particles on a line. We then proposed a method to create a lattice of Rydberg atoms using multiple STIRAP schemes. This method consisted of looking for suitable eigenvectors of the Hamiltonian and following them adiabatically. Subsequently we applied this method on a configuration of three atoms on a line. We then showed that suitable eigenvectors for the STIRAP schemes do exist, but can be hard to follow adiabatically in practice. Lastly, we concluded by discussing several other systems and a discussion of creating Rydberg crystals with general lattices.

One of the important conclusions of this chapter is the fact that the van der Waals interactions induce components of the eigenvectors which contain the intermediate state. It is therefore important that we take spontaneous emission into account in future simulations. In the next chapter, we will discuss a method in which this can be done.
Chapter 4

Multi-particle STIRAP with spontaneous emission

In this chapter we reconsider the three-level system from figure 2.4. In this three-level system we have a ground state $|g\rangle$, an excited state $|e\rangle$ and a Rydberg state $|r\rangle$. As mentioned in section 1.2, the lifetime of a Rydberg state is proportional to $n^3$, which means that the rate of spontaneous emission from the relevant Rydberg state is of the order of several hundreds of $\mu$s. It can therefore be neglected on the scale of our excitation dynamics. However, we cannot neglect spontaneous emission from the excited state $|e\rangle$. Firstly this is the case because we have an experimental spontaneous emission rate for $|e\rangle$ of 6 Mrad/s, or a spontaneous life time of about 1 $\mu$s. Secondly, we showed that the eigenvectors for many-particle systems obtain components in the $|e\rangle$-state, so that spontaneous emission from this state cannot be ignored.

In this chapter, we therefore seek a way to include spontaneous emission from $|e\rangle$ in our description of the system. We start with a thorough derivation of a general expression for the spontaneous emission rate from an excited state of an atom, in the Wigner-Weisskopf approximation. We compare the theoretical spontaneous emission rate for a Rb-85 system with the experimental value. We proceed with a review of density operator theory. By implementing a superoperator, we arrive at a master equation for the system. However, the computation time to solve the master equation is too large. We therefore consider the stochastic quantum Monte Carlo algorithm and use it to perform some simulations.

4.1 Rate of spontaneous emission

In 1917 Einstein postulated spontaneous emission. He considered a given number of atoms in a cavity and was calculating the evolution of the atomic population under the influence of a constant light source shining on the system. In order to reach thermal equilibrium in such a system, he had to postulate spontaneous emission. Up until that moment, only stimulated emission and absorption were known from experiments. Einstein also deduced the rate of spontaneous emission. His answer is correct to this day, but the derivation does not show the true nature of the spontaneous emission process.

After the quantization of the electromagnetic field by Paul Dirac in 1927, it was realized
that the fluctuations of the quantized vacuum field cause excited atoms to spontaneously emit a photon. We shall now describe the more correct Wigner-Weisskopf theory.

Consider a two-level atom, with ground state $|g\rangle$ and excited state $|e\rangle$, placed in a cavity of volume $V$. The atom is in its excited state for $t = 0$. We assume there to be no photons in the radiation field of the cavity. The system is therefore in the state $|\Psi(t = 0)\rangle = |e\rangle \otimes |\{0\}\rangle \equiv |e, \{0\}\rangle$, where we defined the multi-mode vacuum state by: $|\{0\}\rangle = |0, 0, \ldots, 0, \ldots\rangle$. Intuitively, we know that as time progresses, the probability of spontaneous emission shall increase. When an atom spontaneously decays, it emits a photon into the radiation field. This photon is emitted into one of the infinite number of vacuum modes $j$. One such mode corresponds to both a specific direction in which the photon travels, denoted with the wavevector $\vec{k}$, and the polarization of the photon, denoted with $\epsilon$. At a time $t$ the state vector of the system shall therefore be given by:

$$|\Psi(t)\rangle = C_e(t) |e, \{0\}\rangle + \sum_j C_{g,j}(t) |g, \{j\}\rangle,$$

in which the sum runs over all possible decay modes $j = (\vec{k}, \epsilon)$. Furthermore we have defined $|\{j\}\rangle \equiv |0_1, 0_2, \ldots, 0_{j-1}, 1_j, 0_{j+1}, \ldots\rangle$. The reader should also note that we have absorbed the time-dependent complex exponentials including the energies of the states in the expansion coefficients in equation (4.1).

We can now calculate the time evolution of the probability amplitudes by inserting the state vector into the time-dependent Schrödinger equation. As Hamiltonian we use the two-level Hamiltonian from equation (2.1). We note that we do not need the precise expression for the free-atom and free-field Hamiltonians, because only the interaction Hamiltonian couples the two states and the vacuum field. This procedure yields the following expressions:

$$\begin{cases}
\dot{C}_e(t) &= -\sum_j \Omega_j^* C_{g,j} e^{i\Delta_j t} \\
\dot{C}_{g,j}(t) &= \Omega_j C_e e^{-i\Delta_j t},
\end{cases}$$

where we defined the single mode Rabi frequency $\Omega_j \equiv d \cdot E_j / \hbar$ and $\Delta_j = \omega_{eg} - \omega_j$ is the detuning for every mode $j$.

Noting that the atom starts in the excited state, i.e. $C_e(t = 0) = 1$ and $C_{g,j}(t = 0) = 0$, formally integrating the equation for $\dot{C}_{g,j}(t)$ and plugging this into the equation for $\dot{C}_e(t)$ yields:

$$\dot{C}_e(t) = -\sum_j |\Omega_j|^2 \int_0^t C_e(\tau) e^{i\Delta_j(t-\tau)} d\tau.$$  \hspace{1cm} (4.3)

From this expression we immediately see an exponential decay for $\dot{C}_e(t)$, as expected. If we want to say something about the decay rate of the exponential we need to evaluate the sum in equation (4.3). In a different context, it is shown in [6] how a sum over all vacuum modes can be transferred into an integral over all frequencies $\omega$ by considering the quantized electromagnetic field inside a cavity with volume $V$. Using this, we get the following expression:

$$\dot{C}_e(t) = -\frac{d^2}{6\pi^2 c_0 e^2 \hbar} \int_0^\infty \omega^3 d\omega \int_0^t C_e(\tau) e^{i\Delta(\omega)(t-\tau)} d\tau,$$ \hspace{1cm} (4.4)
where $d_{ge} \equiv \langle g | d | e \rangle$. In order to evaluate the integrals that are left, we assume that the dipole moment slowly changes as a function of the frequency $\omega$, so that evaluation at $\omega = \omega_{ge}$ is justified. This approximation is called the Wigner-Weisskopf approximation and it yields:

$$
\int_{0}^{\infty} \omega^3 d\omega \int_{0}^{t} e^{i\Delta(t-\tau)} d\tau \approx \omega_{eg}^3 \int_{-\infty}^{\infty} d\omega e^{i(\omega_{eg}-\omega)(t-\tau)} = 2\pi \omega_{eg}^3 \delta(t-\tau).
$$

Plugging this into equation (4.4) finally yields:

$$
d_{ge}(t) \frac{dt}{d\omega} = -\frac{\gamma}{2} c_{e}(t) \text{ with } \gamma = \frac{\omega_{eg}^3 d_{eg}^2}{3\pi \epsilon_0 \hbar^3},
$$

where $\gamma$ is called the spontaneous emission rate. Note that the amplitude of the excited states decays at a rate $\gamma/2$, so that the population (which scales as the square of the absolute value of the amplitude) decays with $\gamma$. The spontaneous emission life time is therefore $\tau \equiv 1/\gamma$.

We can now use the expression for $\gamma$ to calculate a theoretical value for the spontaneous emission rate for the system we described in section 2.3. The relevant constants and experimental parameters are the following:

- Transition wavelength: $\lambda_{eg} = 780$ nm.
- Dipole moment transition: $d_{eg} = 2.9 \cdot 10^{-12}$ C-m
- Elementary electron charge: $e = 1.602 \cdot 10^{-19}$ C
- Bohr radius: $a_0 = 5.29 \cdot 10^{-11}$ m
- Reduced Planck’s constant: $\hbar = 1.05 \cdot 10^{-34}$ J-s
- Speed of light: $2.998 \cdot 10^8$ m/s
- Vacuum permittivity: $\epsilon_0 = 8.854 \cdot 10^{-12}$ F/m.

Using these values yields the following number for the rate of spontaneous emission: $\gamma \approx 36$ Mrad/s = 5.73 MHz. This is a theoretically calculated value. Experimentally, a value of 5.98 MHz is observed [20], which yields a relative error of 4.4%. This small deviation is most probably a result of the Wigner-Weisskopf approximation. Now that we know how spontaneous emission emerges and at what rate it takes place for a Rb-85 system, we are ready to look at an implementation in the simulations. Firstly, we shall consider the theory of density operators.

### 4.2 Review of density operator theory

Suppose we would like to describe a set of two-level atoms, which all start in the excited state. After a certain time $t$, there is a certain probability for every atom that it has spontaneously decayed. Unfortunately, this yields a big problem. When spontaneous emission occurs, the photon is emitted into a random vacuum mode, as described earlier. When considering a set of atoms and the surrounding vacuum, one therefore has to
include an infinite number of vacuum modes in the description. This greatly complicates
the wavefunction formalism.

A way to overcome this problem is to use the density operator formalism. This for-
malism is a generalization of the wavefunction formalism to include the possibility of
uncertainty in its preparation. Suppose that we do not know the state a system is pre-
pared in. However, we do know that the system is in state from an ensemble of states
\{ | \Psi_n \rangle \} with corresponding probability \{ p_n \}. The density operator \( \hat{\rho} \) is then defined as:

\[
\hat{\rho}(t) = \sum_n p_n |\Psi_n(t)\rangle \langle \Psi_n(t)| .
\] (4.7)

We immediately note that the density operator is Hermitian: \( \hat{\rho} = \hat{\rho}^\dagger \), so that it has real
eigenvectors. Furthermore, the density operator can be represented as a matrix for a
given set of complete orthonormal basis vectors, say \{ \phi_i \}. The elements of the density
matrix then become:

\[
\rho_{ij} = \langle \phi_i | \hat{\rho} | \phi_j \rangle .
\] (4.8)

The eigenvalues of the density matrix are just the probabilities \( p_n \), which sum to one.
The trace of the density matrix is therefore always one:

\[
\text{Tr}[\hat{\rho}] = \sum_i \langle \phi_i | \hat{\rho} | \phi_i \rangle = \sum_n p_n \sum_i |\langle \phi_i | \Psi_n \rangle|^2 = \sum_n p_n = 1.
\] (4.9)

If and only if the system is in a pure state, i.e. we know that it is prepared in one
state \( \Psi \), the density operator is equal to its square: \( \hat{\rho}^2 = \hat{\rho} \). This gives us a method to
check whether or not a system is in a statistical mixture of states or just in one pure state.

Just as in the wave function formalism we can use the density operator to calculate
the expectation value for any operator \( \hat{A} \):

\[
\langle \hat{A} \rangle = \sum_n p_n \langle \Psi_n | \hat{A} | \Psi_n \rangle
= \sum_{i,j} \sum_n \langle \Psi_n | \phi_i \rangle \langle \phi_i | \hat{A} | \phi_j \rangle \langle \phi_j | \Psi_n \rangle
= \sum_{i,j} \hat{A}_{ij} \rho_{ji}
= \text{Tr}[\hat{A} \hat{\rho}],
\] (4.10)

which just constitutes of writing the density matrix and the operator in their matrix
representation, multiplying the matrices and calculating the trace of the product. We
note that the diagonal element \( \hat{\rho}_{ii} \) is the probability of finding the system in the state
\( | \phi_i \rangle \), whereas the off-diagonal elements \( \hat{\rho}_{ij} \) are called the coherences between the states
| \phi_i \rangle and | \phi_j \rangle. They are called this way because they depend on the phase difference
between the two states.

Finally, we can derive a differential equation for the time-dependence of the density

51
operator:
\[
\frac{d\hat{\rho}}{dt} = \sum_n \left( \frac{d}{dt} |\Psi_n(t)\rangle \langle \Psi_n(t)| + |\Psi_n(t)\rangle \langle \Psi_n(t)| \frac{d}{dt} \right)
\]
(4.11) = \frac{1}{i\hbar} \sum_n p_n \hat{H} |\Psi_n(t)\rangle \langle \Psi_n(t)| - \frac{1}{i\hbar} \sum_n p_n |\Psi_n(t)\rangle \langle \Psi_n(t)| \hat{H}
= \frac{1}{i\hbar} [\hat{H}, \hat{\rho}],
which is called the quantum Liouville equation, because of the analogy with the Liouville equation in theoretical classical mechanics. In this compact discussion we used elements from [24]. The reader is referred to this book for further details on density operator theory.

4.3 Using density operator theory to describe spontaneous emission

Now we have reviewed basic density operator theory, we are almost ready to use it to describe spontaneous emission. There is only one point of attention left.

Spontaneous emission is a so-called dissipative quantum process. It is called this way because the energy of the atom is not conserved when spontaneous emission occurs: the potential energy of the electron in an excited state is converted into a photon, which is emitted randomly into space. The atom returns to its ground state after emitting the photon and has thus lost its energy to the environment, i.e. the photon in the vacuum. Therefore, we can only describe the atom and its environment together as a system with a Hamiltonian, so that the total energy is conserved.

One way to overcome these problems is to split the total system in two parts: on one hand the atom - or more general a quantum system in which dissipation can occur - and on the other hand the bath or reservoir to which the energy of the quantum system shall flow. Because of the flow of energy there is a coupling between the two subsystems. In order to have a flow only from the quantum system to the bath, i.e. a irreversible flow of energy, the bath has to contain an infinite number of degrees of freedom, as we know from the second law of thermodynamics. Obviously, this is the case with the infinite number of vacuum modes. The crux of this description is to calculate the coupling between quantum system and bath correctly [25].

We can include this coupling correctly by simply adding a phenomenological term - in contrast to the tedious discussion in [26] - to the quantum Liouville equation:
\[
\frac{\partial\hat{\rho}}{\partial t} = \frac{1}{i\hbar} [\hat{H}, \hat{\rho}] \Rightarrow \frac{\partial\hat{\rho}}{\partial t} = \frac{1}{i\hbar} [\hat{H}, \hat{\rho}] + \mathcal{L}(\hat{\rho}),
\]
(4.12) which is often referred to as the master equation. The \(\mathcal{L}\) is called a Lindblad superoperator [27], [28] and it operates on the density operator: \(\mathcal{L}(\hat{\rho})\). Its purpose is to cause a relaxation of the atomic excitation. To do that it operates on atomic transition operators
$C_m$: $\mathcal{L}(\hat{\rho}) = \mathcal{L}(\hat{\rho}, C_m)$. The explicit form of the Lindblad superoperator is:

$$\mathcal{L}(\hat{\rho}, C_m) = -\frac{1}{2} \sum_m \left( C_m^\dagger C_m \hat{\rho} + \hat{\rho} C_m^\dagger C_m + \sum_m C_m \hat{\rho} C_m^\dagger \right).$$  \hbox{(4.13)}$$

There are several things to note about this expression. Firstly, the summation goes over all decay modes $m$. For instance: for a set of $N$ two-level atoms, the sum goes from $m = 1$ to $N$, as each atom has one decay mode. In the case of $N$ atoms in a three-level system, with two levels from which spontaneous emission can occur per atom, the sum goes from $m = 1$ to $m = 2N$. Furthermore, let us consider a specific decay channel from state $|e\rangle$ to $|g\rangle$. The first term of equation (4.13) describes the loss of population from the excited state $|e\rangle$ due to the spontaneous emission of a photon. The second term describes the change of the coherence terms in the density matrix due to this spontaneous emission. Finally, the last term ensures the restoration of population in the excited state so that $\text{Tr}[\hat{\rho}(t)] = 1 \forall t$. This property makes it by definition a superoperator: an operator which conserves the trace of the system it works on [8].

### 4.3.1 Example: a dissipative two-level system

Let us consider the easiest dissipative system one can think of as an example: one atom in a two-level system with spontaneous emission from its excited state $|e\rangle$. In this case there is only one decay channel: the decay from $|e\rangle$ to $|g\rangle$ for the sole atom. The relevant transition operator and its Hermitian conjugate are therefore:

$$C_e = \sqrt{\gamma_e} |g\rangle \langle e| \hspace{1cm} C_e^\dagger = \sqrt{\gamma_e} |e\rangle \langle g|,$$

with $\gamma_e$ the spontaneous emission rate from state $|e\rangle$. We immediately see that the transition operator working on $|g\rangle$ yields zero. When it works on $|e\rangle$ we get spontaneous decay to the ground state. Plugging these expression into the Lindblad superoperator (4.13) yields:

$$\mathcal{L}(\hat{\rho}) = -\frac{1}{2} \gamma_e \left( |e\rangle \langle g| + |g\rangle \langle e| \right) + \gamma_e |g\rangle \langle e| \hat{\rho} |e\rangle \langle g| = \gamma_e \left( |g\rangle \rho_{ee} \langle g| - \frac{1}{2} |e\rangle \langle e| \right) \hat{\rho} + \frac{1}{2} |e\rangle \langle e| - \frac{1}{2} |e\rangle \langle e| \hat{\rho} \right),$$

Choosing as our basis $\{|e\rangle, |g\rangle\} = \{(1, 0)^T, (0, 1)^T\}$, the Lindblad operator can be written in a matrix representation:

$$\mathcal{L}(\hat{\rho}) = \gamma_e \left( \begin{array}{cc} \rho_{ee} & -\frac{1}{2} \rho_{eg} \\ -\frac{1}{2} \rho_{ge} & \rho_{gg} \end{array} \right).$$

(4.16)

We can write down the Hamiltonian in a matrix representation for a two-level atom interacting with laser light in analogy to Chapter 2:

$$\hat{H} = \frac{\hbar}{2} \begin{pmatrix} -2\Delta & \Omega \\ \Omega & 0 \end{pmatrix}.$$  \hbox{(4.17)}

The density matrix in this basis is given by:

$$\hat{\rho} = \begin{pmatrix} \rho_{ee} & \rho_{eg} \\ \rho_{ge} & \rho_{gg} \end{pmatrix}.$$  \hbox{(4.18)}
Upon plugging in equations (4.16), (4.17) and (4.18) into the quantum Liouville equation, we arrive at the following set of equations:

\[
\begin{align*}
\frac{d\rho_{gg}}{dt} &= \gamma_e \rho_{ee} + \frac{i\Omega}{2} (\rho_{eg} - \rho_{ge}) \\
\frac{d\rho_{ee}}{dt} &= -\gamma_e \rho_{ee} + \frac{i\Omega}{2} (\rho_{ge} - \rho_{eg}) \\
\frac{d\rho_{eg}}{dt} &= -\left(\gamma_e^2 + \frac{i\Delta}{2}\right) \rho_{eg} + \frac{i\Omega}{2} (\rho_{gg} - \rho_{ee}) ,
\end{align*}
\]

where \( \left( \frac{d\rho_{eg}}{dt} \right)^* = \left( \frac{d\rho_{ge}}{dt} \right) \). These are the famous optical Bloch equations \cite{3} for a dissipative two-level atom interacting with an electromagnetic field via a Rabi frequency \( \Omega \) and detuning \( \Delta \).

It is instructive to solve this system of equations. Consider a two-level atom which starts in the ground state: \( \rho_{gg}(t = 0) = 1 \). We can solve the system by using a Runge-Kutta method. The script we used is displayed in appendix C.5. Plotting the results yields figure 4.1.

![Figure 4.1: Plot of the population of the excited state |e⟩ of a two-level atom as a function of time, normalized on the spontaneous emission rate \( \gamma_e \). The atom starts in the ground state and is driven by the electromagnetic field to perform Rabi oscillations. The Rabi oscillation are damped in time, as the probability that spontaneous emission has occurred increases in time. In the figure we see a damped Rabi oscillation in time. The atom starts in the ground state and driven by the electromagnetic field starts to go to the excited state. However, it never reaches that state with probability 1, although the detuning is zero. This is because of the damping due to spontaneous emission: as time progresses, it becomes more and more likely that the atom has spontaneously decayed. The horizontal asymptote lies around 0.5, which corresponds to a superposition of the ground state and excited state, as is expected. The precise value of the horizontal asymptote depends on the fraction \( \gamma/\Omega \) and can be calculated. We shall not go into details, as we prefer a qualitative discussion here. The reader is referred to \cite{3} for further details.](image)
We conclude that using the master equation provides a good way of describing systems
of atoms which can spontaneously decay. However, there is a computational problem
associated with the usage of the master equation. The size of the density matrix is
2^N with N the number of particles for atoms in a two-level system and even 3^N for
atoms in three-level system. This causes the computation time to grow dramatically for
the number of particles. Typically, the computation times goes with \( O(Z^2) \), where Z is
the dimension of the relevant Hilbert space of the system. This quickly gets out of hand.

It is therefore necessary to use an algorithm which needs less computation time. One
such algorithm is the quantum Monte Carlo wavefunction method. It is a routine which
evolves the wavefunction in time. This method has a computation time which scales as
\( O(Z) \). We introduce the method in the next section.

4.4 The quantum Monte Carlo approach

The Quantum Monte Carlo (abbreviated as QMC) approach has been pioneered by
Mølmer, Dalibard and Castin in the 1990s [29], [30]. It is inspired by the master equa-
tion, which was discussed in the previous section. Instead of adding the full Lindblad
superoperator to the quantum Liouville equation, a part of the Lindblad superoperator
is added to the Hamiltonian of the system. Via a stochastic method the simulation
decides in every time step whether or not decay mode \( m \) has been activated. It can
be shown that this method converges to the 'exact' solution of the master equation, if
the number of time steps becomes infinite. We start with discussing the algorithm and
continue with reconsidering the single two-level atom, to verify the correctness of the
algorithm.

4.4.1 The QMC algorithm

Suppose we are given the wavefunction of the system at time \( t \): \( |\psi(t)\rangle \). The QMC
approach presents an algorithm to stochastically calculate the wavefunction at \( t + \delta t: \nolimits\)
\( |\psi(t + \delta t)\rangle \). After averaging over many of such stochastic calculations, we expect to find
a wavefunction that converges to the solution of the master equation.

Let \( \hat{H}_S \) be the Hamiltonian of the system we are concerning. We define a non-Hermitian
Hamiltonian inspired by the Lindblad superoperator:

\[
\hat{H} = \hat{H}_S - \frac{i\hbar}{2} \sum_m C_m^\dagger C_m, \tag{4.20}
\]

where the sum is again over all decay modes \( m \). In this expression we recognize the
first term of the Lindblad superoperator, with the absence of the density operator.
Plugging in this Hamiltonian in the Schrödinger equation yields the new unnormalized
wavefunction:

\[
|\psi^{(1)}(t + \delta t)\rangle = \exp\left(-\frac{i\hat{H}\delta t}{\hbar}\right) |\psi(t)\rangle \approx \left(1 - \frac{i\hat{H}\delta t}{\hbar}\right) |\psi(t)\rangle, \tag{4.21}
\]
where the last approximation is justified if $\delta t \ll 1$. From this equation we can intuitively understand the choice of adding the non-Hermitian term to equation (4.20). For, if we subtract the original wavefunction on both sides of equation (4.21) and divide by $\delta t$ we get:

$$\frac{|\psi^{(1)}(t + \delta t)\rangle - |\psi(t)\rangle}{\delta t} \approx -\frac{i}{\hbar} \hat{H} |\psi(t)\rangle,$$

(4.22)

where we can approximate the left-hand side with the derivative of $|\psi(t)\rangle$, which is justifiable as we assumed $\delta t$ to be small:

$$\frac{d |\psi(t)\rangle}{dt} \approx -\frac{i}{\hbar} \hat{H} |\psi(t)\rangle = -\left( \frac{1}{2} \sum_m C_m^\dagger C_m + \frac{i}{\hbar} \hat{H}_S \right) |\psi(t)\rangle.$$

(4.23)

We immediately see that this differential equation has exponentials as its solution. However, the first term yields a real, negative eigenvalue, whereas the second term yields a purely imaginary eigenvalue. The added non-Hermitian term therefore yields a negative exponential in the solution. This represents spontaneous decay in time.

Now that we understand the addition of the non-Hermitian term, we can proceed with the algorithm. As the used Hamiltonian is non-Hermitian, the new wavefunction is not normalized. We can calculate the new normalization constant $A$:

$$A = \left\langle \psi^{(1)}(t + \delta t)|\psi^{(1)}(t + \delta t)\right\rangle$$

$$\approx \langle \psi(t)| \left( 1 + \frac{i\hat{H}^\dagger \delta t}{\hbar} \right) \left( 1 - \frac{i\hat{H} \delta t}{\hbar} \right) |\psi(t)\rangle$$

$$= 1 - \delta p + O(\delta t^2),$$

with

$$\delta p = \sum_m \delta p_m = \delta t \sum_m \langle \psi(t)|C_m^\dagger C_m|\psi(t)\rangle \geq 0.$$  

(4.25)

The reader should note that as $\delta t \ll 1$, so is $\delta p \ll 1$. We are now ready to decide whether or not a quantum jump has occurred in the interval $[t, t + \delta t]$. We therefore generate a uniformly distributed random number $\epsilon \in [0, 1]$ and compare it with $\delta p$.

There are two options:

1. If $[\delta p < \epsilon]$, no spontaneous emission or quantum jump occurs and we just normalize the new wavefunction. This shall happen in most of the cases, as $\delta p \ll 1$. In this case:

$$|\psi(t + \delta t)\rangle = \frac{|\psi^{(1)}(t + \delta t)\rangle}{\sqrt{A}},$$

(4.26)

where $A$ is the normalization constant from equation (4.24).

2. If $[\delta p \geq \epsilon]$, a photon is spontaneously emitted. We now have to choose which decay mode $m$ was activated and project the new wavefunction by using the relevant transition operator $C_m$. We choose to do this via the probability distribution $\Pi_m = \delta p_m/\delta p$. Then:

$$|\psi(t + \delta t)\rangle = \frac{C_m |\psi(t)\rangle}{(\delta p_m/\delta t)^{1/2}}.$$

(4.27)
The reader should note that the case of $\delta p = \epsilon$ is highly unlikely to happen and it can be included arbitrarily into option one or two. This finalizes the QMC algorithm. As it is a stochastic procedure, it should be run several times and then averaged over all runs. We have implemented the algorithm in a Matlab script, which is displayed in appendix C.7.

### 4.4.2 Example: a dissipative two-level system

To investigate the convergence of the QMC algorithm, we reconsider the two-level atom, with spontaneous emission from its excited state $|e\rangle$. In section 4.3.1 we already considered this system and we derived the optical Bloch equations. We numerically solved those to obtain figure 4.1. Can we obtain the same results by using the QMC algorithm? Using a slightly altered version of the script of appendix C.7 we obtained figure 4.2. It contains plots of the population of the excited state for an atom that starts in the ground state and is subjected to an electromagnetic field on resonance. The different plots are made for an increasing number of runs.

![Plots of the population in the excited state $|e\rangle$](image_url)

(a) $N = 1$  
(b) $N = 10$

(c) $N = 100$  
(d) $N = 10000$

**Figure 4.2:** Plots of the population in the excited state $|e\rangle$ as a function of the time, scaled on $\gamma^{-1}$, with $\gamma$ the spontaneous emission rate. The plots are made for four different numbers of runs: $N = 1$, $N = 10$, $N = 100$ and $N = 10000$. In (a) the points in time at which spontaneous emission takes place are marked with a red circle. The ratio of Rabi frequency to spontaneous emission was chosen to be: $\Omega/\gamma = 2.5$, which is the same as in figure 4.1.
Let us interpret figure 4.2. In subfigure (a) we see the stochastic process at work: at every red circle spontaneous emission occurs. This is completely random: every time the simulation is run for \( N = 1 \) the number of decays and their positions in time shall be different. If we increase \( N \) to 10 and 100, we see that the curve flattens out and starts to resemble the solution to the optical Bloch equations. However, it is only for a large number of runs, i.e. \( N = 10000 \), that the curve really approaches the exact solution.

It is important to note that the plots in figure 4.2 are made for \( \Omega/\gamma = 2.5 \). This means that spontaneous emission happens on a characteristic time scale which is smaller than the time scale of the Rabi oscillations. It is interesting to look at the two remaining regimes: (1) \( \Omega/\gamma = 1 \) and (2) \( \Omega/\gamma < 1 \). A plot for both situations is made in figure 4.3. We see that the steady-state population of \(|e\rangle\) decreases as a function of smaller \( \Omega/\gamma \). This is logical as spontaneous emission becomes more important in this regime. For \( \Omega/\gamma = 1 \) we see a small Rabi-like overshoot, but this quickly dies out. For \( \Omega/\gamma = 0.5 \) we do not see any Rabi oscillations: spontaneous emission has become completely dominant. As a rule of thumb we can therefore conclude that \( \Omega/\gamma \) should be larger than one for Rabi oscillations to dominate spontaneous emission.

Figure 4.3: A plot of the population of the excited state for two different \( \Omega/\gamma \). The atom starts in the ground state and the simulation is run \( N = 10000 \) times in both cases.

Finally, we can perform one last check. What happens if the lasers are switched off, i.e. \( \Omega = 0 \)? If the atom starts in the ground state, we expect the population of \(|e\rangle\) to decrease as an exponential, with a characteristic time \( \gamma^{-1} \). In figure 4.4 a comparison is made between the expected solution and the results from a QMC simulation. They align almost perfectly.

We have now experimentally compared the QMC approach thoroughly with the 'exact' solutions to the master equation - or optical Bloch equations in this case. We can conclude that the QMC approach yields the correct results, if the number of runs is large enough. It can also be proven formally that in the limit \( N \to \infty \Leftrightarrow \delta t \to 0 \) the QMC solution converges to the master equation solution. This is shown in appendix B.2. We are now ready to use the proposed algorithm for more general systems, including \( N > 1 \) atoms in a three-level system.
4.5 Simulating using the QMC algorithm

Consider three atoms in a three-level system, placed on a line. In section 3.4 we thoroughly discussed the eigenvectors of the three-particle Hamiltonian for several values of the relevant parameters. STIRAP turned out to be successful in accomplishing a population inversion from $|ggg\rangle$ to $|rgr\rangle$. This was the case because there was a suitable eigenvector for this population inversion. However, it was shown in figure 3.7 that this eigenvector also has a component $|rer\rangle$ with a finite amplitude during the STIRAP scheme. At that point we did not include spontaneous emission, so it was not a problem. The question now is: how will spontaneous emission influence the result?

To investigate this we run a QMC simulation. A plot of the relevant wavefunction components as a function of time is displayed in figure 4.5. We used the same parameters as in figure 3.7 and included a spontaneous emission rate of $\gamma = 5.98$ MHz. This rate is realistic and has been discussed in section 2.3 in an experimental context and in section 4.1 in a theoretical context.

From figure 4.5 we conclude that the effectiveness of the STIRAP scheme is completely destroyed by spontaneous emission from the intermediate state $|e\rangle$. The system follows the correct eigenvector perfectly for $t < -10 \mu s$. However, as soon as the eigenvector acquires a $|rer\rangle$-component, the system cannot follow the eigenvector anymore due to spontaneous emission. This takes places for $t \in [-10, 10] \mu s$. In this interval there is so much spontaneous emission, that almost all population has failed to follow the correct eigenvector, long before two-Rydberg states are reached. At the end, we see small populations in $|rgg\rangle$, $|ggr\rangle$ and $|grg\rangle$, but virtually nothing in the desired state $|rgr\rangle$. Furthermore, the ground state $|ggg\rangle$ is populated a lot. These are all aspects we explicitly do not want.
Figure 4.5: A QMC simulation of three atoms in a three-level system, placed on a line. The atoms start in their ground states. The parameters are: $\Delta_S + \Delta_P = 0$, $\Omega_S = \Omega_P = -\Delta_P = 10 \text{ Mrad/s}$, $r = 5 \mu m$ and $\gamma = 5.98 \text{ MHz}$. The number of runs was $N = 100$.

So, why does the STIRAP scheme perform so badly? The answer lies in figure 3.7. In this plot the eigenvector is displayed, which we are trying to follow for the parameters in figure 4.5. It has a $|e\rangle$-bump with a maximum population of 0.1 during the process. Apparently, the spontaneous emission rate of $\gamma = 5.98 \text{ MHz}$ is too high for this bump to be tolerated in the STIRAP scheme.

We would like to change the available parameters, i.e. the detunings $\Delta_S$ and $\Delta_P$, the Rabi frequencies $\Omega_S$ and $\Omega_P$, the spontaneous emission rate $\gamma$ and the inter-particle separation $r$ as to arrive at a working STIRAP scheme. There are three possible routes to follow:

1. **Try to get as little population as possible in states with an $|e\rangle$-component during the whole STIRAP process.** The idea is simple: if no atoms ever populate the intermediate state $|e\rangle$, it cannot spontaneously decay from that state, no matter how large $\gamma$ is. In this approach we are free to change the detunings $\Delta_S$ and $\Delta_P$ and also the Rabi frequencies $\Omega_S$ and $\Omega_P$. The spontaneous emission rate $\gamma$ shall be kept at the experimental value. However, for changing the values of the Rabi frequencies we are restricted by experimental constraints as discussed in section 2.3. We follow this route in the following section.

2. **Try to get the stimulated emission rate as low as possible.** In this case, an atom may populate the intermediate state $|e\rangle$ during the scheme. For if the spontaneous emission life time is large enough compared to the timescale of the STIRAP scheme, we expect a negligible amount of decays. In this case, we only need to make sure that at the end of the scheme, the eigenvector is completely in the $|rgr\rangle$-state. In
principle, the spontaneous emission rate $\gamma$ has a fixed value for the Rb-85 atom we are considering. However, if we find an optimal set of parameters, we can reconsider our system. We might find an other atom which satisfies our experimental needs for STIRAP to work. We discuss this route in the second following section.

3. Change the inter-particle separation so that the van der Waals blockade effect is smaller. We saw in chapter 3 that the Rydberg blockade effect is responsible for the occurrence of $|e\rangle$-components in the eigenvectors. The van der Waals interaction energy scales as $C_6/r^6$, so that enlarging the inter-particle separation yields a smaller interaction energy and thus smaller $|e\rangle$-components. This might induce a working STIRAP scheme. We investigate this idea in the third following section.

4.5.1 Varying the detunings and Rabi frequencies

In section 3.4 we investigated the eigenvectors of the Hamiltonian as a function of the variable parameters. In figure 3.7 we observed a non-zero population in the intermediate state, which caused the STIRAP of figure 4.5 to fail in establishing a population inversion from $|ggg\rangle$ to $|rgr\rangle$. In figure 3.8 we saw that making $\Omega_S/\Omega_P > 1$ and $\Delta_P/\Omega_P < -1$ yielded an eigenvector which causes a complete population transfer from $|ggg\rangle$ to $|rgr\rangle$, whereas the population of the intermediate state stays negligible small. This seems promising. A simulation for both sets of parameters from figure 3.8 is displayed in figure 4.6.

Figure 4.6: A QMC simulation for the three-level system containing three atoms. For both figures holds: $r = 5 \mu m$, $\Omega_S = 5\Omega_P$, $\Delta_P = -10\Omega_P$ and the number of runs $N = 100$.

We see that the STIRAP process from 4.6(a) clearly is not adiabatic enough. There is a correct eigenvector, as is displayed in figure 3.8, but this changes to rapidly in time for the chosen STIRAP procedure, and is thus difficult to follow adiabatically. However, in 4.6(b) we do manage to follow the eigenvector adiabatically by increasing the strength of the lasers, i.e. the Rabi frequencies. Note that although figure 4.6(b) is the result we wish to accomplish, it is unfortunately only possible to achieve in theory. We argued in section 2.3 that realistic values for the Rabi frequencies are: $\Omega_S = 10$ Mrad/s and $\Omega_P = 40$ Mrad/s. The values used in this plot exceed the realistic values by a factor fifty. If this procedure is to be used, stronger lasers should be made available.
We observe a great resemblance to the simulations without spontaneous emission in figure 3.9. This is not so strange, as the parameters were chosen specifically to remove the $|rer\rangle$-component in the eigenvector. Clearly, we have succeeded, as running the simulation with spontaneous emission yields almost the same result. However, we obtain from figure 4.6(b) that $\langle rgr | \psi \rangle$ obtains a steady-state value of 0.965, whereas this was a full 1 when spontaneous emission was not included. So, even when the $|ere\rangle$-component is incredibly small, the high spontaneous emission rate still has some influence. We can also observe this influence by noting that in figure 4.6(a) the steady-state values have a different order.

Although we can only obtain correct results for unrealistically strong lasers, we can still investigate whether or not the full excitation scheme works if we include spontaneous emission. Assuming we can perform one pulse to get to the $|rgr\rangle$ state, can we then obtain full population inversion to $|rrr\rangle$ by applying a second pulse? The answer is a partial yes: in figure 4.7 we obtained such a population inversion for realistic parameters. However, the steady-state value is: $\langle rrr | \psi \rangle_{\text{end}} = 0.98$. In the simulation we observed two runs with an extreme number of decays, whereas the other 98 did not decay at all. It seems that if one particle decays at some time in a run, it is constantly re-excited and decays again during the rest of the run. This is curious and interesting for further research. Again we conclude that even though we have an almost perfect eigenvector to follow, the spontaneous emission is still large enough to have some influence on the course of the population inversion.

![Figure 4.7: A QMC simulation in which we obtain almost full population inversion from $|rgr\rangle$ to $|rrr\rangle$. The steady-state value is: $\langle rrr | \psi \rangle_{\text{end}} = 0.98$. The used parameters are: $\Omega_P = \Omega_S = 10$ Mrad/s and $\hbar(\Delta_P + \Delta_S) = 2C_6/r^6$ with $r = 5$ µm and $\Delta_P = 0$. The simulation has been run $N = 100$ times.]

4.5.2 Varying the spontaneous emission rate

In this section we investigate the influence of the size of the spontaneous emission rate $\gamma$ on the success of the STIRAP scheme. We saw in figure 4.5 that the effect of spontaneous emission was too significant for STIRAP to be successful. In that case we had: $\Omega_P/\gamma = \Omega_S/\gamma = 10/(2\pi \cdot 5.98) \approx 0.27$. When we considered a dissipative two-level
system in section 4.4.2 we already noted that for STIRAP to work, we need to be in the regime where \( \Omega/\gamma > 1 \). This is not the case for figure 4.5.

In figure 4.8 we plotted the end population of the desired state \( |rgr\rangle \) for three particles on a line with a inter-particle separation of 5 \( \mu m \), as a function of a new spontaneous emission rate \( \Gamma \in [0, 1] \cdot \gamma \). This new rate \( \Gamma \) is normalized on the old rate \( \gamma \). We would expect the graph to be a smooth transition from zero end population in \( |rgr\rangle \) for \( O(10^{-1} - 10) \) to a probability of 1 for the end population in \( |rgr\rangle \) for small fractions of the original spontaneous emission rate. Globally, we observe such behaviour, but locally the graph is not really smooth and not always monotone decreasing. This is just the consequence of too few runs to average out stochastic errors.

Besides this, we observe that for \( \Gamma < \gamma/100 \) the end population in \( |rgr\rangle \) goes \( \sim 1 \). If we were to find a three-level system with the same useful properties as Rb-85, but a spontaneous emission rate of \( \gamma/100 \), we might be able to perform a working STIRAP scheme in practice.

![Graph](image.png)

Figure 4.8: A semi-logarithmic plot of the end population of \( |rgr\rangle \), i.e. \( |\langle rgr|\psi\rangle|_{\text{end}} \) against the normalized spontaneous emission rate \( \Gamma \). The number of runs is \( N = 75 \) for each value of \( \Gamma \). The inter-particle separation is still 5 \( \mu m \).

### 4.5.3 Varying the inter-particle separation

We now know that there is a regime for the spontaneous emission rate for STIRAP to work, i.e. \( \Gamma < \gamma/100 \), in the case of three particles on a line with an inter-particle separation of 5 \( \mu m \). We argued earlier that this distance is a realistic value for the system. However, it is interesting to look at the consequences of enlarging this separation. For a larger separation, the van der Waals interaction becomes smaller. Therefore, the Blockade effect becomes less important. As the interaction of particles via the Blockade effect creates \( |e\rangle \)-components in the eigenvectors, we expect to reduce the influence of spontaneous emission for larger separations \( r \).
Let us define the van der Waals interaction energy to be \( V \) between two atoms in a the system of three particles on a line with 5 \( \mu \text{m} \) separation. What happens if we place the particles further apart so that the van der Waals interaction energy becomes a fraction of its initial value? To answer this question we made a plot like in figure 4.8 for \( V/2, V/4, V/8 \) and \( V/16 \). It is displayed in figure 4.9.

![Figure 4.9: Several plots of the end population of \(|rgr\rangle\), i.e. \(|\langle rgr|\psi\rangle|_{\text{end}}\) against the normalized spontaneous emission rate \( \Gamma \), for different van der Waals interaction energies. The number of runs is \( N = 100 \) for each value of \( \Gamma \). The inter-particle separations are respectively 5 \( \mu \text{m} \), 5.6 \( \mu \text{m} \), 6.3 \( \mu \text{m} \), 7.07 \( \mu \text{m} \) and 7.94 \( \mu \text{m} \). The used parameters are: \( \Omega_P = \Omega_S = -\Delta_P = 20 \text{Mrad/s} \) and \( \Delta_S = 0 \).

The first thing we can conclude from figure 4.9 is that for smaller \( V \), i.e. larger inter-particle separations, the end population of \(|rgr\rangle\) becomes smaller and smaller. This is observable when we compare the green and red plots in the figure. The explanation is easy: for larger separations, the particles influence each other less. We showed in figure 3.6 that for three particles on a line that do not interact, there is an eigenvector from \(|ggr\rangle\) to \(|rrr\rangle\). Therefore the end population in \(|rgr\rangle\) becomes less as the interaction weakens and the population in \(|rrr\rangle\) therefore increases. This is exactly what we observe in figure 4.9.

Furthermore, we observe that for \( V/2 \) and \( V/4 \) the end population in \(|rgr\rangle\) is higher than for \( V \), for all \( \Gamma \). The effect is tiny: the blue and yellow curve seem to lie slightly higher than the black curve. This might be a sign that in this regime, the \(|e\rangle\)-components are the smallest in the eigenvectors, so that the influence of spontaneous emission is the smallest. Further simulations with more runs and more intermediate fractions of \( V \) might shed more light on this. As the required number of runs is quite large: \( N \gg 27 \), it is faster to use the density matrix approach in this specific system for further inquiries.

64
4.6 Summary

In this chapter we have studied the STIRAP scheme for multi-particle three-level systems with a non-zero spontaneous emission rate $\gamma$. We started with deriving a semi-classical expression for the spontaneous emission rate of an excited level in an atom. In this derivation we used the Wigner-Weisskopf approximation. Using this expression, we calculated a spontaneous emission rate which deviated 4.4% from the experimental value.

We continued with a review of density operator formalism, which can be used to describe a wider range of phenomena than the more standard wavefunction formalism. We derived the quantum Liouville equation, which is the equivalent of the Schrödinger equation in the wavefunction formalism. Whereas spontaneous emission is notoriously hard to include in the wavefunction formalism, it is easy in the density operator formalism: we just add a Lindblad superoperator to the quantum Liouville equation. This operator is a function of the decay mode operators. As an example of this approach we consider a dissipative two-level system. We show that the optical Bloch equations can be derived by including the Lindblad superoperator. We then numerically solve these so-called master equations for several ratios of the spontaneous emission rate and the Rabi frequency. We conclude that $\Omega/\gamma > 1$ is a rule of thumb for STIRAP to work.

The density operator approach has one big disadvantage: the number of variables scales as $O(N^2)$, with $N$ the dimension of the Hilbert space of the system. We therefore resorted to a stochastic wavefunction approach, called the quantum Monte Carlo (QMC) procedure, for which the number of variables scales as $O(N)$. This way the computation time stays manageable. We introduced the algorithm and wrote a Matlab script to implement it. As a check, we recalculated the dissipative two-level system with the QMC procedure and compared it with the master equation approach. The QMC approach converges to the master equation solution for a large number of runs $N$, or short timesteps $\delta t$: $N \to \infty \Leftrightarrow \delta t \to 0$.

Finally, using the QMC algorithm, we performed several simulations for three particles on a line with spontaneous emission. It turned out that spontaneous emission ruins the STIRAP scheme. The rate is so large, that all the atoms immediately fall back down to the ground state if they obtain a component in the excited state. We investigated the influence of the Rabi frequencies and detunings and concluded that for unrealistic high laser intensities - and thus Rabi frequencies - we can get a working STIRAP scheme. Furthermore, we looked into the critical value of the spontaneous emission rate and found that it should be significantly lower for the STIRAP scheme to work under realistic parameters. Finally, we varied the inter-particle separation. It was shown that for small values of the van der Waals interaction energy, the end population in $|rgr\rangle$ is small, whereas the end population of $|rrr\rangle$ is large. This is because the particles are no longer interacting at large separations. For values between five and seven $\mu m$ there might be a tendency for STIRAP to work better. The effect is small and should be investigated further with more and better simulations.
Chapter 5

Conclusions and discussion

The goal of this research project was to investigate the theoretical possibilities of creating strongly-coupled Rydberg crystals by exciting atoms in an ultra-cold atomic gas from their ground state to the Rydberg state.

Rydberg atoms are atoms with one electron excited to a state with a high principal quantum number. These atoms have extreme properties, one of which is a life time of up to several hundreds of $\mu$s. We noted that in an ultra-cold atomic gas the frozen-gas limit holds on the same time scale. This means that atoms have such a low kinetic energy, that they barely move at certain time scales. We concluded that one demand for our excitation scheme is a maximum time scale of a few hundred $\mu$s. If this condition is fulfilled, we can consider the atoms to be motionless and without spontaneous emission from the Rydberg state during the excitation scheme.

One other important aspect for our excitation scheme is the van der Waals blockade effect. This effect arises because an atom in a Rydberg state has a high polarizability, so that it raises the energy landscape around it due to possible induced dipole interactions between two Rydberg atoms. In a lattice, this means that the excitation energy for an atom is a function of the number of nearest neighbours in the Rydberg state. It is therefore impossible to excite all atoms in a lattice to a Rydberg state with one monochromatic laser pulse. This is something we have to account for in our excitation scheme.

A potential excitation scheme for the creation of Rydberg crystals makes use of the STIRAP procedure. Because a direct transition from the ground to the Rydberg state is not possible, the STIRAP scheme requires the use of an additional intermediate state. However, this intermediate level must remain unpopulated at all times, as it has a high spontaneous emission rate. We showed that the STIRAP procedure can succeed at causing a transition of one atom from the ground state to the Rydberg state, without populating the intermediate state at any time. The STIRAP method uses two lasers which induce time-dependent Rabi frequencies and depends on adiabatic following of the proper eigenstate. By performing simulations we found that using higher Rabi frequencies and a lower detuning of the intermediate state increases the adiabaticity of the process.
Because of the van der Waals interactions, we cannot excite two adjacent atoms to the Rydberg state using only one pulse. We therefore proposed an excitation scheme which uses multiple STIRAP pulses with an increased total detuning for each subsequent scheme, to account for the van der Waals blockade effect. During each STIRAP scheme, a suitable eigenvector must be followed adiabatically. Upon application of the STIRAP scheme to a multi-particle system consisting of three particles on a line, we found that the van der Waals interactions induce components of the intermediate level in the eigenstates. By increasing the detuning of the intermediate state, these components can be suppressed. However, we showed that by using this increased detuning, we require very large Rabi frequencies to be able to follow the appropriate eigenstate on reasonable timescales.

Investigating other configurations of atoms yielded similar results for the eigenstates: all required a detuning of the intermediate state to suppress population of the intermediate level. For the symmetric systems we considered, we found that the STIRAP scheme yields superpositions of states which only differ from each other by a symmetry operation. For further research, it would be interesting to investigate STIRAP schemes on more complex systems and see whether we can exploit the symmetry to predict the eigenvectors induced by the STIRAP schemes.

As we observed that the van der Waals interactions induce eigenstate components which allow population of the intermediate state, we included spontaneous emission in our subsequent models. We therefore calculated the spontaneous emission rate from an excited state for a general two-level system in the Wigner-Weisskopf model. Using density operator theory, we showed that it is easy to include spontaneous emission to the description of a system. To accomplish this, we added a Lindblad superoperator to the quantum Liouville equation, which ensures a relaxation of the excited state of the atom. This rather general treatment was applied to a two-level system to yield the well-known optical Bloch equations, a specific form of the more general master equations.

However, the problem with a density operator treatment is that the computation time goes as $\mathcal{O}(Z^2)$, with $Z$ the dimension of the relevant Hilbert space. This quickly diverges for larger systems. We therefore proposed to use a stochastical Quantum Monte Carlo algorithm (QMC), which has $\mathcal{O}(Z)$ parameters and basically evolves the wavefunction in time. We showed both by simulations and and theoretically that solving the master equations is equivalent to using the QMC approach, if the number of runs is large enough.

Using the QMC algorithm, we showed that the STIRAP pulse scheme does not work when we include spontaneous emission. The reason is the fact that interacting particles obtain components of the intermediate state. For the system of Rb-85 we studied, the spontaneous emission was so high that if an atom enters the $|e\rangle$ state, it immediately falls back again. Solving this problem might be done in two ways. Firstly, one can adjust the Rabi frequencies and detunings. We showed that using very large parameters can result in a working STIRAP scheme. Secondly, one can search for a three-level system with a smaller spontaneous emission rate. We found that a system with a spontaneous emission rate of one hundredth of the value for Rb-85 would suffice. It would be interesting to look for such a system in the atomic tables. Finally, it was shown that there
might be an optimal inter-particle separation. This has to be confirmed by running more simulations.
Bibliography


Appendix A

List of symbols and definitions

This list displays the used symbols and their definitions in a chronological order:

- $C_6$: coefficient which determines the 'strength' of the van der Waals interaction. For Rydberg atoms, it scales as $\sim n^{11}$, with $n$ the principal quantum number.

- $\Delta \equiv \omega_0 - \omega$: the detuning, defined as the difference in frequency between the relevant transition frequency $\omega_0$ and the frequency of the light $\omega$.

- $(\mathbf{d} \cdot \mathbf{E}_0)_{eg} \equiv \langle e | \mathbf{d} \cdot \mathbf{E}_0 | g \rangle$: the dipole transition moment from the ground state to the excited state.

- $\Omega_R = \sqrt{\Delta^2 + (\mathbf{d} \cdot \mathbf{E}_0/\hbar)^2}$: the generalized Rabi frequency.

- $\Omega_0 = \mathbf{d} \cdot \mathbf{E}_0/\hbar$: the 'ordinary' Rabi frequency for zero detuning.

- $\Delta_S = \omega - (E_r - E_e)/\hbar$: the detuning between the frequency of the laser $\omega$ and the atomic transition frequency between $|e\rangle$ and $|r\rangle$.

- $\Delta_P = \omega - (E_e - E_g)/\hbar$: the detuning between the frequency of the laser $\omega$ and the atomic transition frequency between $|g\rangle$ and $|e\rangle$.

- $\Omega_P$: the Rabi frequency induced by the Pulse laser between level $|g\rangle$ and $|e\rangle$.

- $\Omega_S$: the Rabi frequency induced by the Stokes laser between level $|e\rangle$ and $|r\rangle$.

- $\omega^\pm$ and $\omega^0$: eigenvalues of the Hamiltonian for one particle in a three-level system.

- $|a^0\rangle, |a^\pm\rangle$: eigenvector of the Hamiltonian for one particle in a three-level system. For one-particle STIRAP $|a^0\rangle$ should be followed adiabatically.

- $\Theta \equiv \arctan (\Omega_S/\Omega_P)$: the mixing angle which is varied adiabatically from $0$ to $\pi/2$ for a successful STIRAP scheme.

- $\Phi = \Phi(\Omega_P, \Omega_S, \Delta_P, t)$: variable which determines amplitude of different components in the eigenfunctions $|a^{0,\pm}\rangle$.

- $\Omega_j \equiv \mathbf{d} \cdot \mathbf{E}_j/\hbar$: single-mode Rabi frequency.

- $\gamma$: spontaneous emission rate. For Rb-85 it has a value of 5.98 MHz.
• $\tau \equiv 1/\gamma$: spontaneous emission life time. For Rb-85 it has a value of 1 $\mu$s.

• $\hat{\rho}$ and $\hat{\sigma}$: general symbols for density operators. A subscript "S" symbolizes that the density operator works on the system and a subscript "R" symbolizes that it works on the reservoir.

• $\mathcal{L}(\hat{\rho})$: the Lindblad superoperator which works on the density matrix $\hat{\rho}$.

• $C_m$: the transition operator for decay mode $m$. For one atom in a two-level system it is: $C = \sqrt{\gamma} \, |g\rangle \langle e|$.

• $|\psi(1)(t + \delta t)\rangle$: unnormalized wavefunction after time-evolution with the non-Hermitian Hamiltonian as defined in the QMC algorithm.

• $\epsilon \in [0, 1]$: stochastic parameter used in the QMC algorithm to decide whether or not an atom has decayed.
Appendix B

Derivations

B.1 The Hamiltonian in a rotating frame

Consider an $M$-level system with 1 particle. We define $\epsilon_m$ as the one-particle energy of the state $|m\rangle$, which describes one particle in the $m$th level. Here, $|m\rangle$ denotes one of the $M$ orthonormal basis states in the $M$-dimensional Hilbert space $\mathcal{H}$. We take $\epsilon_1 \equiv 0$ and $m_2 > m_1 \Rightarrow \epsilon_{m_2} > \epsilon_{m_1}$. Furthermore, a laser with frequency $\omega_{j,k}$ is present, which can cause transitions between $|j\rangle$ and $|k\rangle$. The Hamiltonian for this system can be written as

$$\hat{H}_1 = \sum_{m=1}^{M} \epsilon_m |m\rangle \langle m| + \frac{\hbar}{2} \sum_{j=1}^{M} \sum_{k=1}^{M} \Omega_{j,k} \cos (\omega_{j,k} t) (|j\rangle \langle k| + |k\rangle \langle j|).$$  \hspace{1cm} (B.1)

Here, $\Omega_{j,k}$ is the Rabi frequency of the corresponding laser. Because every pair of states appears twice in the double sum in (B.1), a factor $\frac{1}{2}$ is added.

Next, consider an $M$-level system with $N$ particles. In this case, the system can be described by an $M^N$-dimensional Hilbert space, which is the tensor product of $N M$-dimensional Hilbert spaces $\mathcal{H}$:

$$\mathcal{H}^{\otimes N} = \bigotimes_{n=1}^{N} \mathcal{H}.\hspace{1cm} (B.2)$$

We can now write the Hamiltonian as

$$\hat{H} = \sum_{i=1}^{M^N} E_i |i\rangle \langle i| + \frac{\hbar}{2} \sum_{j=1}^{M^N} \sum_{k=1}^{M^N} \Omega_{j,k} \cos (\omega_{j,k} t) (|j\rangle \langle k| + |k\rangle \langle j|).$$  \hspace{1cm} (B.3)

Let us now split this Hamiltonian in two parts as follows:

$$\hat{H} = \hat{H}_E + \hat{H}_c.$$  

Here, the first operator

$$\hat{H}_E = \sum_{i=1}^{M^N} E_i |i\rangle \langle i|$$  \hspace{1cm} (B.4)

contains the energies of the states. The energy of a state now consist of two parts. Firstly, it contains the sum the one-particle energies of the $N$ individual particles. Secondly,
interaction terms are added, depending on the energy levels and the spatial configuration of all atoms. We can write this as follows:

\[ \hat{H}_E = \sum_{i=1}^{MN} E_i \ket{i} \bra{i} = \sum_{n=1}^{N} \sum_{m=1}^{M} \epsilon_{nm} \ket{m_n} \bra{m_n} + \sum_{i=1}^{MN} V_i \ket{i} \bra{i}. \]  

(B.5)

Here \( \ket{m_n} \bra{m_n} \) is a projection operator which projects a vector in \( \mathcal{H}^N \) on the subspace where the \( n \)-th atom is in the \( m \)-th level. For example, \( \ket{3_2} \bra{3_2} \) operating on \( \ket{\psi} = \ket{k} \otimes \ket{l} \otimes \ket{m} \otimes \ldots \) returns a zero vector if \( l \neq 3 \) and \( \ket{k_2} \) if \( l = 3 \).

For the following discussion it is not necessary to specify the interaction terms \( V_i \).

The second operator

\[ \hat{H}_c = \frac{\hbar}{2} \sum_{j=1}^{MN} \sum_{k=1}^{MN} \Omega_{j,k} \cos (\omega_{j,k} t) (\ket{j} \bra{k} + \ket{k} \bra{j}) \]  

(B.6)

describes the coupling of the states. In case a transition between two states \( \ket{j} \) and \( \ket{k} \) is impossible, we can define \( \Omega_{j,k} \equiv 0 \).

To transform the Hamiltonian (B.3) to a rotating frame, we define an operator \( \hat{U} = e^{i\hat{A}t} \). The operator \( \hat{A} \) is a diagonal matrix and can be written as

\[ \hat{A} = \sum_{i=1}^{MN} \alpha_i \ket{i} \bra{i}, \]  

(B.7)

where the \( \alpha_i \) are angular frequencies which are yet to be determined. Because \( \alpha_i \in \mathbb{R} \) for all \( i \), \( \hat{A} \) is Hermitian and consequently \( \hat{U} \) is unitary. We can transform a state vector \( \ket{\psi} \) to a state vector \( \ket{\tilde{\psi}} \) in a rotating frame by use of the following transformation: \[ \ket{\tilde{\psi}} = \hat{U} \ket{\psi}. \]  

(B.8)

Note that since \( \hat{U} \) is a unitary transformation, inner products in the rotating frame are preserved. Using this definition, we can write out the Schrödinger equation in the rotating frame:

\[ i\hbar \frac{d}{dt} \ket{\tilde{\psi}} = i\hbar \frac{d}{dt} \left( \hat{U} \ket{\psi} \right) = i\hbar \hat{U} \frac{d}{dt} \ket{\psi} + i\hbar \frac{d\hat{U}}{dt} \ket{\psi} = \hat{U} \hat{H} \ket{\psi} - \hbar \hat{A} \ket{\psi} = \hat{U} \hat{H} \hat{U}^\dagger \ket{\tilde{\psi}} - \hbar \hat{A} \ket{\tilde{\psi}} \equiv \hat{H}_{\text{rot}} \ket{\tilde{\psi}}, \]  

(B.9)

where we have used the Schrödinger equation for \( \ket{\psi} \). It follows that the Hamiltonian in the rotating frame is given by

\[ \hat{H}_{\text{rot}} = \hat{U} \hat{H} \hat{U}^\dagger - \hbar \hat{A}. \]  

(B.10)
Our next step will be to find an expression for $\hat{H}_{\text{rot}}$ and choosing suitable values for the frequencies $\alpha_i$.

We will again start by splitting the Hamiltonian in two parts:

$$\hat{H} = \hat{U} \left( \hat{H}_E + \hat{H}_c \right) \hat{U}^\dagger - \hbar \hat{A} \equiv \hat{H}_{E,\text{rot}} + \hat{H}_{c,\text{rot}} \quad (B.11)$$

where

$$\hat{H}_{E,\text{rot}} = \hat{H}_E - \hbar \hat{A} \quad (B.12)$$

$$\hat{H}_{c,\text{rot}} = \hat{U} \hat{H}_c \hat{U}^\dagger \quad (B.13)$$

Note that we have used the fact that $\hat{U} \hat{H}_E \hat{U}^\dagger = \hat{H}_E$, which stems from the fact that both $\hat{H}_E$ and $\hat{U}$ are diagonal and diagonal matrices commute.

Using the definitions (B.6) and (B.7), we determine the $\hat{H}_{c,\text{rot}}$:

$$\hat{H}_{c,\text{rot}} = \hat{U} \hat{H}_c \hat{U}^\dagger = \exp \left( \frac{i \hbar}{2} \sum_{l=1}^{M} \sum_{n=1}^{N} \alpha_l \left| l \right\rangle \left\langle l \right| \right) \exp \left( -i \hbar \sum_{l=1}^{M} \sum_{n=1}^{N} \Omega_{n,l,k} \cos (\omega_{n,l,k} \hbar) \left( \left| j \right\rangle \left\langle k \right| + \left| k \right\rangle \left\langle j \right| \right) \right)$$

$$= \frac{\hbar}{2} \sum_{j,k=1}^{M} \Omega_{j,k} \cos (\omega_{j,k} \hbar) \left( e^{it(\alpha_j - \alpha_k)} \left| j \right\rangle \left\langle k \right| + e^{it(\alpha_k - \alpha_j)} \left| k \right\rangle \left\langle j \right| \right)$$

$$= \frac{\hbar}{4} \sum_{j,k=1}^{M} \Omega_{j,k} \left( e^{it(\omega_{j,k} + \alpha_j - \alpha_k)} + e^{it(-\omega_{j,k} + \alpha_j - \alpha_k)} + e^{it(\omega_{j,k} + \alpha_k - \alpha_j)} + e^{it(-\omega_{j,k} + \alpha_k - \alpha_j)} \right) \left| j \right\rangle \left\langle k \right|,$$  

(B.14)

where we have used the fact that the basis states are orthonormal: $\langle j|k \rangle = \delta_{jk}$ for all $j$ and $k$.

Next, we aim to choose suitable values for $\alpha_i$ in order to simplify the Hamiltonian. We write the basis state $\left| i \right\rangle$ as

$$\left| i \right\rangle = \left| m_1 m_2 m_3 ... m_N \right\rangle,$$

so with the first atom in the $m_1$th level, the second atom in the $m_2$th level, et cetera. The choice for the corresponding $\alpha_i$ is as follows:

$$\alpha_i = \sum_{n=1}^{N} \left( \sum_{j=1}^{m_n-1} \omega_{j,j+1} \right) \quad (B.15)$$

where $\omega_{j,j+1}$ is the angular frequency of the laser that causes a transition between the levels $j$ and $j+1$. We note that throughout the entire report, the Hamiltonian only
includes transitions between states that have only one atom in a different one-particle state. Moreover, this atom can only undergo a transition from level \( j \) to \( j + 1 \) or \( j - 1 \). Therefore, for all pairs of basis states \( |j\rangle \) and \( |k\rangle \) which require another kind of transition, we take \( \Omega_{j,k} \equiv 0 \).

Consider a pair of states \( |j\rangle \) and \( |k\rangle \) with \( \Omega_{j,k} \neq 0 \). In this case, there is one atom which undergoes a transition from level \( m \) to \( m + 1 \), or from \( m + 1 \) to \( m \). Naturally, the corresponding laser frequency is \( \omega_{j,k} = \omega_{m,m+1} \). Using the chosen values for \( \alpha_i \) \((B.15)\), it follows that \( |\alpha_j - \alpha_k| = \omega_{m,m+1} \). The result is that in equation \((B.14)\), the pairs of exponentials preceding the projection operators yield 1 and \( e^{\pm 2i\omega_{m,m+1}t} \). We then apply the rotating wave approximation. This approximation indicates that the exponentials \( e^{\pm 2i\omega_{m,m+1}t} \) oscillate so fast that on the time scales we are considering, we can replace them with their average values 0. For this approximation to be valid, we require that all the cumulative detunings and Rabi frequencies are negligible compared to all the laser frequencies \((32)\):

\[
\forall m \in \{1, 2, ..., M - 1\} : \omega_{m,m+1} \gg \max_{k \in \{1, 2, ..., M - 1\}} \{|\Delta_k + 1|, \Omega_{k,k+1}\}.
\]

Using this approximation, the following expression for \( \hat{H}_{c,\text{rot}} \) is found:

\[
\hat{H}_{c,\text{rot}} = \frac{\hbar}{2} \sum_{n=1}^{N} \sum_{m=1}^{M-1} \Omega_{m,m+1} (|m_n\rangle \langle (m+1)_n| + |(m+1)_n\rangle \langle m_n|).
\]

(B.17)

Using equations \((B.20)\) and \((B.17)\) we finally obtain an expression for the Hamiltonian
in the rotating frame:

\[
\hat{H}_{\text{rot}} = \hat{H}_{E,\text{rot}} + \hat{H}_{c,\text{rot}}
\]

\[
= -\hbar \sum_{n=1}^{N} \sum_{m=1}^{M} \Delta_m |m_n\rangle \langle m_n|
\]

\[
+ \frac{\hbar}{2} \sum_{n=1}^{N} \sum_{m=1}^{M-1} \Omega_{m,m+1} (|m_n\rangle \langle (m+1)_n| + |(m+1)_n\rangle \langle m_n|)
\]

\[
+ \sum_{i=1}^{M^N} V_i |i\rangle \langle i| , \tag{B.20}
\]

where \(\sum_m\) sums over the \(M\) levels, \(\sum_n\) sums over the \(N\) atoms and \(\sum_i\) sums over all \(M^N\) states.
B.2 Equivalence master equation and QMC procedure

Reconsider the master equation:

\[
\frac{d\hat{\rho}_S}{dt} = \frac{1}{i\hbar} [\hat{H}_S, \hat{\rho}_S] + \mathcal{L}(\hat{\rho}_S), \quad (B.21)
\]

where \( \hat{\rho}_S \) is the density operator of the system, \( \hat{H}_S \) the Hamiltonian of the system and \( \mathcal{L}(\hat{\rho}_S) \) the Lindblad superoperator from equation (4.13). We show in this appendix that solving the master equation is equivalent to propagating the wavefunction as purposed in the QMC algorithm, provided the number of runs is large enough.

Let us assume that the system is in a pure state. Assuming this is not crucial, the discussion is easily generalized to a statistical mixture. Consider a given wavefunction \( |\psi(t)\rangle \) and density operator \( \sigma(t) = |\psi(t)\rangle \langle \psi(t)| \), calculated with the QMC procedure. We can calculate the average density operator at time \( t + \delta t \):

\[
\sigma(t + \delta t) = P_1 \sigma(t + \delta t)|_{\text{No jump}} + \sum_m P_m \sigma(t + \delta t)_m|_{\text{jump atom } m}
\]

\[
= (1 - \delta p) \left| \frac{\psi^{(1)}(t + \delta t)}{\sqrt{1 - \delta p}} \right| \left| \frac{\psi^{(1)}(t + \delta t)}{\sqrt{1 - \delta p}} \right| + \sum_m \delta p_m C_m \langle \psi(t) | C_m^\dagger \langle \psi(t) | C_m^\dagger,
\]

where we calculated the expectation value for the density operator at time \( t + \delta t \) by multiplying the possible outcomes with their probabilities. When we introduced the QMC procedure, we showed that for sufficiently small \( \delta t \), the following unnormalized wavefunction follows from time-evolvement of the Schrödinger equation with the non-Hermitian matrix \( \hat{H} \):

\[
|\psi^{(1)}(t + \delta t)\rangle = \left( 1 - \frac{i \hat{H} \delta t}{\hbar} \right) |\psi(t)\rangle + \mathcal{O}(\delta t^2). \quad (B.23)
\]

The non-Hermitian Hamiltonian was chosen to be:

\[
\hat{H} = \hat{H}_S - \frac{i\hbar}{2} \sum_m C_m^\dagger C_m \leftrightarrow \hat{H}^\dagger = \hat{H}_S + \frac{i\hbar}{2} \sum_m C_m^\dagger C_m, \quad (B.24)
\]

where it was used that the system has a Hermitian Hamiltonian. Plugging in these expressions into equation (B.22) yields:

\[
\sigma(t + \delta t) = \left( 1 - \frac{i \hat{H} \delta t}{\hbar} \right) |\psi(t)\rangle \langle \psi(t)| + \delta t \sum_m C_m \sigma(t) C_m^\dagger
\]

\[
= \left( \sigma(t) - \frac{i \delta t}{\hbar} \hat{H} \sigma(t) \right) + \delta t \sum_m C_m \sigma(t) C_m^\dagger
\]

\[
= \sigma(t) + \frac{i \delta t}{\hbar} [\sigma(t), \hat{H}_S] + \delta t \left( \sum_m C_m \sigma(t) C_m^\dagger - \frac{1}{2} (C_m^\dagger C_m + C_m C_m^\dagger \sigma) \right) + \mathcal{O}(\delta t^2)
\]

\[
\equiv \sigma(t) + \frac{i \delta t}{\hbar} [\sigma(t), \hat{H}_S] + \mathcal{L}(\sigma(t)) + \mathcal{O}(\delta t^2), \quad (B.25)
\]
where we recognized the definition of the commutator and the Lindblad superoperator in the last two steps. As $\delta t \ll 1$ we shall neglect $O(\delta t^2)$. Upon subtracting $\sigma(t)$ on both sides of equation (B.25), dividing by $\delta t$ and averaging, we obtain:

$$\frac{d\sigma}{dt} = i\frac{\hbar}{\hbar}[\sigma, \hat{H}_S] + L(\sigma)$$

(B.26)

where the definition of the derivative was used under the assumption that $\delta t \to 0$. This effectively means that the number of runs should be infinite. Provided that the density operator of the system and $\bar{\sigma}$ coincide at $t = 0$, we have now proven that the two methods are equivalent for $\delta t \to 0$. In this discussion we followed [30].
Appendix C

Matlab scripts

C.1 Two small, but useful functions

These scripts were written by ir. R.M.W. van Bijnen.

In this appendix we define two useful functions, which were used in virtually all following scripts. The function \texttt{getState} calculates the state from a number called 'index', as an N-digit q-base number. It returns a column vector.

\begin{verbatim}
function state = getState(index, N, q)
    % Construct an array with powers of q
    Q = uint32(q .^ (0 : N));

    % Compute the state array
    state = zeros(N, length(index), 'uint32');
    for k = N : -1 : 1
        % Get the factor of q^k present in the index
        state(k, :) = idivide(uint32(index), Q(k));
        % Subtract the factor of q^k
        index = index - bsxfun(@times, state(k, :), Q(k));
    end
end
\end{verbatim}

The function \texttt{getIndex} is the complement of the function \texttt{getState}: given a column vector 'state', a basis 'q' and the number of digits 'N', it returns the index of the state.

\begin{verbatim}
function index = getIndex(state, N, q)
    % Calculates the index (number) represented by the array 'state', \% which is an N-digit, base-q number

    % Construct an array with powers of q
    Q = uint32(q .^ (0 : (N - 1)));

    % Compute the value of the index
    index = sum(bsxfun(@times, uint32(state), Q));
end
\end{verbatim}
C.2 Calculating the Hamiltonian

This script was written by ir. R.M.W. van Bijnen.

The function `calcMatrices` has as its input the two Rabi frequencies of a three-level system: 'Omega01' and 'Omega12'. Besides that, it requires as input the vector 'r', which denotes the position of all the atoms in the excitation volume. It returns:

- 'Hexc1' - The number of atoms in state 1 (vector of diagonal elements)
- 'Hexc2' - The number of atoms in state 2 (vector of diagonal elements)
- 'Hint' - Interaction energies (vector of diagonal elements)
- 'Hc01' - Couplings between level 0 and 1
- 'Hc02' - Couplings between level 1 and 2

```matlab
function [Hexc1, Hexc2, Hint, Hc01, Hc12] = calcMatrices(Omega01, Omega12, r)

% The final Hamiltonian is given by
% H = -diag(Hexc1) * Delta1 - diag(Hexc2) * Delta2 + diag(Vint)
% + Omega01 * Hc01 + Omega12 * Hc12

% Get the number of particles
N = length(Omega01);

% The number of states and number of couplings
Ns = 3^N;
Nc = 2 * 3^(N-1) * N;

% pre-allocate index and value arrays of the coupling matrices
colindex01 = zeros(Nc, 1);
colindex12 = zeros(Nc, 1);
rowindex01 = zeros(Nc, 1);
rowindex12 = zeros(Nc, 1);
coupling01 = zeros(Nc, 1);
coupling12 = zeros(Nc, 1);

% pre-allocate the interaction energies and excitations
Hexc1 = zeros(Ns, 1);
Hexc2 = zeros(Ns, 1);
Hint = zeros(Ns, 1);
```

% counters for the number of couplings
Nc01 = 0;
Nc12 = 0;

82
% compute the pairwise interactions between all particles
rrx = bsxfun(@minus, r(:, 1), r(:, 1))' + eye(N, N);
rry = bsxfun(@minus, r(:, 2), r(:, 2))';
rrz = bsxfun(@minus, r(:, 3), r(:, 3))';
rr = rrx .^ 2 + rry .^ 2 + rrz .^ 2;
pairint = (1 ./ rr) .^ 3;

% fill the matrix, by looping over all possible states
for k = 0 : (Ns - 1)
    % get the state (i.e. the digits of the ternary representation of k)
    state = getState(k, N, 3);
    % get the indices of all particles in the 2 state
    indices2 = (state == 2);
    % store the number of excitations in this state
    Hexc1(k + 1) = sum(state == 1);
    Hexc2(k + 1) = sum(indices2);

    % check each particle and in this state, see if there is a coupling
    % possible (and add it)
    for l = 1 : N
        % copy the state
        cstate = state;

        % check if there is a coupling
        if state(l) == 0
            % yes, there is a coupling from 0 to 1:
            % increase the number of couplings
            Nc01 = Nc01 + 1;
            % calculate the coupled state index, add it to the index list
            cstate(l) = 1;
            colindex01(Nc01) = getIndex(cstate, N, 3) + 1;
            rowindex01(Nc01) = k + 1;

        elseif state(l) == 1
            % yes, there is a coupling from 1 to 2:
            % increase the number of couplings
            Nc12 = Nc12 + 1;
            % calculate the coupled state index
            cstate(l) = 1;
            colindex01(Nc01) = getIndex(cstate, N, 3) + 1;
            rowindex01(Nc01) = k + 1;

    end
end
cstate(l) = 2;
colindex12(Nc12) = getIndex(cstate, N, 3) + 1;
rowindex12(Nc12) = k + 1;

% calculate the coupling strength
coupling12(Nc12) = Omega12(l);

% see if the coupled state already has a known interaction
% energy
if Hint(colindex12(Nc12)) == 0
% no, calculate it from the current state’s energy by
% adding the pairwise interactions of the new particle
% with the particles in the old state
    Hint(colindex12(Nc12)) = Hint(k + 1)
    + sum(pairint(1, indices2));
end
end
end

% copy the indices and couplings to make the matrix symmetric
colindex01((Nc01 + 1) : end) = rowindex01(1 : Nc01);
rowindex01((Nc01 + 1) : end) = colindex01(1 : Nc01);
coupling01((Nc01 + 1) : end) = conj(coupling01(1 : Nc01));
Nc01 = 2 * Nc01;
colindex12((Nc12 + 1) : end) = rowindex12(1 : Nc12);
rowindex12((Nc12 + 1) : end) = colindex12(1 : Nc12);
coupling12((Nc12 + 1) : end) = conj(coupling12(1 : Nc12));
Nc12 = 2 * Nc12;

% create the coupling matrices
Hc01 = sparse(rowindex01(1 : Nc01), colindex01(1 : Nc01),
coupling01(1 : Nc01), Ns, Ns);
Hc12 = sparse(rowindex12(1 : Nc12), colindex12(1 : Nc12),
coupling12(1 : Nc12), Ns, Ns);
end
C.3 A time-evolve function

This script was written by ir. R.M.W. van Bijnen

In this appendix we present an alternative differential equation solver. The widely
used Runge-Kutta Matlab function 'ode45' did not correspond to our expectations, so
we defined a new one. This became the function 'timeEvolve'. Offering to this function
a wave function at a time \( t_{\text{min}} \), timeEvolve calculates the wave function at the time
\( t_{\text{max}} \). The input parameters are:

- 'tmin' - the starting time
- 'tmax' - the ending time
- 'Delta01' - the detuning of the intermediate level
- 'Delta12' - the detuning of the Rydberg level
- 'Omega01' - coupling strength between \( |e\rangle \) and \( |g\rangle \)
- 'Omega12' - coupling strength between \( |r\rangle \) and \( |e\rangle \)
- 'Psi0' - the initial state of the system
- 'Hexc1' until 'Hc12' - the parts of the Hamiltonian as calculated in 'calcMatrices'

function [Psi, t] = timeEvolve(tmin, tmax, Delta01, Delta12, Omega01, Omega12,
Psi0, Hexc1, Hexc2, Hint, Hc01, Hc12)

% Get a measure for the order of magnitude of the elements of Psi (for the
% error calculation and step adjustment)
err0 = max(abs(Psi0));

% The Cash-Karp Runge Kutta Butcher tableau
a = [0, 1/5, 3/10, 3/5, 1, 7/8];
b = [0, 0, 0, 0, 0, 0; 1/5, 0, 0, 0, 0, 0; 3/40, 9/40, 0, 0, 0, 0; 3/10, -9/10, 6/5, 0, 0, 0; -11/54, 5/2, -70/27, 35/27, 0, 0; 1631/55296, 175/512, 575/13824, 44275/110592, 253/4096, 0];

% The stencil factors for the fourth and fifth order scheme
c4 = [2825/27648, 0, 18575/48384, 13525/55296, 277/14336, 1/4];
c5 = [37/378, 0, 250/621, 125/594, 0, 512/1771];

% Initialize the array for time values
t = tmin;
h0 = 1e-6;
reitol = 1e-8;
Psi = Psi0;

% Calculate the (diagonal) matrix due to the detunings and interactions
Hdiag = spdiags(-Hexc1 * Delta01(tmin) - Hexc2 * 
(Delta01(tmin) + Delta12(tmin)) + Hint, 
0, length(Hexc1), length(Hexc1));

% Calculate Psi until the final detuning value is reached 
while t(end) < tmax 
    % Initialize step size larger than initial step size to initiate the 
    % while-loop below 
    h = 2 * h0; 
    numsteps = 0; 
    % Make sure we don’t step over the end of the time interval 
    if t(end) + h0 >= tmax 
        h0 = tmax - t(end); 
    end 
    % Keep taking teststeps of decreasing size until the step is accepted 
    while h0 < h 
        % Use the estimate for the stepsize as the actual stepsize 
        h = h0; 

        % Keep track of the number of refinements 
        numsteps = numsteps + 1; 

        % Calculate the RK 'k' values 
        k = zeros(length(Psi), length(b(1, :))); 
        for i = 1 : length(b(1, :)) 
            ti = t(end) + a(i) * h; 
            Hdiag = spdiags(-Hexc1 * Delta01(ti) - Hexc2 * 
(Delta01(ti) + Delta12(ti)) + Hint, 0, Hdiag); 
            k(:, i) = -1i * (0.5 * Omega01(ti) * Hc01 + 0.5 * Omega12(ti) * Hc12 + Hdiag) 
* (Psi + h * k(:, 1 : i) * b(i, 1 : i)'); 
        end 
        % Get the new estimate for Psi and error estimate vector 
        Psi4 = Psi + h * k * c4; 
        Psi5 = Psi + h * k * c5; 
        dPsi = Psi5 - Psi4; 
        err = max(abs(dPsi)) / err0; 
        if err < 1e-14 
            h0 = 10 * h; 
        else 
            h0 = h * abs(reltol / err).^0.2; 
        end 
    end 
end 
% Store the newly calculated 5th order estimate of Psi + time point 
Psi = Psi5; 
t = [t, t(end) + h];
C.4 Calculating the wave function

This script was written by ir. R.M.W. van Bijnen

In this function 'runSim' we use all previous function in order to calculate the wavefunction of the system as a function of time, when subjected to STIRAP procedure. The output parameters are the wave function as a function of time 'Psit' and the points in time at which the wavefunction is calculated 'tt'. The input parameters are:

- 'N' - the number of atoms
- 'tdiff' - the time delay between the two Gaussian laser pulses
- 'tmin' and 'tmax' - begin and ending time of the simulation
- 'Delta01amp' - the detuning from the intermediate state
- 'fmax01' - the strength of the Pump laser in MHz
- 'fmax12' - the strength of the Stokes laser in MHz
- 'plotresults' - if one: plot graphs

function [Psit, tt] = runSim(N, tdiff, twidth, tmin, tmax, Delta01amp, fmax01, fmax12, plotresults)

% Planck's constant
hbar = 1.05457 * 1e-34;
% C_6 in J m^6 of the n=65 s Rydberg state
C6 = 2.4 * 1e-58;

% the system size in micrometers
a = 10;
R = a * 1e-6;
% the particle positions; set other two dimensions to zero
r = [0 : 0.5 : 1]';
r = [r, zeros(N, 2)];

% E0 is an energy scale we use to scale all our energies with, and we take % it equal to the interaction energy of a pair of atoms sitting at opposite % ends of our 1D system of size R
E0 = C6 / R^6;

% our frequencies are expressed in units of Omega0
Omega0 = E0 / hbar;

% one MHz is equal to 2pi * 10^-6 / Omega0 of our dimensionless units
MHz = 2 * pi * 1e6 / Omega0;
% our time scale is equal to \( \hbar / E_0 \) seconds
t0 = \( \hbar / E_0 \);

% which is equal to \( 10^6 \) t0 microseconds
tmus = t0 * 1e6;

% Scale all times
t01 = twidth / tmus;
t12 = twidth / tmus;
tdiff = tdiff / tmus;
tmin = tmin / tmus;
tmax = tmax / tmus;

% the peak intensity of the laser pulses
Omax01 = fmax01 * MHz;
Omax12 = fmax12 * MHz;

% the detunings
Delta01 = @(t) Delta01amp * MHz;
Delta12 = @(t) -Delta01(t);

% Laser pulses: a smooth sequence with two gaussian pulses
Omega01 = @(t) Omax01*(exp(-0.5 * t .^2 / t01^2)
   + exp(-0.5 * (t - 3*tdiff).^2/t01^2));
Omega12 = @(t) Omax12*(exp(-0.5 * (t + tdiff) .^2 / t12^2)
   + exp(-0.5 * (t -4*tdiff) .^2 / t12^2));
Omega01r = @(rr) ones(length(r(:, 1)));
Omega12r = @(rr) ones(length(r(:, 1)));

if plotresults == 1
    % plot the pulses
    tt = tmin : (tmax - tmin) / 1024 : tmax;
    figure;
    hold on;
    plot(tt * tmus, Omega01(tt) / MHz, 'r-', 'Linewidth', 3);
    plot(tt * tmus, Omega12(tt) / MHz, 'b-', 'Linewidth', 3);
    xlabel('t [\mu s]');
    ylabel('\Omega [MHz]');
    title('Laser pulses')
end

% Calculate the matrices that make up the Hamiltonian
[Hexc1, Hexc2, Hint, Hc01, Hc12] = calcMatrices(Omega01r(r), Omega12r(r), r);

% Initialize Psi with unit amplitude in ground state
Psi0 = zeros(length(Hint), 1);
Psi0(7, 1) = 1;

% Create an array of time points, at which we store the state vector and
% the eigenvector we want to match
Nt = 128;
tt = tmin : (tmax - tmin) / (Nt - 1) : tmax;

% Create arrays for the state vector and eigenvector
Psit = zeros(3^N, length(tt));
Psit(:, 1) = Psi0;

% run the simulation!
for k = 1 : (length(tt) - 1)
    % evolve the system for one timestep
    [Psi, abcd] = timeEvolve(tt(k), tt(k + 1), Delta01, Delta12, Omega01,
                             Omega12, Psit(:, k), Hexc1, Hexc2, Hint,
                             Hc01, Hc12);
    % store the state vector
    Psit(:, k + 1) = Psi;
end

Psit

if plotresults == 1
    % Define particle states
    allground = zeros(1, length(tt));
    oneexcited = zeros(1, length(tt));
    twoexcited = zeros(1, length(tt));
    allexcited = zeros(1, length(tt));

    for k = 1 : 3^N
        % calculate the single particle states for this many-body state
        state = getState(k - 1, N, 3);

        % get the logical indices of particles in states 0, 1, and 2
        i0 = (state == 0);
        i1 = (state == 1);
        i2 = (state == 2);

        % Calculate total wavefunctions for 0, 1, 2, 3 Rydbergs
        if i0(1) + i0(2) + i0(3) == 3
            allground = abs(Psit(k,:)).^2;
        elseif i2(1) + i2(2) + i2(3) == 1
            oneexcited = oneexcited + abs(Psit(k,:)).^2;
        end
    end
end
```matlab
elseif i2(1) + i2(2) + i2(3) == 2
    twoexcited = twoexcited + abs(Psit(k,:).^2);
elseif i2(1) + i2(2) + i2(3) == 3
    allexcited = abs(Psit(k,:)).^2;
end

%Plot probabilities of getting 0,1,2 and 3 Rydberg atoms.
figure;
hold on;
plot(tt * tmus, allground(1,:), 'r-', 'Linewidth', 3);
plot(tt * tmus, oneexcited(1,:), 'b-', 'Linewidth', 3);
plot(tt * tmus, twoexcited(1,:), 'y-', 'Linewidth', 3);
plot(tt * tmus, allexcited(1,:), 'g-', 'Linewidth', 3);
xlabel('t [\mu s]');
ylabel('Probability');
end
```
C.5 Solving the optical Bloch equations

In this script we use a Runge-Kutta method to solve the coupled system of optical Bloch equations (OBE). We start with a function which defines the OBE for a given spontaneous emission rate $\gamma = g$, a given Rabi frequency $\Omega = \omega$ and a given detuning $\Delta = \delta$:

```matlab
function drho = DV(t, rho, g, omega, delta)
    % Define empty differential vector
    drho = zeros(4,1);
    % Define coupled optical Bloch equations
    drho(1) = -g*rho(1) + 1i/2*(omega*rho(3) - omega*rho(2));
    drho(2) = -(g/2 + 1i*delta)*rho(2) + 1i*omega/2*(rho(4) - rho(1));
    drho(3) = -(g/2 - 1i*delta)*rho(3) + 1i*omega/2*(rho(1) - rho(4));
    drho(4) = g*rho(1) + 1i/2*(omega*rho(2) - omega*rho(3));
end
```

We then solve the OBE in the following script:

```matlab
% In this script we solve a two level system, in which spontaneous emission occurs.

% Input parameters
    tmin = 0;
    tmax = 18;
    g = 1;
    omega = 2.5 * g;
    delta = 0;

    % The initial condition: atom starts in ground state
    y0 = [0 0 0 1];
    f1 = @(t,rho) DV(t, rho, g, omega, delta);

    % Options for ode45-solver
    options = odeset('RelTol',1e-6,'AbsTol',1e-6);

    % Use Runge-Kutta method to solve system
    [ a,b ] = ode45(f1, [tmin tmax], y0, options);

    % Plot solution
    figure;
    hold on;
    plot(a, abs(b(:,1)), 'k-', 'Linewidth', 3)
xlabel('Time ($\gamma^{-1}$)');
ylabel('\rho_{ee}');
```

91
### C.6 Calculating eigenvectors and eigenvalues of the Hamiltonian

This script is based on the script by ir. R.M.W. van Bijnen for the case $N=2$.

In this script we calculate the instantaneous eigenvalues and eigenvectors of a three-level Hamiltonian for a system with $N$ particles on a line, where we used a STIRAP scheme for the course of the Rabi frequency. The output parameters are the final probability of the $|rg\rangle$ state, the final probability of the $|grg\rangle$ state and the maximum probability of a state that contains atoms in $|e\rangle$. All parameters are scaled with $\Omega_{P0}$, so an input of $\Delta_1 = -1$ is equivalent with $\Delta_P = -\Omega_P$. The input parameters are:

- 'N' - the number of atoms
- 'Delta1' - the detuning from the intermediate state $\Delta_P$
- 'Deltatot' - the total detuning $\Delta_P + \Delta_S$
- 'Vint' - The van der Waals interaction parameter
- 'plotresults' - if one: plot the eigenvectors graphs
- 'fast' - if 1, the script will only calculate the eigenvectors which start in the required state

```matlab
function [rgrend, grgend, epop] = plotNParticleEigenvectors(N, Delta1, Deltatot, Vint, plotresult, fast)

% Initialise parameters
rgrend = 0;
grgend = 0;
epop = 0;

% Choose the component in which we want the eigenvector to start
startcomp = 1;

% Get the pieces of the Hamiltonian
Omega01vector = ones(1, N);
Omega12vector = ones(1, N);

% Positions of the particles
rx = [0 : 0.5 : 0.5*(N-1)]';
r = [rx, zeros(N, 2)];

% Calculate Hamiltonian pieces
[Hexc1, Hexc2, Hint, Hc01, Hc12] = calcMatrices(Omega01vector, Omega12vector, r);

% Define two laser pulses for the STIRAP sequence.
```
% Amplitude of Omega01 is equal to 1, since we will
% only consider the parameters Delta/Omega and V_int/Omega
% etc. Time scale in a.u.
Omega01 = @(t) exp(-0.5 * (t).^2);
Omega12 = @(t) exp(-0.5 * (t+2).^2);

% Create an array of time points
tt = -2.3 : 0.01 : 2;

% Create option to plot the pulses
if plotresult == 2;
    figure;
    hold on;
    plot(tt,Omega01(tt),'r-')
    plot(tt,Omega12(tt),'b-')
end

% Preallocate arrays to keep track of the eigenvectors and eigenvalues
Vv = zeros(3 ^ N, length(tt));
Vprev = zeros(3 ^ N, 1);
eigenvalue = zeros(3^N, length(tt));

% Calculate each eigenvector and plot its components
for v = 1 : length(Vv(:, 1))
    % Loop over all time points
    for k = 1 : length(tt)
        % Piece together the Hamiltonian
        H = -diag(Hexc1) * Delta1 - diag(Hexc2) * Deltatot + diag(Vint*Hint) + 0.5 * Omega01(tt(k)) * Hc01 + 0.5 * Omega12(tt(k)) * Hc12;
        % get the eigenvectors and -values of H
        [V, D] = eig(full(H));

        % We are only interested in the eigenvector which starts in
        % startcomp, so if this is not the case, break (only works if
        % fast=1)
        if (k == 1)
            if abs(V(startcomp,v)^2) < 0.31 && fast == 1;
                break
            end
        % Initialise the tracking system with eigenvector nr. 'v'
        Vprev = V(:, v);
    end
end
% take inner products, to find the eigenvector we're tracking
ip = zeros(1, length(V(1, :)));
for vv = 1 : length(V(1, :))
    ip(vv) = abs(V(:, vv)' * Vprev);
end

% Find the maximum overlap, this must be the eigenvector that
% evolved from Vprev
[m, i] = max(ip);

% Copy the eigenvector and its eigenvalue to the storage array
Vv(:, k) = V(:, i);
eigenvalue(v,k) = D(i, i);

% Update the Vprev vector
Vprev = V(:, i);
end

% We are only interested in eigenvectors which start in startcomp
if abs(Vv(startcomp,1)^2) > 0.31 || fast == 0;

% Store maximum population of any state with |e>
for l = 1:3^N
    state = (getState(l-1,N,3)==1);
    if abs(state)~=0
        max_estate(l) = max(abs(Vv(l,:).^2));
    end
end
epop = max(max_estate);

% Create a figure for the eigenvector
if plotresult == 1;
    figure;
    title(sprintf('Vector: %i', v));
    hold on;
    for n = 1 : length(Vv(:, 1))
        % plot all its components in a different color
        c = (n - 1) / (length(Vv(:, 1)) - 1);
        plot(tt, abs(Vv(n, :)) .^2, '-', 'Color',
            [c, 0, 1 - c], 'Linewidth', 3);
    end
    xlabel('t (a.u.)');
% String array to help labeling the curves
statelabel = ['g', 'e', 'r'];

% Find a suitable timepoint at which we will add labels to the curves
ti = find(tt >= -1.4, 1);

% Loop over all the components of the eigenvector
for n = 1 : length(Vv(:, ti));

    % we are going to add spacing if there are multiple components
    % with the same value, let's start with one space
    space = ' ';

    % find the number of equal-valued components
    numprev = find(abs(abs(Vv(1 : (n - 1), ti))
            - abs(Vv(n, ti))) < 1e-2);

    % for each equal-valued component, we add 4N spaces
    for k = 1 : (length(numprev) * 4*N)
        space = sprintf('%s ', space);
    end

    % plot color helper variable
    c = (n - 1) / (length(Vv(:, 1)) - 1);

    % get the single-particle states associated with the n-th basis
    % vector
    state = getState(n - 1, N, 3);

    % print a label for the n-th basis vector
    s = sprintf('\leftarrow %s [\%s, \%s, \%s]', space,
            statelabel(state(1) + 1), statelabel(state(2) + 1),
            statelabel(state(3) + 1));

    % add the label at the position of the curve
    text(tt(ti), abs(Vv(n, ti)).^2, s, 'color', [c, 0, 1-c]);

end
end

% Clear Vv(startcomp,1) to prevent multiple plots
Vv(startcomp,1)=0;
end
end

% plot the eigenvalues
figure;
hold on;
for k = 1 : 3^N
    plot(tt,eigenvalue(k, :));
C.7 Quantum Monte Carlo

In this script we have implemented the quantum Monte Carlo algorithm. In it, we put \( N \) particles on a line, with a distance of 5 \( \mu m \) between adjacent particles. The total system size is \( 5(N - 1) \mu m \). We scale all relevant parameters in the characteristic energy \( E_0 \), as used before in the 'runSim' simulation. The structure of the script is the following:

- Define parameters
- Calculate non-Hermitian evolution matrix as proposed in discussion QMC algorithm
- Calculate 'normal' interaction and detuning matrices
- Time-evolve system using Monte-Carlo procedure
- Plot the obtained results.

However, first we define a function which returns the matrix representation of the transition operators \( C_m \). The function takes as input the number of levels \( M \), the number of atoms \( N \), the spontaneous emission rate \( gamma \) and the number of the level from which spontaneous emission occurs \( l \). The output consists of a cell \( Carray \) which contains the individual \( C_m \)'s and a matrix \( CCtot \) which contains the sum over the products \( C_m C_m^\dagger \).

```matlab
function [CCtot, Carray] = DecayMatrix(M, N, gamma, l)
% Define empty matrix and empty cell
CCtot = zeros(M ^ N, M ^ N);
Carray = cell(1, N);

% Initialize numexc
numexc = 0;

% First calculate the total decay matrix
for k = 1 : M ^ N
    % Get the k-1-state
    state = getState(k - 1, N, M);

    % How many atoms in |e>?
    numexc = sum(state == l);

    % Put the number of atoms in |e> which may decay
    % on the k-th diagonal
    CCtot(k,k) = CCtot(k,k) + numexc;
end

% Multiply matrix by gamma
CCtot = CCtot * sqrt(gamma);
```

end
% Then calculate the separate C_m’s

for a = 1 : N
    % Initialize counter and help parameters
    count = 0;
    A = zeros(M ^ N, M ^ N);
    stateground = zeros(N,1);
    stateexcited = zeros(N,1);

    for b = 1 : M ^ N
        % Get the k-1-state
        state = getState(b-1, N, M);

        % Check if n-th atom is in ground-state
        if state(a,1) == 0
            % yes: save state
            stateground = state;
        else
            % No: continue (Note: continue makes sure rules underneath are not read and loop goes to b = b+1)
            continue
        end

        % Get the corresponding excited state
        stateexcited = stateground;
        stateexcited(a,1) = 1;

        % Get indices
        x = getIndex(stateexcited, N, M) + 1;
        y = getIndex(stateground, N, M) + 1;

        % Put decay-one in matrix and add 1 to counter
        A(y, x) = 1;
        count = count + 1;
    end

    Carray{1,a} = sqrt(gamma) * A;
end
end

The full quantum Monte Carlo script:

% First: define all parameters
% Number of particles (N), number of levels (M) and
% number of times to run (z):

N = 3;
M = 3;
z = 10;
% Choose running times in microseconds
  tmin = -30;
  tmax = 20;
  tstep = 0.001;
  twidth = 5;
  tdiff = 10;
  Delta01amp = -10 / (2 * pi);
  fmax01 = 10 / (2 * pi);
  fmax12 = fmax01;

% Planck’s constant and C_6 in J m^6 of the n=65s Rydberg state
  hbar = 1.05457 * 1e-34;
  C6 = 2.4 * 1e-58;
  gamma = 5.98;

% The system size in micrometers R;
  a = 10;
  R = a * 1e-6;

% The particle positions; set other two dimensions to zero
  r = [0 : 0.5 : 1]’;
  r = [r, zeros(N, 2)];

% E0 is an energy scale we use to scale all our energies with,
% and we take it equal to the interaction energy of a pair
% of atoms sitting at opposite ends of our 1D system of size R
  E0 = C6 / R^6;

% Our frequencies are expressed in units of Omega0
  Omega0 = E0 / hbar;

% One MHz is equal to 2pi*10^6/Omega0 of our dimensionless units
  MHz = 2 * pi * 1e6 / Omega0;

% Our time scale is equal to hbar / E0 seconds
  t0 = hbar / E0;

% which is equal to 10^6 t0 microseconds
  tmus = t0 * 1e6;

% Scale all times
  t01 = twidth / tmus;
  t12 = twidth / tmus;
  tdiff = tdiff / tmus;
  tmin = tmin / tmus;
  tmax = tmax / tmus;
tstep = tstep / tmus;

% The peak intensity of the laser pulses
Omax01 = fmax01 * MHz;
Omax12 = fmax12 * MHz;

% Scale gamma
gamma = gamma * MHz;

% The detunings
Delta01 = @(t) Delta01amp * MHz;
Delta12 = @(t) -Delta01(t);

% Define lasers pulses: two Gaussians
Omega01 = @(t) (exp(-0.5 * t .^2 / t01^2)) * Omax01;
Omega12 = @(t) (exp(-0.5 * (t + tdiff) .^2 / t12^2)) * Omax12;

% Create vector with time points:
tt = tmin : tstep : tmax;

% Plot laser pulses
figure;
hold on;
plot(tt * tmus , Omega01(tt) / MHz, 'r-', 'Linewidth', 3);
plot(tt * tmus , Omega12(tt) / MHz, 'b-', 'Linewidth', 3);
xlabel('t [\mus]');
ylabel('
Omega [MHz]');
title('Laser pulses')
legend('Pump laser pulse','Stokes laser pulse',
2, 'Location', 'NorthEast');

% Second: calculate non-Hermitian decay matrix, the '1' makes sure that
% decay takes place from |e>
[CCtot, Carray] = DecayMatrix(M, N, gamma, 1);

% Third: calculate 'normal' Hamiltonian for the system

% Calculate the matrices that make up the interaction Hamiltonian
[Hexc1, Hexc2, Hint, Hc01, Hc12] =
calcMatrices(ones(N,1)', ones(N,1)', r);

% Calculate the (diagonal) xxmatrix due to
% the detunings and interactions
Hdiag = spdiags( -Hexc1 * Delta01(tmin)
+ Hexc2 * (Delta01(tmin) + Delta12(tmin))
+ Hint, 0, length(Hexc1), length(Hexc1) );
% Full Hamiltonian with decay matrix
Hamiltonian = @(t) 0.5 * Omega01(t) * Hc01 + 0.5 * Omega12(t) * Hc12 - 1i * (1 / 2) * CCtot + Hdiag;

% Fourth: time-evolve the non-Hermitian total matrix

% Define state vector of system
psi = zeros(M ^ N, length(tt));

% Define initial state vector of system: starting in full ground-state
psi0 = zeros(M ^ N, 1);
psi0(1,1) = 1;
psi(:,1) = psi0;

% Define initial norm
norm = 0;

% Define empty vector for all, which shall hold the average result
psitot = zeros(M ^ N, length(tt));

% Run simulation!
for j = 1 : z

% Create vector to store probabilities
dp = zeros(N,1);

for k = 2 : 1 : length(tt)

% Time-evolve the vector
[Psi, t] = timeEvolveMC3( tt(k-1), tt(k), psi(:, k-1), Hamiltonian );

% Calculate dp_m’s as defined in article Molmer.
% Note that Carray(1,i) holds C_i
for i = 1 : N
dp(i,1) = abs( tstep * psi(:, k-1)' * Carray{1,i}' * Carray{1,i} * psi(:, k-1) );
end

% Sum to get whole dp
dptot = sum( dp(:,1) );

% Calculate the norm
norm = sqrt(Psi' * Psi);

% Generate random number between 0 and 1
eps1 = rand(1);

end
if dptot <= eps1
  % No quantum jump
  psi(:,k) = Psi / norm;
else % A quantum jump occured
  for h = 1 : N
    % Check: is the sum of dp_1 until dp_h larger
    % than eps?
    if eps1 < sum(dp(1 : h, 1))
      %yes: decay
      psi(:,k) = Carray{1,h} * Psi;
      norm2 = sqrt( psi(:,k)' * psi(:,k) );
      psi(:,k) = psi(:,k) / norm2;
      fprintf('C%i acted at t = %.4f\n', h, tt(k));
      break
    else
      % No: h = h + 1
      continue
    end
  end
end
% Update average vector
psitot = psitot + abs(psi).^2;
end

% Fifth: make a plot
figure;
hold on;
plot(tt*tmus, psitot(1,:)/z, 'r-', 'Linewidth', 3);
plot(tt*tmus, psitot(7,:)/z, 'y-', 'Linewidth', 3);
plot(tt*tmus, psitot(21,:)/z, 'b-', 'Linewidth', 3);
plot(tt*tmus, psitot(27,:)/z, 'k-', 'Linewidth', 3);
plot(tt*tmus, psitot(3,:)/z, 'g-', 'Linewidth', 3);
plot(tt*tmus, psitot(19,:)/z, 'c-', 'Linewidth', 3);
xlabel('Time (\mus)');
ylabel('\langle\psi(1,2,3)|\psi\rangle');
legend('|ggg>','|grg>','|rgr>','|rrr>','|rgg>','|ggr>',
6,'Location','NorthEast');