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Collective State Representation of Atoms in Quantum Computing and
Precision Metrology

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ABSTRACT

Collective State Representation of Atoms in Quantum Computing and Precision Metrology

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When $N$ non-interacting atoms interact with a single frequency laser with no phase difference between the photons interacting with the atoms, their interaction can be described collectively \[1\]. For instance, suppose that there is a two level atom with state $|\Psi_1\rangle = \alpha_1|a\rangle + \beta_1|b\rangle$, and another two level atom with state $|\Psi_2\rangle = \alpha_2|a\rangle + \beta_2|b\rangle$. We assume that the two internal states $|a\rangle$ and $|b\rangle$ are indistinguishable between the atoms. Since they are non-interacting atoms, the total state of the system with the two atoms is $|\Psi\rangle_C = \alpha_1\alpha_2|aa\rangle + \alpha_1\beta_2|ab\rangle + \beta_1\alpha_2|ba\rangle + \beta_1\beta_2|bb\rangle$. By rotating the states $|ab\rangle$ and $|ba\rangle$, we can redefine the system using two new basis states, $|+\rangle = (|ab\rangle + |ba\rangle)/\sqrt{2}$ and $|−\rangle = (|ab\rangle − |ba\rangle)/\sqrt{2}$. The state of the system with these states is $|\Psi\rangle_C = \alpha_1\alpha_2|aa\rangle + (\alpha_1\beta_2 + \beta_1\alpha_2)/\sqrt{2}|+\rangle + (\alpha_1\beta_2 − \beta_1\alpha_2)/\sqrt{2}|−\rangle + \beta_1\beta_2|bb\rangle$. If the two atoms interact with the same field, they evolve in the same way, so that $\alpha_1 = \alpha_2 \equiv \alpha$ and $\beta_1 = \beta_2 \equiv \beta$. Hence, the $|−\rangle$ state, which is the antisymmetric state, vanishes, and only the symmetric states remain in the system, so that the total state of the system can be described by $|\Psi\rangle_C = \alpha^2|aa\rangle + \sqrt{2}\alpha\beta|+\rangle + \beta^2|bb\rangle$. The remaining states are what are known as the symmetric Dicke states, symmetric collective states, or collective spin states. This two atom case can be generalized to $N$ atoms; for $N$ atoms, there are $N + 1$ symmetric collective states. They have been studied since Dicke’s seminal paper in the 1950s, especially with respect to superradiance \[2, 3, 4, 5, 6, 7\] and squeezed states \[7, 8, 9, 10\].

We first studied the symmetric collective states for the purpose of quantum computing. Using Rydberg atoms, we showed that with the proper choice of experimental parameters, the excitation of the collective states can be confined to just the ground state and the first excited state by way of differential light shifts. We called this the Rydberg assisted
light shift imbalance induced blockade. Such a two level system is important in quantum computing because it can be used as a qubit, a building block of quantum computers. Although the collective state description of Rydberg atoms is quite complicated, since it requires more than just the two traditional hyperfine ground states of an alkali atom, we were able to successfully simplify the system and find the conditions necessary for the proper light shifts to occur to our advantage. The simulations supported our results and we published the results [11].

We then moved on to study whether the collective states could be used to make atomic clocks and interferometers. In the case of a collective state atomic clock (COSAC), we found that the Ramsey fringes narrowed by a factor of $\sqrt{N}$ compared to a conventional clock – $N$ being the number of non-interacting atoms – without violating the uncertainty relation. This narrowing is explained as being due to interferences among the collective states, representing an effective $\sqrt{N}$ fold increase in the clock frequency, without entanglement. We discuss the experimental inhomogeneities that affect the signal and show that experimental parameters can be adjusted to produce a near ideal signal. The detection process collects fluorescence through stimulated Raman scattering of Stokes photons, which emits photons predominantly in the direction of the probe beam for a high enough optical density. By using a null measurement scheme, in which detection of zero photons corresponds to the system being in a single collective state, we detect the population in a collective state of interest. The quantum and classical noise of the ideal COSAC is still limited by the standard quantum limit and performs only as well as the conventional clock. However, when detection efficiency and collection efficiency are taken into account,
the detection scheme of the COSAC increases the quantum efficiency of detection significantly in comparison to a typical conventional clock employing fluorescence detection, yielding a net improvement in stability by as much as a factor of 10. For the off-resonant Raman excitation based COSAC, the theory and results from simulations were published together [12]; the experiment is underway, and we hope to publish the results in a few months. The COSAC can also be described in terms of the coherent population transfer (CPT) states. The theory behind it is being polished and will be published soon. The collective state atomic interferometer is also possible, with similar inhomogeneities being present in such system, as well [13].

This dissertation is organized as follows: In Chapter 1, the fundamental atomic interactions with the electric field and magnetic field are used to derive the interaction Hamiltonian and the density matrix formulation. Chapter 2 comprises of the theoretical work and simulation results regarding Rydberg assisted light shift imbalance induced blockade. This chapter introduces collective states. For a more thorough investigation into these states, recommended reading includes Dicke’s seminal paper [11], and other references [14, 15]. Chapter 3 discusses the off-resonant Raman-Rabi excitation based COSAC, and Chapter 4 follows it up with the discussion on coherent population trapping based COSAC. In Chapter 5 I discuss the ongoing experimental progress and the preliminary results we have obtained thus far. I conclude in Chapter 6 with future work. Finally, some of the key programs used for simulations are included in the appendices. In Appendix A some of the MATLAB programs used in the evolution of the density matrix, and the steady state solution of the master equation, in Chapter 2 are included. In Appendix B the more sophisticated Python programs used for Chapter 3 are included.
Despite the rumors I have heard about no one actually reading anyone else’s dissertations, I hope that someone will find the information in here useful in the future.
Acknowledgements
If I had to work alone without the support that people around me gave me, I would not have been able to make it to this point in my career. I would like to thank my advisor, Professor Selim M. Shahriar, for believing in me when he hired me to work with him in the atomic lab. I was new to atomic physics and experimental work, but he gave me a chance to learn everything from scratch. I also want to thank him for his guidance and tough mentorship. My committee members, Professor John Ketterson and Professor Brian Odom, have also been extremely helpful in my journey as a graduate student. Just by being part of a collaboration with them, and by attending the weekly meetings, I learned more than I could have from reading up on my own. I respect Professor Ketterson for his hands-on approach; he tried to be in the lab with me and Jon Trossman when we were working in the IGERT (National Science Foundation Integrative Graduate Education and Research Traineeship) lab. Jon Trossman was in the same boat as I was when we started at Northwestern together, and we learned to build a vacuum chamber. I would like to thank him for all the good times. Professor Odom’s group members, David Taylor, Chien-Yu Lien, Yen-Wei Lin, Jason Nguyen, Joan Marler, Vaishnavi Rajagopal, Ming-Feng Tu and Zeke Tung, were extremely helpful. I went to them whenever I needed advice or a sounding board; and they were sometimes much needed familiar faces to eat lunch and dinner with at conferences.

When I first joined Professor Selim Shahriar’s Laboratory for Atomic and Photonic Technology in 2009, there was a 3 meter long vacuum chamber, with a chirp cooling arm and 30 A current supply for the magnetic coils, and no student or post doc who knew how it all worked. Ye Wang had taken some notes down from Ken Salit and Mary Salit before they left, but he was not the primary student on the project and did not know the
ins and outs. However, he, along with Mary and Ken via phone and email, helped me and Yanfei Tu, who later joined as a post doc in January 2010, helped us to get the apparatus to eventually do what we wanted it to do. We were able to actually make an atomic fountain in the huge chamber before we dismantled it. I would also like to thank Shih Tseng for his help throughout the years with the electronics. If we had problems with the tapered amplifier or an AOM, we would ask him for his help, and they would magically work within fifteen minutes of his working on it. Mohamed Fouda, besides building the Zeeman slower with me and Renpeng Fang, also helped out with any questions I had regarding electronics since he is an expert. I would like to thank him for his time and his patience with my ignorance.

For the actual work on the experiment, I would like to thank Daniel Villalon for working with Renpeng on the Labview program. We use it all the time to collect data. Jeff Sundwall and the crew at the Northwestern University Scientific Instrument shop made us our custom built flange holders and magnetic coil holders, among other things. I would like to thank them for their patience in trying to understand me, who has very little experience with machining anything. I especially want to thank Yanfei Tu and Renpeng for their dedication to the projects that we were on together. Yanfei helped check all my simulations regarding the light shift blockade, and worked with me on not only the huge vacuum chamber, but to build the smaller metal chamber. I always appreciated that whenever we had to deal with new equipment, she understood how to use it right away. She was also very quick at setting up optics and put my slow (and anal) progress to shame. Thank you for dealing with my obsessive compulsive disorder. Out of my group members, Renpeng has been the most helpful. Even though we only really worked together for the
last year or so, he is a fast learner, and a hard worker. He was like clockwork - always arriving at school between 9 and 9:30 everyday. He always stayed as late as he could for the sake of our experiment. He also took the data while I have been writing my thesis; the data that I am including in this dissertation would not be present without Renpeng. I am most grateful to him.

When we got into sticky situations and did not know how to get out of them, we were lucky in that Selim had collaborators who knew a lot about our work than I did. I would like to thank Professor Phil Hemmer at Texas A&M, who visited us periodically to check up on our progress and to lend us his hand in the lab. I would also like to express my deep gratitude to Dr. Frank Narducci at Naval Air Systems Command. Last year around this time, our experiment was in deep trouble and we did not know what was wrong. Frank welcomed me to visit his lab in Patuxent River, MD. When I forgot to bring my passport, he even used his powers to get me in through the gates. From then until now, he has continued to share with us all he knows and has been an amazing mentor. Our weekly collaboration meetings have been so helpful that I am certain beyond any doubt that we would not have any experimental results now if not for his mentorship.

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\[ |a\rangle \equiv |F = 2, m_f = 0\rangle, |b\rangle \equiv |F = 3, m_f = 0\rangle, |c\rangle \equiv |F' = 2, m_{f'} = 1\rangle, \]

\[ |d\rangle \equiv |F' = 3, m_{f'} = 1\rangle, \text{ and } |e\rangle \equiv |F' = 4, m_{f'} = 1\rangle. \]

The states \( |a\rangle \) and \( |b\rangle \) are the lower legs of the Λ transition, and the states \( |c\rangle \) and \( |d\rangle \) form an effective single excited level in the Λ transition. The state \( |e\rangle \) is brought into discussion because the Raman beams of frequency \( \omega_1 \) and \( \omega_2 \) are brought from a laser that is locked 90 MHz away from the \( |b\rangle \rightarrow |e\rangle \) transition. The values of \( \delta_c \) and \( \delta_d \) are discussed in the main text.

5.2 Optical paths of the MOT beams and the optical pumping beam.

Laser 1 is Toptica TA PRO 100; Laser 2 is Toptica TA 100; D1 to D4 are Thorlabs PD36A; AOM 1 is Isomet 80 MHz AOM, AOM 2 is IntraAction 80 MHz AOM; and AOM 3 is IntraAction 40 MHz AOM.

FC stands for fiber coupler; PBS is polarized beam splitter; QWP is quarter wave plate; HWP is half wave plate; and Rb is the rubidium vapor cell from Triad Technology that includes mixed species of rubidium.

5.3 Optical paths of the off-resonant Raman beams. Laser 1, AOM 1, D1, D2, and Rb are the same as in the previous figure. The Raman beams come from the same laser as the MOT beams. The Raman beams go through polarizers, marked P in the figure, and then through AOM 4 and AOM 5, which are Brimrose GPF-1500-200-630 models. They have efficiency of well over 15% when they are at their best.
A non-polarizing beam splitter, NPBS, combines the two Raman beams, dumps half the power, and send the other half through a fiber coupler, FC, to ensure that the beams are co-propagating. They then go through a tapered amplifier (after an Electro Optic Tech Isolator not drawn in the figure), and then through a switching IntraAction 40 MHz AOM, AOM 6. After a two lens system to enlarge the beams, they combine with the MOT-z beam and go through the cell.

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varied while current stayed fixed. In the bottom plots, the distance
was fixed while the current was varied.

5.7 Diagrams of switching circuit for (a) MOT coils; (b) Bias coils.

Although they were originally designed for another set of coils, where
the voltage supplied is around 60 V and 5 V, respectively, they work
for the coils designed in Fig. 5.5 as well.

5.8 A snapshot of the switching circuit for the bias coils.

5.9 Magnetic field decay test performed with the MOT coils in the metal
chamber, before we migrated to the glass cell. It can be seen that,
even with lower current, the decay time does not shorten all that
much; it remains at around 30 ms. The eddy current in the metal
chamber is to blame.

5.10 The first vacuum chamber that we ran experiments on in the lab.

5.11 The second vacuum chamber that required a smaller, 150 l/s ion
pump and had the MOT coils fit outside the chamber.

5.12 Another view of the second vacuum chamber

5.13 Single run result of single Raman pulse experiment in the MOT
chamber depicted in Fig. 5.11.

5.14 Averaged result of single Raman pulse experiment in the MOT
chamber depicted in Fig. 5.11.

5.15 The final vacuum chamber that we ended up with, which is a 1 cm by
1 cm by 4 cm experimental chamber with a turbo pump and a 2 ml/s
Gamma Vacuum ion pump. The feedthroughs are attached in series to rubidium getters inside the chamber.

5.16 The glass cell vacuum system with the coils and the optics around it.

5.17 A side view of the glass cell.

5.18 A view of the optics table from far away to show the setup from far away. The turbo pump, camera, and MOT optics are labeled.

5.19 The paths of the MOT beams and the repump beams. The Raman beam combines with the MOT beam that goes up, but is not in place yet.

5.20 A captured image of the MOT.

5.21 Experimental timing pulse sequence. 1. (189 ms) Magneto optic trap collects cold atoms. 2. (10 ms) MOT coils are turned off for two decay periods. 3. (90 µs) Optical pumping into. 4. (5 µs) Everything off. 5. (25 µs) Raman beams on. 6. (5 µs) Everything off. 7. (875 µs) MOT beam turned on as probe.

5.22 Result from Raman Rabi oscillation test by varying the Raman pulse width (while keeping the total duration from the Raman pulse to the detection time constant). The credit for taking this data goes to Renpeng Fang.

5.23 Result from single Raman pulse test. The credit for taking this data goes to Renpeng Fang.
5.24 Result from single Raman pulse test. The credit for taking this data goes to Renpeng Fang.

5.25 Results of the Ramsey fringe test for off-resonant Raman-Rabi interactions of width 12.5 µs separated by (a) 25 µs, (b) 50 µs, and (c) 75 µs of dark, free precession, time. The credit for taking this data goes to Renpeng Fang.
CHAPTER 1

Atomic interaction with external fields

The information in this chapter deals with fundamental properties of atom-field interaction, and can be found in such sources as [16, 17, 18, 19, 14].

1.1. Atomic interaction with electric field

1.1.1. Two level atom

1.1.1.1. Dipole approximation. The Hamiltonian for an electron in the potential of the nucleus is \( \hat{H}_0 = \frac{\hat{P}^2}{2m} + V(r) \) where \( \hat{P} = -i\hbar\nabla \) is the center of mass momentum and \( m \) is the mass of the atom. In the presence of an external electromagnetic field, the Hamiltonian becomes

\[
\hat{H} = \frac{[\hat{P} + eA(r,t)]^2}{2m} - e\Phi(r,t) + V(r)
\]

(1.1)

where \( A(r,t) \) is the vector potential and \( \Phi(r,t) \) the scalar potential of the external field. The field is invariant under the gauge transformations \( A \rightarrow A + \nabla \chi \) and \( \Phi \rightarrow \Phi - \partial \chi / \partial t \), where \( \chi(r,t) \) is an arbitrary scalar field. The Hamiltonian can be simplified under the Coulomb gauge, which has the constraint \( \nabla \cdot A = 0 \). The scalar potential under the Coulomb gauge is a Poissonian, \( -\nabla^2 \Phi = \rho / \epsilon_0 \), where \( \rho \) is a charge density from outside the atom, and \( \epsilon_0 \) is permittivity of free space. Therefore, with no extra charge around the atom, the scalar potential can be ignored, and the vector potential obeys the wave
equation,
\[
\nabla^2 A - \frac{1}{c^2} \frac{\partial^2 A}{\partial t^2} = 0
\]

The plane wave solution of the wave equation is of the form
\[
A(r, t) = A_0 e^{i(k \cdot r - \omega t)} + c.c.
\]

where \(\omega\) is the frequency, \(k = 2\pi/\lambda\) is the wave vector, and \(\lambda\) is the wavelength of the field. For an atom, which is on the order of \(10^{-10}\), in an electric field with the wavelength on the order of \(10^{-7}\) or so, it sees a spatially uniform field. Putting the atom at the origin, \(k \cdot r \ll 1\) and the dipole approximation leads to \(A(r, t) \simeq A(t)\). Using \(\chi(r, t) = -A(t) \cdot r\) as the scalar field of choice, we find:
\[
\nabla \chi(r, t) = -A(t)
\]
\[
\frac{\partial \chi(r, t)}{\partial t} = -r \cdot \frac{\partial A}{\partial t} = r \cdot E(t)
\]

where \(E(t)\) is the external electric field acting on the atom. Putting all these elements together, the Hamiltonian simplifies to
\[
\hat{H} = \frac{\hat{p}^2}{2m} + V(r) + e r \cdot E(t) = \hat{H}_0 - \hat{d} \cdot E(t)
\]

where \(\hat{d}\) is the dipole moment operator, and the last term represents the interaction between the atomic dipole moment and the external field.

1.1.1.2. Rabi model of the atom, rotating wave approximation, and rotating wave transformation. Interacting with the external electric field, we first consider a
two level atom (Fig. 1.1(a)). As an example, a microwave pulse can couple two hyprefine ground states of an alkali atom and achieve this kind of coupling. The state of the system can be written \( |\Psi\rangle = c_1(t)|1\rangle + c_2(t)|2\rangle \), where \( |1\rangle \) is the ground state, with energy \( E_1 = \hbar \omega_1 \), and \( |2\rangle \) the excited state, with energy \( E_2 = \hbar \omega_2 \). The transition frequency is \( \omega_0 = \omega_2 - \omega_1 \), and the frequency of the electric field is \( \omega \). The unperturbed Hamiltonian is \( \hat{H}_0 = E_1 \sigma_{11} + E_2 \sigma_{22} \), where \( \sigma_{ii} = |i\rangle \langle i| \) for \( i = 1, 2 \). Using the Schrodinger equation, \( \hat{H}_0|\Psi\rangle = i\hbar (\partial |\Psi\rangle / \partial t) \), the amplitudes of the states evolve as \( c_1(t) = c_{10} \exp(-i\omega_1 t) \) and \( c_2(t) = c_{20} \exp(-i\omega_2 t) \) where \( c_{10} \) and \( c_{20} \) are the initial state amplitudes. The interaction Hamiltonian with this two level system is \( \hat{H}_I = \langle 1| \hat{d} |2\rangle \cdot E(t) \sigma_x \) where \( \sigma_x \) is the Pauli-x matrix. With \( E(t) = E_0 \cos(\omega t) \), the combined Hamiltonian is

\[
(1.7) \quad \hat{H} = \hbar \begin{pmatrix}
\omega_1 & -\Omega_0/2 \left( e^{i\omega t} + e^{-i\omega t} \right) \\
-\Omega_0/2 \left( e^{i\omega t} + e^{-i\omega t} \right) & \omega_2
\end{pmatrix}
\]

where \( \Omega_0(t) = \langle 1| \hat{d} |2\rangle \cdot E_0(t) / \hbar \) is the Rabi frequency.

Before the rotating wave approximation (RWA) and rotating wave transformation (RWT), as an exercise, the Schrodinger equation for \( |\Psi\rangle = c_1(t)|1\rangle + c_2|2\rangle \) can be solved
with this Hamiltonian. Because the system is in a periodic potential due to the periodic electric field, we can use the Bloch form of the wave equation to describe its coefficients as $c_1(t) = \sum_n c_{1n} \exp[-i(\omega_1 - n\omega)t]$ and $c_2(t) = \sum_n c_{2n} \exp[-i(\omega_2 - n\omega)t]$. Hence, for $\tau = 2\pi/(\omega_1 - n\omega)$, we would get the periodic condition $c_1(t) = c_1(t + \tau)$ and $c_2(t) = c_2(t + \tau)$.

The derivatives of these equations are $\dot{c}_1(t) = -i \sum_n c_{1n} (\omega_1 - n\omega) \exp[-i(\omega_1 - n\omega)t]$ and $\dot{c}_2(t) = -i \sum_n c_{2n} (\omega_2 - n\omega) \exp[-i(\omega_2 - n\omega)t]$. Plugging these into the Schrodinger equation, we find:

\begin{align}
\sum_n c_{1n} (\omega_1 - n\omega) e^{i\omega_1 t} &= \omega_1 e^{-i\omega_1 t} \sum_n c_{1n} e^{i\omega_1 t} + \frac{\Omega_0}{2} \left( e^{-i(\omega_2 - \omega)t} + e^{-i(\omega_2 + \omega)t} \right) \sum_n c_{2n} e^{i\omega_1 t} \\
\sum_n c_{2n} (\omega_2 - n\omega) e^{i\omega_2 t} &= \omega_2 e^{-i\omega_2 t} \sum_n c_{2n} e^{i\omega_2 t} + \frac{\Omega_0}{2} \left( e^{-i(\omega_1 - \omega)t} + e^{-i(\omega_1 + \omega)t} \right) \sum_n c_{1n} e^{i\omega_2 t}
\end{align}

These equations can be simplified into:

\begin{align}
-\omega e^{-i\omega_1 t} \sum_n c_{1n} e^{i\omega_1 t} &= \frac{\Omega_0}{2} \left( e^{-i(\omega_2 - \omega)t} + e^{-i(\omega_2 + \omega)t} \right) \sum_n c_{2n} e^{i\omega_1 t} \\
-\omega e^{-i\omega_2 t} \sum_n c_{2n} e^{i\omega_2 t} &= \frac{\Omega_0}{2} \left( e^{-i(\omega_1 - \omega)t} + e^{-i(\omega_1 + \omega)t} \right) \sum_n c_{1n} e^{i\omega_2 t}
\end{align}

If we assume that for each $n$,

\begin{align}
-\omega e^{-i\omega_1 t} c_{1n} e^{i\omega_1 t} &= \frac{\Omega_0}{2} \left( e^{-i(\omega_2 - \omega)t} + e^{-i(\omega_2 + \omega)t} \right) c_{2n} e^{i\omega_1 t} \\
-\omega e^{-i\omega_2 t} c_{2n} e^{i\omega_2 t} &= \frac{\Omega_0}{2} \left( e^{-i(\omega_1 - \omega)t} + e^{-i(\omega_1 + \omega)t} \right) c_{1n} e^{i\omega_2 t}
\end{align}
then they can be further simplified:

\[(1.14) \quad -\omega e^{-i\omega_1 t}c_{1n} = \frac{\Omega_0}{2} \left( e^{-i(\omega_2 - \omega)t} + e^{-i(\omega_2 + \omega)t} \right) c_{2n} \]

\[(1.15) \quad -\omega e^{-i\omega_2 t}c_{2n} = \frac{\Omega_0}{2} \left( e^{-i(\omega_1 - \omega)t} + e^{-i(\omega_1 + \omega)t} \right) c_{1n} \]

Substituting Eq. (1.15) into Eq. (1.14), we arrive at:

\[(1.16) \quad \frac{2n^2\omega^2}{\Omega_0^2} - 1 = \cos(2\omega t) \]

However, this implies that \(|\frac{2n^2\omega^2}{\Omega_0^2} - 1| \leq 1\). Even if this were to be true, the solution is time dependent so that solutions exist only at certain times. Instead, we make the RWA, which allows the Hamiltonian to be simplified to:

\[(1.17) \quad \hat{H} = \hbar \begin{pmatrix} \omega_1 & -\Omega_0 e^{i\omega t} \\ -\Omega_0 e^{-i\omega t} & \omega_2 \end{pmatrix} \]

The RWA can be made because the Schrodinger equation has terms that are, for \(|1\rangle\), a low frequency term \((\omega - \omega_1)\) and a high frequency term \((\omega + \omega_1)\) (for \(|2\rangle\), \((\omega - \omega_2)\) and \((\omega + \omega_2)\)). The high frequency term effectively averages to zero and can be taken out of the Hamiltonian.

Next, the Hamiltonian can become time independent through the RWT. We define a rotation operator, or the \(Q\) matrix, \(\hat{Q} = \exp(i\theta_1 t)\sigma_{11} + \exp(i\theta_2 t)\sigma_{22}\). The new state after the RWT is \(|\tilde{\Psi}\rangle = \hat{Q}(t)|\Psi\rangle\). The Schrodinger equation, \(i|\dot{\tilde{\Psi}}\rangle = i\frac{\partial}{\partial t} [\hat{Q}(t)|\Psi\rangle] = i\hat{Q}|\Psi\rangle + \hat{Q}\hat{H}|\Psi\rangle\). Taking the derivative of the \(Q\) matrix, \(\dot{Q} = (i\theta_1 \sigma_{11} + i\theta_2 \sigma_{22})\hat{Q} \equiv \hat{M}\hat{Q}\).
Inserting this into the Schrödinger equation, along with \( \hat{Q}^{-1} \hat{Q} = 1 \), we find
\[
i \hat{\dot{\Psi}} = i \hat{\dot{M}} \hat{Q} |\Psi\rangle + \hat{Q} \hat{\dot{H}} \hat{Q}^{-1} |\Psi\rangle = (i \hat{\dot{M}} + \hat{Q} \hat{\dot{H}} \hat{Q}^{-1}) |\Psi\rangle.
\]
The Hamiltonian for the rotated state is \( \hat{H} \equiv i \hat{\dot{M}} + \hat{Q} \hat{H} \hat{Q}^{-1} \). Solving for the second term with \( \hat{H} \) as it appears in Eq. (5.2):

\[
(1.18) \quad \hat{Q} \hat{\dot{H}} \hat{Q}^{-1} = \hbar \begin{pmatrix}
\omega_1 & -\frac{\Omega_0}{2} e^{i(\omega + \theta_1 - \theta_2)\omega t} \\
-\frac{\Omega_0}{2} e^{-i(\omega + \theta_1 - \theta_2)\omega t} & \omega_2
\end{pmatrix}
\]

The time dependent argument of the exponents in the coherence terms vanish when \( \theta_2 - \theta_1 = \omega \). Hence,

\[
(1.19) \quad \hat{H} \equiv i \hat{\dot{M}} + \hat{Q} \hat{\dot{H}} \hat{Q}^{-1} = \hbar \begin{pmatrix}
\omega_1 - \theta_1 & -\frac{\Omega_0}{2} \\
-\frac{\Omega_0}{2} & \omega_2 - \theta_2
\end{pmatrix}
\]

Setting \( \theta_1 = \omega_1, \theta_2 = \omega_1 + \omega, \) and \( \delta = \omega - (\omega_2 - \omega_1) \), the final form of the Hamiltonian after dipole approximation, RWA, and RWT is:

\[
(1.20) \quad \hat{H} = \hbar \begin{pmatrix}
0 & -\frac{\Omega_0}{2} \\
-\frac{\Omega_0}{2} & -\delta
\end{pmatrix}
\]

The new rotated states are \( |\tilde{\Psi}\rangle = \tilde{c}_1(t) |\tilde{1}\rangle + \tilde{c}_2(t) |\tilde{2}\rangle \) where the coefficients are \( \tilde{c}_1(t) = e^{i\omega_1 t} c_1(t) \) and \( \tilde{c}_2(t) = e^{i(\omega_2 + \delta) t} c_2(t) \).

The Schrödinger equation with this Hamiltonian can be solved, first, for the case \( \delta = 0 \), with the initial conditions \( c_1(0) = 1 \) and \( c_2(0) = 0 \) – which lead to \( \tilde{c}_1(0) = 1 \) and \( \tilde{c}_2(0) = 0 \).
the initial conditions, we can set $B = 0$. Taking the first derivative and putting it back into the equation for $\tilde{c}_1$, with the initial conditions, we find that $A = -i$, so that the state evolves as:

\begin{equation}
|\tilde{\psi}\rangle = \cos\left(\frac{\Omega_0 t}{2}\right) |\tilde{1}\rangle - i \sin\left(\frac{\Omega_0 t}{2}\right) |\tilde{2}\rangle
\end{equation}

Similarly, in the case where $\delta \neq 0$, the state evolves as

\begin{equation}
|\tilde{\psi}\rangle = e^{i\delta t/2} \left[ \cos\left(\frac{\Omega t}{2}\right) - i \frac{\delta}{\Omega} \sin\left(\frac{\Omega t}{2}\right) \right] |\tilde{1}\rangle - i \frac{\Omega_0}{\Omega} e^{i\delta t/2} \sin\left(\frac{\Omega t}{2}\right) |\tilde{2}\rangle
\end{equation}

where $\Omega \equiv \sqrt{\Omega_0^2 + \delta^2}$. When we set $\delta = 0$, this reduces to Eq. (1.21). In theory, the system can be controlled by changing $\delta$ and $\Omega_0$. The presence of $\delta$, even in the dark zone when there is no $\Omega_0$ and populations are unaffected, the phase of the system will change. When there is external field present and $\Omega_0$ is no longer zero, the population of the system will oscillate between the ground state and the excited state governed by the period of rotation $\Omega t$.

Before moving onto the next section, we consider two additional cases for completeness. First, we consider the Hamiltonian with terms that were dropped from the RWA, or:

\begin{equation}
\hat{H} = \hbar \begin{pmatrix}
\omega_1 & -\frac{\Omega_0}{2} e^{-i\omega t} \\
-\frac{\Omega_0}{2} e^{i\omega t} & \omega_2
\end{pmatrix}
\end{equation}

After the RWT, this becomes

\begin{equation}
\hat{\tilde{H}} = \hbar \begin{pmatrix}
0 & -\frac{\Omega_0}{2} \\
-\frac{\Omega_0}{2} & 2\omega
\end{pmatrix}
\end{equation}
Solving the Schrödinger equation with this Hamiltonian, we find that the state evolves as

\[ |\tilde{\psi}\rangle = e^{-i(\omega+\omega_1)t} \left[ \cos \left( \frac{\chi t}{2} \right) + i \frac{2\omega}{\chi} \sin \left( \frac{\chi t}{2} \right) \right] |1\rangle - i \frac{\Omega_0}{\chi} e^{-i\omega t} \sin \left( \frac{\chi t}{2} \right) |2\rangle \]

where \( \chi = \sqrt{(2\omega)^2 + \Omega_0^2} \). The maximum value that \( \tilde{c}_2 \) can have is \( \tilde{c}_{2,\text{max}} = \Omega_0/\chi \). With typical values \( \Omega_0 \simeq 2\pi \cdot 10^6 \text{ sec}^{-1} \) and \( \omega \simeq 2\pi \cdot 10^{14} \text{ sec}^{-1} \), \( \tilde{c}_{2,\text{max}} \sim 10^{-8} \), which is negligible, justifying the RWA.

1.1.1.3. Density matrix and Bloch sphere representation. Another approach to understanding the evolution of the two level system is by representing the state of the system with a Bloch vector. We first define the density matrix \( \hat{\rho} \equiv |\psi\rangle \langle \psi| \). The first time derivative of the density matrix is \( \dot{\hat{\rho}} = |\dot{\psi}\rangle \langle \psi| + |\psi\rangle \langle \dot{\psi}| \). Substituting the Schrödinger equation and its conjugate into the equation, we arrive at the Liouville equation for the density matrix (without the decay terms):

\[ \dot{\hat{\rho}} = i \frac{\hbar}{\hbar} [\hat{\rho}, \hat{H}] \]

However, it is more realistic to add decay terms to Eq. (1.26). Moreover, a unique steady state solution does not exist for Eq. (1.30) when decay terms are not included. Hence, we require a full Liouville equation with the Hamiltonian that includes decay term, and a source matrix \( L \):

\[ \dot{\hat{\rho}} = i \frac{\hbar}{\hbar} [\hat{\rho}, \hat{H}] + \hat{L} \]
where the Hamiltonian is now:

\[ \hat{H} = \hbar \begin{pmatrix} \omega_1 & -\frac{\Omega_0}{2} e^{i\omega t} \\ -\frac{\Omega_0}{2} e^{-i\omega t} & \omega_2 - i\Gamma/2 \end{pmatrix} \]

where \( \Gamma \) is the spontaneous decay term, and the source matrix is:

\[ \tilde{L} = \hbar \begin{pmatrix} \Gamma \rho_{22} & 0 \\ 0 & 0 \end{pmatrix} \]

Expanding Eq. (1.27), the density matrix evolves as:

\[ \dot{\rho} = \hbar \begin{pmatrix} -i\frac{\Omega_0}{2} (\rho_{21} - \rho_{12}) + \Gamma \rho_{22} & -i\frac{\Omega_0}{2} (\rho_{22} - \rho_{11}) - i\delta \rho_{12} - \frac{\Gamma}{2} \rho_{12} \\ -i\frac{\Omega_0}{2} (\rho_{11} - \rho_{22}) + i\delta \rho_{21} - \frac{\Gamma}{2} \rho_{21} & -i\frac{\Omega_0}{2} (\rho_{12} - \rho_{21}) - \Gamma \rho_{22} \end{pmatrix} \]

We can find the steady state solution by setting \( \dot{\rho} = 0 \), along with \( \rho_{11} + \rho_{22} = 0 \):

\[ \rho = \begin{pmatrix} \frac{\Omega_0^2 + 4\delta^2 + \Gamma^2}{2\Omega_0^2 + 4\delta^2 + \Gamma^2} & \frac{2\Omega_0 \delta + i\Omega_0 \Gamma}{2\Omega_0^2 + 4\delta^2 + \Gamma^2} \\ \frac{2\Omega_0 \delta - i\Omega_0 \Gamma}{2\Omega_0^2 + 4\delta^2 + \Gamma^2} & \frac{\Omega_0^2}{2\Omega_0^2 + 4\delta^2 + \Gamma^2} \end{pmatrix} \]

Many insights can be gathered from this information. When the decay is large compared to the other terms, \( \Gamma \gg \Omega_0, \delta \), the steady state solution is \( \rho_{11} \sim 1 \) and \( \rho_{22} \sim 0 \). The system will remain in state \(|1\rangle\). When the Rabi interaction term is large compared to the other terms, we find \( \rho_{11} = \rho_{22} \sim 1/2 \), and the system has equal chance of occupying either state.
The system, in terms of density matrix, can be mapped onto a Bloch sphere with the three dimensional Bloch vector defined as:

\[
R \equiv R_1 \hat{e}_1 + R_2 \hat{e}_2 + R_3 \hat{e}_3
\]

where \(R_1 \equiv 2\text{Re}(\rho_{12})\), \(R_2 \equiv 2\text{Im}(\rho_{12})\), and \(R_3 \equiv \rho_{11} - \rho_{22}\). Solving for the density matrix elements, \(\rho_{11} = (1 + R_3)/2\), \(\rho_{12} = (R_1 + iR_2)/2\), \(\rho_{21} = (R_1 - iR_2)/2\), and \(\rho_{22} = (1 - R_3)/2\). Then (1.27) can be solved in terms of the Bloch vector elements:

\[
\begin{pmatrix}
\dot{R}_1 \\
\dot{R}_2 \\
\dot{R}_3
\end{pmatrix} =
\begin{pmatrix}
0 & \delta & 0 \\
-\delta & 0 & -\Omega_0 \\
0 & \Omega_0 & 0
\end{pmatrix}
\begin{pmatrix}
R_1 \\
R_2 \\
R_3
\end{pmatrix}
\]

By introducing \(Q \equiv \Omega_0 \hat{e}_1 - \delta \hat{e}_3\), the Bloch vector will now evolve as

\[
\dot{R} = Q \times R
\]

As was seen previously, the presence of \(\delta\) will change the phase but not the population of the system; alternatively, the presence of \(\Omega_0\) will change the population but not the phase of the system.

1.1.1.4. Separated field Ramsey fringe experiment. The Bloch sphere representation of a two level atom can be used to calculate the time evolution of the system, as
in the separated field three zone Ramsey experiment: an interaction zone with a $\pi/2$
pulse, followed by a dark zone, followed by another interaction zone with a $\pi/2$ pulse. We
consider the system with the Hamiltonian after dipole approximation, RWA, and RWT,
as in Eq. (5.2). When $\Omega_0 \gg \Gamma$, this is valid for systems with decay, as well. Initially, the
atom is in its ground state so that $\mathbf{R} = R_3 \hat{e}_3$. In the first interaction zone, the condition
$\Omega_0 \gg \delta$ allows for the approximation $\delta \to 0$. Then $\mathbf{Q} \simeq \Omega_0 \hat{e}_1$ and the Bloch vector with
the given initial condition in the first interaction zone is:

\begin{equation}
\mathbf{R} = - \sin (\Omega_0 t) \hat{e}_2 + \cos (\Omega_0 t) \hat{e}_3
\end{equation}

After $\Omega_0 T_1 = \pi/2$, the system can be described by the Bloch vector $\mathbf{R} = - \hat{e}_2$. In the
dark zone, the system’s evolution is governed by $\mathbf{Q} = - \delta \hat{e}_3$. Solving for the Bloch vector
with the initial condition in the dark zone:

\begin{equation}
\mathbf{R} = \sin (\delta t) \hat{e}_1 + \cos (\delta t) \hat{e}_2
\end{equation}

After a time $T_2$ in the dark zone, the system is in state $\mathbf{R} = \sin (\delta T_2) \hat{e}_1 + \cos (\delta T_2) \hat{e}_2$.
In the third interaction zone, the approximation $\mathbf{Q} \simeq \Omega_0 \hat{e}_1$ is valid again. Then the Bloch
vector in the second interaction zone is:

\begin{equation}
\mathbf{R} = \sin (\delta T_2) \hat{e}_1 + \cos (\delta T_2) \cos (\Omega_0 t) \hat{e}_2 - \cos (\delta T_2) \sin (\Omega_0 t) \hat{e}_3
\end{equation}

Therefore, as a result of passing through the three zones, the ground state population
will be:

\begin{equation}
\rho_{11} = [1 - \cos (\delta T_2) \sin (\Omega_0 t)]/2
\end{equation}
For different values of $\Omega_0 t$ and $\delta T_2$, the ground state population will show oscillatory behavior. When $\Omega_0 T_1 = \pi/2$, $\rho_{11} = \sin^2(\delta T_2/2)$ and $\rho_{22} = \cos^2(\delta T_2/2)$. When $\Omega_0 T_1 \gg \delta T_2$, the population will show Ramsey fringes around $\Omega_0 T_1 = \pi/2$. The presence of $\delta T_2$ in the final term modulates the population and increases the frequency, and therefore, the accuracy of the clock.

The picture is more complicated when $\delta$ is taken into account in both interaction zones. In this case, the Bloch vector in the first interaction zone as a function of time is:

\[
R = \frac{2\delta \Omega_0}{\Omega^2} \sin^2 \frac{\Omega t}{2} \hat{e}_1 + \frac{\Omega_0}{\Omega} \sin \Omega t \hat{e}_2 + \left( \frac{2\delta \Omega_0}{\Omega^2} \sin^2 \frac{\Omega t}{2} - 1 \right) \hat{e}_3
\]

where as earlier, $\Omega \equiv \sqrt{\Omega_0^2 + \delta^2}$. Hence, at the end of the $\Omega T_1 = \pi/2$ pulse, $R = \frac{\delta \Omega_0}{\Omega^2} \hat{e}_1 + \frac{\Omega_0}{\Omega} \hat{e}_2 - \frac{\delta^2}{\Omega^2} \hat{e}_3$. In the dark zone, solving for the components of the Bloch vector with the given initial condition yields at the end of time $T_2$:

\[
R = \alpha \hat{e}_1 + \beta \hat{e}_2 + \xi \hat{e}_3
\]

where

\[
\alpha = \frac{\delta \Omega_0}{\Omega^2} \cos \delta T_2 + \frac{\Omega_0}{\Omega} \sin \delta T_2
\]

\[
\beta = -\frac{\delta \Omega_0}{\Omega^2} \sin \delta T_2 + \frac{\Omega_0}{\Omega} \cos \delta T_2
\]

\[
\xi = -\frac{\delta^2}{\Omega^2}
\]
After the second interaction zone, the elements of the Bloch vector become:

\begin{align}
R_1 &= \frac{\beta\delta}{\Omega} \sin \Omega t + \frac{\delta}{\Omega^2} (\alpha\delta + \xi \Omega_0) \cos \Omega t + \alpha - \frac{\delta}{\Omega^2} (\alpha\delta + \xi \Omega_0) \\
R_2 &= \beta \cos \Omega t - \frac{\alpha\delta + \xi \Omega_0}{\Omega} \sin \Omega t \\
R_3 &= \frac{\beta \Omega_0}{\Omega} \sin \Omega t + \frac{\Omega_0}{\Omega^2} (\alpha\delta + \xi \Omega_0) \cos \Omega t + \xi - \frac{\Omega_0}{\Omega^2} (\alpha\delta + \xi \Omega_0)
\end{align}

Substituting equations (1.41) - (1.43) into the above yields:

\begin{align}
R_3 &= \frac{\Omega_0^2}{\Omega^3} (-\delta \sin \delta T + \Omega \cos \delta T) \sin \Omega t + \frac{\delta \Omega_0^2}{\Omega^3} (\delta \cos \delta T_2 - \delta + \Omega \sin \delta T) \cos \Omega t \\
&- \frac{\delta \Omega_0^2}{\Omega^4} (\delta \cos \delta T_2 + \Omega \sin \delta T) - \frac{\delta^4}{\Omega^4}
\end{align}

The ground state population is then:

\begin{align}
\rho_{11} &= \frac{\Omega_0^2}{2\Omega^3} (-\delta \sin \delta T_2 + \Omega \cos \delta T_2) \sin \Omega t \\
&+ \frac{\delta \Omega_0^2}{2\Omega^4} (\delta \cos \delta T_2 - \delta + \Omega \sin \delta T_2) \cos \Omega t \\
&- \frac{\delta \Omega_0^2}{2\Omega^4} (\delta \cos \delta T_2 + \Omega \sin \delta T_2) - \frac{\delta^4}{2\Omega^4} + \frac{1}{2}
\end{align}

Although a complicated expression, the variables can still be manipulated to give meaningful results. If again, we set \( \delta T_2 = 2n\pi \) and \( \Omega T_1 = \pi/2 \), then \( \rho_{11} = \Omega_0^2/\Omega^2 \) and it will only be unity in the limit \( \delta \to 0 \).
1.1.2. Three level atomic system

The hyperfine ground states of an atom can be coupled with a microwave. Another method, which we employ for our clock, is to use an optically off-resonant Raman transition with three hyperfine energy levels in a Λ scheme depicted in Fig. 1.1 (b). The ground states $|1\rangle$ and $|2\rangle$ of this atom interact with an excited state $|3\rangle$ via two coherent electromagnetic light fields of frequencies $\omega_1$ and $\omega_2$, respectively, detuned from resonance by $\delta_1$ and $\delta_2$, respectively. The energy of each level is $\omega_{\mu 0}$ for $\mu = [1, 2, 3]$. The Hamiltonian after the dipole approximation and RWA is

\begin{equation}
\hat{H} = \hbar \begin{pmatrix}
\omega_{10} & 0 & -\frac{\Omega_1}{2} e^{i\omega_1 t} \\
0 & \omega_{20} & -\frac{\Omega_2}{2} e^{i\omega_2 t} \\
-\frac{\Omega_1}{2} e^{-i\omega_1 t} & -\frac{\Omega_2}{2} e^{-i\omega_2 t} & \omega_{30}
\end{pmatrix}
\end{equation}

where $\Omega_{1,2}$ are the Rabi frequencies. Furthermore, RWT can be performed on this Hamiltonian with $\hat{Q} = \sum_\mu e^{(i\theta_\mu t)} \sigma_{\mu\mu}$ where $\sigma_{\mu\mu} = |\mu\rangle\langle\mu|$. By choosing $\theta_1 = (\omega_{10} + \omega_{20} - \omega_1 + \omega_2)/2$, $\theta_2 = (\omega_{10} + \omega_{20} + \omega_1 - \omega_2)/2$, and $\theta_3 = (\omega_{10} + \omega_{20} + \omega_1 + \omega_2)/2$, Eq. \(1.49\) can be expressed as \(20\):

\begin{equation}
H = \frac{\hbar}{2} [(\delta \sigma_{11} - \delta \sigma_{22} - 2\Delta \sigma_{33}) - (\Omega_1 \sigma_{13} + \Omega_2 \sigma_{23} + \text{h.c.})]
\end{equation}

where $\sigma_{\mu\nu} = |\mu\rangle\langle\nu|$, $\delta \equiv \delta_1 - \delta_2$ is the two photon detuning, and $\Delta \equiv (\delta_1 + \delta_2)/2$ is the average detuning. Here, we have also assumed a phase transformation applied to the Hamiltonian so that $\Omega_{1,2}$ are real.

We assume next that $\Delta \gg \Gamma, \Omega_1$, and $\Omega_2$ (where $\Gamma$ is the decay rate of state $|3\rangle$) so that the effect of $\Gamma$ can be neglected, and state $|3\rangle$ can be eliminated adiabatically.
(in Chapter 2) we will consider the residual effect of spontaneous emission).

Under these conditions, the Hamiltonian of the reduced two level system can be expressed as

\[ H_{\text{red}} = \frac{\hbar \delta}{2} \sigma_z - \frac{\hbar \Omega}{2} \sigma_x, \]

where \( \Omega \equiv \Omega_1 \Omega_2 / 2 \Delta \) is the Raman Rabi frequency, and \( \sigma_z \) and \( \sigma_x \) are Pauli matrices defined as \( \sigma_z = (\sigma_{11} - \sigma_{22}) \) and \( \sigma_x = (\sigma_{12} + \sigma_{21}) \). The quantum state for this system is given by

\[ |\psi(t')\rangle = W_{\delta t} |\psi(t')\rangle \]

and the propagation operator is given by

\[ W_{\delta t} = e^{i\delta t/2} \begin{pmatrix} \cos \phi - i \frac{\delta}{\Omega'} & -i \frac{\Omega}{\Omega'} \sin \phi \\ -i \frac{\Omega}{\Omega'} \sin \phi & \cos \phi + i \frac{\delta}{\Omega'} \sin \phi \end{pmatrix} \]

where \( \phi = \Omega' t / 2 \), and \( \Omega' \equiv \sqrt{\Omega^2 + \delta^2} \) is the generalized Rabi frequency. Hence, a Raman interaction that is characterized by a large average detuning from the excited state can be effectively treated as a two level system, in which the only states that matter are the ground states.

1.1.3. Off-resonant Raman-Rabi excitation and Ramsey fringes

When this system is excited by two pulses of duration \( T_1 \), separated in time by \( T_2 \), we have \( \Omega_1(t) \sim \Omega_2(t) = \Omega_0 [U(t) - U(t - T_1) + U(t - (T_1 + T_2)) - U(t - (2T_1 + T_2))] \)

where \( U(t) \) is the Heaviside step function. When \( \delta \ll \Omega \) and the width of the pulse is chosen to be \( \Omega T_1 = \pi/2 \), each pulse acts on the system as a propagation operator \( W_{\pi/2}^0 = (I - i \sigma_x) / \sqrt{2} \). While the system is between \( t = T_1 \) and \( t = T_1 + T_2 \) where no interaction is present, the propagation operator can be expressed as \( W_{0}^{ST_2} = \sigma_{11} + e^{i\delta T_2} \sigma_{22} \).

After passing through the three zones, the state of the atom that was originally in state \( |1\rangle \) is

\[ |\psi\rangle = W_{\pi/2}^0 W_{0}^{ST_2} W_{\pi/2}^0 |1\rangle = -ie^{i\theta} (\sin \theta |1\rangle + \cos \theta |2\rangle) \]

where \( \theta = \delta T_2 / 2 \) is the dephasing
angle. The probability of the atom being in state $|2\rangle$ is $P_2 \equiv |\langle 2|\psi\rangle|^2 = (\cos \theta)^2$. This is essentially the result that was derived in the previous section where a two level atom was discussed. In Chapter 2, the results of this section will be used as foundation for an off-resonant Raman-Rabi excitation based collective state atomic clock.

### 1.1.4. Coherent population trapping in atomic system

The coherent population trapping (CPT) based atomic clock employs Raman interactions in Λ-type three level atomic system, shown in Fig. 1.3(a). In order to differentiate the CPT based clock from the off-resonant clock, we use different notations for the energy levels. The long lived ground states, $|a\rangle$ and $|b\rangle$, interact separately with the short lived excited state, $|e\rangle$, via electric dipole interactions. Field of frequency $\omega_1$, with detuning $\delta_1$, couples $|a\rangle$ and $|e\rangle$, and field of frequency $\omega_2$, with detuning $\delta_2$, couples $|b\rangle$ and $|e\rangle$. In the Λ-type scheme, the ground states do not directly interact with each other. The strengths of these interactions are given by Rabi frequencies $\Omega_1 \equiv \mu_{ae} \cdot E_1 / \hbar$ and $\Omega_2 \equiv \mu_{be} \cdot E_2 / \hbar$, where $\mu$ is the dipole moment operator of the atom. After dipole approximation and RWA
the Hamiltonian for this system is:

\begin{equation}
H = \sum_{i=a,b,e} \epsilon_i \sigma_{ii} - \frac{\hbar}{2} (\Omega_1 e^{i(\omega_1 t - \phi_1)} \sigma_{ae} + \Omega_2 e^{i(\omega_2 t - \phi_2)} \sigma_{be} + \text{H.c.})
\end{equation}

where \(\epsilon_i\) is the energy of state \(|i\rangle\), \(\sigma_{ij} \equiv |i\rangle \langle j|\), \(\phi_{1,2}\) are the phases of the fields, and the Rabi frequencies are taken to be real. The Hamiltonian can be written in the rotating wave basis for convenience, in which the Hamiltonian is time-independent and the states evolve in time. The basis states in this frame are \(|\tilde{a}\rangle \equiv \exp[-i(\omega_1 t + \phi_1)]|a\rangle\), \(|\tilde{b}\rangle \equiv \exp[-i(\omega_2 t + \phi_2)]|b\rangle\), and \(|\tilde{e}\rangle \equiv |e\rangle\), and the Hamiltonian is:

\begin{equation}
\tilde{H} = \frac{\hbar}{2} [\Delta(\sigma_{\tilde{a}\tilde{a}} - \sigma_{\tilde{b}\tilde{b}}) - (2\delta + i\Gamma)\sigma_{\tilde{e}\tilde{e}}] - \frac{\hbar}{2}(\Omega_1 \sigma_{\tilde{a}\tilde{e}} + \Omega_2 \sigma_{\tilde{b}\tilde{e}} + \text{H.c.})
\end{equation}

where \(\Gamma\) is the decay rate from state \(|\tilde{e}\rangle\). The equation of motion for the density matrix can be written as \(\dot{\rho} = (i/\hbar)[\tilde{H}, \rho] + \tilde{L}\rho_{\tilde{e}\tilde{e}}\), where the source matrix, \(\tilde{L} = \Gamma_{ea}\sigma_{\tilde{a}\tilde{a}} + \Gamma_{eb}\sigma_{\tilde{b}\tilde{b}}\).

Next, we partially diagonalize the Hamiltonian by rotating it by \(R = \cos \theta (\sigma_{aa} + \sigma_{bb}) + \sin \theta (\sigma_{ab} - \sigma_{ba}) + \sigma_{ee}\), where \(\sin \theta = \Omega_1/\Omega\) and \(\cos \theta = \Omega_2/\Omega\), and \(\Omega \equiv \sqrt{\Omega_1^2 + \Omega_2^2}\). The basis states in this frame, as depicted in Fig. 4.1(b), are the coherent superpositions of the ground states, \(|-\rangle = \cos \theta |\tilde{a}\rangle - \sin \theta |\tilde{b}\rangle\) and \(|+\rangle = \cos \theta |\tilde{a}\rangle + \sin \theta |\tilde{b}\rangle\), and the excited state \(|e\rangle = |\tilde{e}\rangle\). The equation of motion in this basis is given by \(\dot{\rho}_R = (i/\hbar)[\rho_R, H_R] + L_R\rho_{ee}\).

The rotated Hamiltonian is:

\begin{equation}
H_R = \frac{\hbar}{2} [C\Delta(\sigma_{--} - \sigma_{++}) - (2\delta + i\Gamma)\sigma_{ee}] + \frac{\hbar}{2}(S\Delta\sigma_{--} - \Omega \sigma_{+e} + \text{H.c.})
\end{equation}

where \(C \equiv \cos 2\theta\) and \(S \equiv \sin 2\theta\), and the rotated source matrix is \(L_R = (\Gamma/2)[(1 + Cd)\sigma_{--} + (1 - Cd)\sigma_{++} + Sd(\sigma_{+-} + \sigma_{-+})]\), where \(d = (\Gamma_{ea} - \Gamma_{eb})/\Gamma\).
In this frame, $|−\rangle$, or the dark state, is decoupled from the excited state, $|e\rangle$. Interaction with fields $\omega_1$ and $\omega_2$ couples only the bright state $|+\rangle$ to $|e\rangle$, and drives the transition between them. Spontaneous emission, however, drives the transition from $|e\rangle$ to both $|−\rangle$ and $|+\rangle$. When $d \neq 0$, or $\Gamma_{ea} \neq \Gamma_{eb}$, the source matrix also contributes to the coherence terms in the Hamiltonian. For simplicity, we only consider the case $d = 0$ for the rest of the chapter. Thus, the interaction with $\omega_1$ and $\omega_2$ will optically pump the system into the dark state.

In the case that $\Gamma \gg S\Delta, \Omega$, the excited state will come into equilibrium with the ground states rapidly at rate $\Gamma$ while the ground states change at a time scale much greater than $1/\Gamma$. Hence, we can adiabatically eliminate $|e\rangle$. For $|\Psi\rangle = c_{−}(t)|−\rangle + c_{+}(t)|+\rangle + c_{e}(t)|e\rangle$, we can make the substitution

$$c_{e} \simeq i[\Omega/(\Gamma - 2i\delta)]c_{+}$$

Hence, the system can be reduced to just two states: the dark state $|−\rangle$ and the damped bright state

$$|+\rangle_{d} \equiv |+\rangle + i\frac{\Omega}{\Gamma - 2i\delta}|e\rangle$$

The wave function is now $|\Psi\rangle = c_{−}|−\rangle + c_{+d}|+\rangle_{d}$, and the Hamiltonian in the new basis states is:

$$H = \frac{\hbar}{2} \begin{pmatrix} C\Delta & S\Delta \\ S\Delta & -(C\Delta - 2\beta) - 2i\alpha \end{pmatrix}$$
where $\alpha$ is the effective damping rate given by,

$$(1.58) \quad \alpha \equiv \frac{1}{2} \frac{\Omega^2 \Gamma}{\Gamma^2 + 4\delta^2} = \left| \frac{\Omega}{\Gamma - 2i\delta} \right|^2 \frac{\Gamma}{2}$$

and $\beta$ is the additional light shift between the states, given by,

$$$(1.59) \quad \beta \equiv \frac{\Omega^2 \delta}{\Gamma^2 + 4\delta^2} = \left| \frac{\Omega}{\Gamma - 2i\delta} \right|^2 \delta$$

The effective coupling rate between $|\rangle_{\uparrow}$ and $|\rangle_{\downarrow}$ is $S\Delta$, and the decay from $|\rangle_{\uparrow}$ to $|\rangle_{\downarrow}$ occurs at rate $\alpha$. The Hamiltonian can be made symmetric by subtracting $\hbar/2$ from the diagonal elements:

$$(1.60) \quad H = \frac{\hbar}{2} [-\beta' \sigma_{--} + (\beta' - 2i\alpha) \sigma_{++} + S\Delta \sigma_{--} + H.c.]$$

where $\sigma_{++} = |\rangle_{\uparrow} \langle \uparrow|$ and $\beta' \equiv \beta - C\Delta$. Hence, the energy difference between $|\rangle_{\downarrow}$ and $|\rangle_{\uparrow}$, or the ac Stark shift, is $\beta'$.

The traditional CPT based clock utilizes atoms that are either in vapor or trapped with laser and magnetic fields, that interact with either a single laser field, or two pulses of laser fields that are separated in time. They initially populate both ground states equally. In the previous case, atoms interact with co-propagating laser fields of frequency $\omega_1$ and $\omega_2$ that are on resonance. The interaction drives the transition from $|\rangle_{\uparrow}$ to $|\rangle_{\downarrow}$, until the population in $|\rangle_{\uparrow}$ is depleted in time scale larger than $1/\alpha$. When $\Delta \neq 0$, the states are coupled at rate $S\Delta$. Plotting $\rho_{\text{ee}}$ as a function of $\Delta$, the integrated signal over the interaction time is that of the electromagnetically induced transparency (EIT) effect.
In the latter case, there is Raman interaction between the atoms and the laser fields for time scale $T_1 \gg 1/\alpha$ to optically pump the atoms into the dark state. During the next period $T_2$, the Raman interaction is turned off, so that the only interaction present is the Rabi flopping between $|\rangle$ and $|+\rangle$ at rate $S\Delta$. In the language of the Bloch vector model, the Bloch vector at the end of the first interaction zone is $\mathbf{R}(T_1) = \hat{e}_3$. Since $\mathbf{R}$ rotates at rate $S\Delta$ around $\hat{e}_1$, the Bloch vector at the end of the dark zone is $\mathbf{R}(T_1 + T_2) = \sin (S\Delta T_2)\hat{e}_2 + \cos (S\Delta T_2)\hat{e}_3$. In the next interaction zone, the atoms interact with the Raman fields with the same phases as in the first zone. Again, there will be decay to the dark zone at the time scale $T_3 \gg 1/\alpha$. Since the signal observed from fluorescence is $S_f = \Gamma \rho_{ee} = 2\alpha \rho_{++}$ from Eq. (4.4), and $\rho_{++} = (1 - R_3)/2$, the fluorescence at time $t$ is

\begin{equation}
S_f(T_1 + T_2 + t) = \alpha[1 - \cos (S\Delta T_2)]e^{-\alpha t}
\end{equation}

Thus, the integrated signal at time $T_3$ is

\begin{equation}
\bar{S}_f(T_1 + T_2 + T_3) = \int_0^{T_3} S_f(T_1 + T_2 + t)dt = 2 \sin^2 (S\Delta T_2/2)
\end{equation}

In other words, the (integrated) signal, as a function of $\Delta$, will produce Ramsey fringes.

1.2. Atomic interaction with magnetic field

In the same manner with which Eq. (1.6) was derived, the Hamiltonian describing the interaction between an atom with an external magnetic field $\mathbf{B}$ can be written [24]:

\begin{equation}
\hat{H}_B = \frac{\mu_B}{\hbar} (g_s \mathbf{S} + g_L \mathbf{L} + g_I \mathbf{I}) \cdot \mathbf{B}
\end{equation}
where $S$, $L$, and $I$ are, respectively, the spin, orbital, and nuclear angular momenta, and $g_S$, $g_L$, and $g_I$ are, respectively, the electron spin, electron orbital, and nuclear g-factors that account for various modifications to the corresponding magnetic dipole moments. Furthermore, some of these properties are fixed: $S = 1/2$, $g_S \simeq 2$, and $g_L = 1 - (m_e/m_{\text{nuc}}) \simeq 1$. If the energy shift due to the magnetic field is small compared to the fine-structure splitting, then $J$ is a good quantum number and the interaction Hamiltonian can be written as [24, 19]:

$$\hat{H}_B = \frac{\mu_B}{\hbar} (g_J \mathbf{J} + g_I \mathbf{I}) \cdot \mathbf{B}$$

where

$$g_J = g_L \frac{J(J+1) - S(S+1) + L(L+1)}{2J(J+1)} + g_s \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)} \simeq 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)}$$

If the energy shift due to the magnetic field is small compared to the hyperfine splittings, then $F$ is a good quantum number, so the interaction Hamiltonian becomes

$$\hat{H}_B = \frac{g_F \mu_B}{\hbar} \mathbf{F} \cdot \mathbf{B}$$

where the hyperfine Lande $g$-factors are given by

$$g_F = g_J \frac{F(F+1) - I(I+1) + J(J+1)}{2F(F+1)} + g_I \frac{F(F+1) + I(I+1) - J(J+1)}{2F(F+1)} \simeq g_J \frac{F(F+1) - I(I+1) + J(J+1)}{2F(F+1)}$$
since \( g_I \ll g_J \). For \(^{85}\text{Rb} \), \( g_J \sim 1 \) and \( g_I \sim 0.001 \). With this information, all the Lande \( g \)-factors can be calculated. For the \(^{85}\text{Rb} \) D1 line \((5^2S_{1/2})\), \( I = 5/2 \), \( L = 0 \), and \( J = I + L = 1/2 \). Hence, for \( F = 2 \), \( g_F = -1/3 \) and for \( F = 3 \), \( g_F = 1/3 \).

Next, rewriting the Hamiltonian,

\[
(1.68) \quad \hat{H}_B = \frac{g_F \mu_B}{\hbar} \mathbf{F} \cdot \mathbf{B} = \alpha_F \left( \hat{F}_x B_x + \hat{F}_y B_y + \hat{F}_z B_z \right)
\]

where the substitution \( \alpha_F \equiv g_F \mu_B / \hbar \) has been made for simplicity. For a given \( F \), the matrix elements of the Hamiltonian can be calculated individually:

\[
(1.69) \quad \langle F, m_F | \hat{H}_B | F, m'_F \rangle = \alpha_F \left( B_x \langle F, m_F | \hat{F}_x | F, m'_F \rangle + B_y \langle F, m_F | \hat{F}_y | F, m'_F \rangle + B_z \langle F, m_F | \hat{F}_z | F, m'_F \rangle \right)
\]

Since \( \hat{F}_z | F, m_F \rangle = \hbar m_F | F, m_F \rangle \), the last part can be written \( \langle F, m_F | \hat{F}_z | F, m'_F \rangle = \hbar m'_F \delta_{m_F, m'_F} \).

To calculate the first and the second terms, we recall that

\[
(1.70) \quad \hat{F}_+ | F, m_F \rangle = \hbar \sqrt{F(F+1) - m_F(m_F+1)} | F, m_F + 1 \rangle
\]

\[
(1.71) \quad \hat{F}_- | F, m_F \rangle = \hbar \sqrt{F(F+1) - m_F(m_F-1)} | F, m_F - 1 \rangle
\]
where \( \hat{F}_+ \equiv \hat{F}_x + i \hat{F}_y \) and \( \hat{F}_- \equiv \hat{F}_x - i \hat{F}_y \). Rearranging the terms, \( \hat{F}_x = \left( \hat{F}_+ + \hat{F}_- \right) / 2 \) and \( \hat{F}_y = \left( \hat{F}_+ - \hat{F}_- \right) / (2i) \). Using these expressions, we can get:

\[
\langle F, m_F | \hat{F}_x | F, m_F' \rangle = \frac{\hbar}{2} \sqrt{F(F + 1) - m_F(m_F + 1)} \delta_{\{m_F, m_F' + 1\}} \\
+ \frac{\hbar}{2} \sqrt{F(F + 1) - m_F(m_F - 1)} \delta_{\{m_F, m_F' - 1\}}
\]

(1.72)

\[
\langle F, m_F | \hat{F}_y | F, m_F' \rangle = -\frac{i \hbar}{2} \sqrt{F(F + 1) - m_F(m_F + 1)} \delta_{\{m_F, m_F' + 1\}} \\
+ \frac{i \hbar}{2} \sqrt{F(F + 1) - m_F(m_F - 1)} \delta_{\{m_F, m_F' - 1\}}
\]

When the external magnetic field is in the direction of the quantization axis, the Zeeman sublevels will shift in energy with respect to the strength of the field. However, when the field is in perpendicular direction to the quantization axis, there is coupling between the closest Zeeman sublevels and will cause mixing between the states’ populations. Therefore, in the presence of an electric field propagating in the direction of the quantization axis, the steady state calculation of the population as a function of detuning will show both splitting and mixing between the levels, which we verified in Fig. 1.4. For two copropagating \( \sigma^+ \) beams in a \( \Lambda \) scheme, the electromagnetic induced transparency (EIT) lines for coherent population transfer are clearly visible in the plots. In Fig. 1.4 (a) when no magnetic field is present, there is no splitting of the Zeeman sublevels and only one line is present. In Fig. 1.4 (b), when there’s field parallel to the quantization axis, there are three EIT lines, corresponding to the transitions \((F = 1, m = 1 \leftrightarrow F' = 2, m = 2 \leftrightarrow F = 2, m = 1)\), \((F = 1, m = 0 \leftrightarrow F' = 2, m = 1 \leftrightarrow F = 2, m = 0)\), and \((F = 1, m = -1 \leftrightarrow F' = 2, m = 0 \leftrightarrow F = 2, m = -1)\). However, when there are fields present in other directions, there is mixing between the states, and the decay channels
from the excited states lead to all other transitions, such that instead of three, we expect to see seven lines. (For \(^{85}\)Rb, we expect to see eleven peaks.) Moreover, the spacing between the peaks is proportional to the magnitude of the total magnetic field, and the ratio between the middle peak to the one on its left, for instance, gives us information on \(B_{\parallel}/B_{\perp}\).

Figure 1.4. The steady state calculation of the master equation in the presence of both electric and magnetic fields for \(^{87}\)Rb D1 line transition, where, in the familiar \(\Lambda\) scheme, the common detuning is zero for the excited state \(F' = 2\), and the ground states are \(F = 1\) and \(F = 2\). The parameters are \(\Omega_1 = \Omega_2 = 0.5\, MHz\). Also included are the decay channels from the excited state on the order of \(\sim 6\) MHz, and the spin relaxation decay between the two hyperfine ground states, on the order of \(\sim 10\) kHz. Plotted are the excited state population, which does not care what the initial state of the system is. Plotting the excited state population is also favorable since it’s the fluorescence from the excited state that is measured in real life. The magnetic fields are (a) \(B_x = B_y = B_z = 0\), (b) \(B_x = B_y = 0\) and \(B_z = 0.5\) G, (c) \(B_x = B_y = 0.5\) G and \(B_z = 0\), and (d) \(B_x = B_y = B_z = 0\) G.
This result is important in two ways. First, we can use this information to understand what lines we expect to see in the experiment. Furthermore, we can possibly use this information to build an atom magnetometer [25, 26, 27, 28]. A single measurement gives us information on $||\mathbf{B}||$, which allows for a scalar magnetometer. It also gives us information on $B_\parallel/B_\perp$. Therefore, obtaining the measurement in two directions will allow us to build a vector magnetometer. This investigation is ongoing in our lab, with the principal student in charge being Renpeng Fang. He has seen the EIT effect using a vapor cell with rubidium and neon buffer gas. We expect more to come from this experiment later, and a paper will soon be drafted for publishing.

The elementary material covered in this chapter has been used throughout this dissertation and one hopes that others would find it helpful.
CHAPTER 2

Rydberg Assisted Light Shift Imbalance Induced Blockade

2.1. Introduction

In most protocols for quantum computing or quantum information processing, the fundamental building block is the quantum bit (qubit). A single, neutral atom behaving as a two-level system can be used as a qubit. Compared to ions, neutral atoms have the advantage that they are highly decoupled from electro-magnetic perturbations. However, coupling two qubits using neutral atoms is difficult to achieve. One approach for such coupling makes use of the Rydberg blockade \[29, 30, 31, 32, 33, 34, 35\]. In another approach, a cavity mode is used to couple atoms held inside the cavity \[36, 37, 38, 39\]. A key parameter in this approach is the single photon Rabi frequency, which must be much larger than atomic and cavity decay rates. This constraint can only be met by making the cavity very small, which in turn makes it difficult to hold many qubits inside.

One approach for circumventing this constraint is to make use of atomic ensembles. The single photon Rabi frequency for an ensemble scales as \(\sqrt{N}\), where \(N\) is the number of atoms, thus making it possible to make use of a much larger cavity. However, in order to use an ensemble for quantum computing, it is necessary to ensure that it behaves as an effective two-level system.
When exposed to only a single photon (or in a Raman transition, where one leg is exposed to a single photon), an ensemble of two-level atoms does indeed behave like a single two-level system. This property has been used to realize quantum memory elements using such an ensemble [40, 41]. However, any protocol that aims to create a two qubit logic gate (such as a CNOT gate) between two ensembles, necessary for realizing a quantum computer, must make use of additional, classical laser fields. Under such excitations, an ensemble no longer behaves like a two-level system. Instead, it exhibits a cascade of energy levels that are equally spaced. When exposed to a classical field, all levels in the cascade get excited [1], making it impossible to realize a quantum logic gate. In order to overcome this constraint, it is necessary to create conditions under which the cascade is truncated to a two-level system.

Previously, our group had proposed a scheme for producing such a blockade, using imbalances in light shifts experienced by the collective states [42, 43]. In that model, the light shifts were calculated by using a perturbation method, keeping terms up to second order in laser intensity. However, it turns out that when the collective excitation is viewed as a product of individual atomic states, an accurate representation for classical laser fields, and in the absence of any interaction between the atoms, the blockade effect disappears. We have verified this conclusion by numerically simulating the evolution of collective states for small values of $N$. It is still possible to produce such a blockade for a laser field described as a superposition of photon number states. However, when the mean photon number in such a field is very large, such as in a classical laser field, the blockade tends to vanish. Thus, in order to produce a blockade under excitation with a classical laser field, we must make use of some interaction between the atoms. In this
chapter, we propose to make use of interaction induced via excitation to Rydberg states to achieve this goal.

The rest of the chapter is organized as follows. In Section 2.2 we review briefly the formulation of collective excitation of lambda-type atoms. In Section 2.3 we summarize the model we had developed previously for light shift blockade (LSB) of collective excitation using second order perturbation approximation. In Section 2.4 we discuss how an alternative formulation of collective excitation allows us to determine the effect of light shift exactly, and identify conditions under which LSB is not possible. In particular, we show that when all excitation fields are classical, there is no blockade. In Section 2.5 we show how the interaction between two Rydberg states can be used to realize LSB even under classical excitation. In section Section 2.6 we generalize this process for $N$ atoms and show how LSB works for $N$-atom ensembles. Finally, in Section 2.7 we summarize our results, and present an outlook for using this approach for realizing a multi-qubit quantum computer.

2.2. Collective State Model

In order to avoid the deleterious effect of spontaneous emission, it is useful to realize a qubit based on two states that are long-lived. A convenient example for such a system consists of a Zeeman sublevel in one of the ground hyperfine state (e.g. $m_F = 0, F = 1, \, 5^2S_{1/2}$ in $^{87}$Rb) and another Zeeman sublevel in another ground hyperfine state (e.g. $m_F = 0, F = 2, \, 5^2S_{1/2}$ in $^{87}$Rb). These levels can be coupled by two laser fields to an intermediate state (e.g. $m_F = 1, F = 2, \, 5^2P_{1/2}$ in $^{87}$Rb). When the interaction is highly
Figure 2.1. Three-level scheme of single atom in an ensemble.

detuned with respect to the intermediate state, the laser fields cause a Raman transition between the two low lying states, thus producing an effective two-level system.

This is generally known as the Λ-system, illustrated schematically in Fig. 2.1. Here, the two ground states are $|a\rangle$ and $|c\rangle$, and the intermediate state is $|g\rangle$. The states $|a\rangle$ and $|g\rangle$ are coupled by a field with a Rabi frequency of $\Omega_1$ and a detuning of $\delta_1$. Likewise, states $|c\rangle$ and $|g\rangle$ are coupled by a field with a Rabi frequency of $\Omega_2$ and a detuning of $\delta_2$.

In the basis of states $|a\rangle$, $|c\rangle$ and $|g\rangle$, the Hamiltonian under electric dipole and rotating wave approximation, and rotating wave transportation, is given by

\[
\tilde{H} = \hbar \begin{bmatrix}
\Delta/2 & 0 & \Omega_1/2 \\
0 & -\Delta/2 & \Omega_2/2 \\
\Omega_1/2 & \Omega_2/2 & -\delta
\end{bmatrix},
\]

where $\delta \equiv (\delta_1 + \delta_2)/2$ is the average detuning and $\Delta \equiv (\delta_1 - \delta_2)$ is the two-photon detuning. In what follows, we will assume that $\delta$ is very large compared to both $\Omega_1$ and $\Omega_2$, as well as the decay rate, $\Gamma$, of the state $|g\rangle$. We will further assume that the two lasers are co-propagating.
For $N$ such non-interacting atoms, the ensemble can be modeled using symmetric collective states, also known as symmetric Dicke states [1]. The first few states are defined as follows:

\[
\begin{align*}
|A\rangle & \equiv |a_1, a_2, \ldots, a_N\rangle, \\
|G_1\rangle & \equiv \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |a_1, a_2, \ldots, g_j, \ldots, a_N\rangle, \\
|C_1\rangle & \equiv \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |a_1, a_2, \ldots, c_j, \ldots, a_N\rangle, \\
|G_2\rangle & \equiv \frac{1}{\sqrt{N}} \sum_{j,k (j \neq k)}^{N} |a_1, a_2, \ldots, g_j, \ldots, g_k, \ldots, a_N\rangle, \\
|C_2\rangle & \equiv \frac{1}{\sqrt{N}} \sum_{j,k (j \neq k)}^{N} |a_1, a_2, \ldots, c_j, \ldots, c_k, \ldots, a_N\rangle, \\
|G_{1,1}\rangle & \equiv \frac{1}{\sqrt{2^N C_2}} \sum_{j,k (j \neq k)}^{2^N C_2} |a_1, a_2, \ldots, g_j, \ldots, g_k, \ldots, a_N\rangle, \\
|G_{2,1}\rangle & \equiv \frac{1}{\sqrt{3^N C_3}} \sum_{j,k,l (j \neq k \neq l)}^{3^N C_3} |a_1, a_2, \ldots, g_j, \ldots, g_k, \ldots, c_l, \ldots, a_N\rangle, \\
|G_{1,2}\rangle & \equiv \frac{1}{\sqrt{3^N C_3}} \sum_{j,k,l (j \neq k \neq l)}^{3^N C_3} |a_1, a_2, \ldots, g_j, \ldots, c_k, \ldots, c_l, \ldots, a_N\rangle.
\end{align*}
\]

where $N C_M \equiv \binom{N}{M} \equiv N!/\left[M!(N-M)!ight]$. 

In Ref [44], we have shown that the system remains confined to a generalized form of these symmetric collective states, independent of the relative separation between the atoms (and hence the size of the ensemble), as long as it is assumed that each atom sees the same amplitude of the Rabi frequency, and the same laser frequency (i.e., any residual Doppler shift of the Raman transition frequency due to the motion of the atoms is negligible). The generalized form of the symmetric states are formally the same as those in Eqn. (2.2), except that the excited states incorporate the relevant spatial phases.
Figure 2.2. Schematic illustration of the relevant collective states and the corresponding coupling rates.

of the fields at the location of a given atom. This can be understood by noting that any phase factors accompanying the Rabi frequencies in the Hamiltonian of Eqn. (2.1) can be transformed out to produce a version of the Hamiltonian where the Rabi frequencies are real. The transformation necessary for this transfers the phases to the basis states. We refer the reader to Ref. [?] for details.

The collective states of Eqn. (2.2) are illustrated schematically in Fig. 2.2. Here, for example, $|G_1\rangle$ represents a state where only one atom on average is excited to state $|g\rangle$, with the rest remaining in state $|a\rangle$. Similarly, $|C_1\rangle$ represents a state where only one atom on average is excited to state $|c\rangle$, with the rest remaining in state $|a\rangle$, and so on. In our blockade scheme, we try to confine the system to the two lowest energy states $|A\rangle$ and $|C_1\rangle$. If we could achieve this and minimize the excitations to the first few higher energy states, then excitations to even higher states will be almost nonexistent. It can be shown
that the total number of symmetric states is $N_S = (N + 2)!/2N!$. For large $N$, $N_S = N^2/2$ so that the size of the Hamiltonian scales as $N^4$. Thus, an analysis of the evolution of the complete system exactly in this picture is computationally intractable. However, a plausible way to explore the possibility of finding the condition for the blockade is to truncate the system to a small size, and show that the excitation to the excluded states are negligible.

Here, we choose to truncate the system to six levels: $|A\rangle$, $|G_1\rangle$, $|C_1\rangle$, $|G_{1,1}\rangle$, $|C_2\rangle$ and $|G_{1,2}\rangle$. If the condition we find for the blockade shows negligible excitation to states that have non-zero coupling to the excluded states, the truncation would then be justified.

The Hamiltonian for these states can be expressed as \[ H = \hbar \begin{bmatrix}
\frac{\Delta}{2} & \frac{\sqrt{N-2}\Omega_1}{2} & 0 & 0 & 0 & 0 \\
\frac{\sqrt{N-2}\Omega_1}{2} & -\delta & \frac{\Omega_2}{2} & 0 & 0 & 0 \\
0 & \frac{\Omega_2}{2} & -\frac{\Delta}{2} & \frac{\sqrt{N-1}\Omega_1}{2} & 0 & 0 \\
0 & 0 & \frac{\sqrt{N-1}\Omega_1}{2} & -(\delta + \Delta) & \frac{\sqrt{2}\Omega_2}{2} & 0 \\
0 & 0 & 0 & \frac{\sqrt{2}\Omega_2}{2} & -\frac{3\Delta}{2} & \frac{\sqrt{N-2}\Omega_1}{2} \\
0 & 0 & 0 & 0 & \frac{\sqrt{N-2}\Omega_1}{2} & -(\delta + 2\Delta)
\end{bmatrix} \]

2.3. Original Model for Light Shift Blockade

The Hamiltonian in Eqn. (2.3) can be further simplified by adiabatically eliminating the states $|G_1\rangle$, $|G_{1,1}\rangle$, and $|G_{1,2}\rangle$ when $\delta \gg \sqrt{N}\Omega_1$, $\Omega_2$, $\Delta$, and $N \gg 1$. The reduced
Hamiltonian in the basis of states $|A\rangle$, $|C_1\rangle$ and $|C_2\rangle$ is

\[
\tilde{H} = \hbar \begin{bmatrix}
\varepsilon_A + \Delta/2 & \Omega/2 & 0 \\
\Omega/2 & \varepsilon_{C1} - \Delta/2 & \sqrt{\frac{2(N-1)}{N}}\Omega/2 \\
0 & \sqrt{\frac{2(N-1)}{N}}\Omega/2 & \varepsilon_{C2} - 3\Delta/2
\end{bmatrix},
\]

where $\varepsilon_A = N\Omega_1^2/4\delta$, $\varepsilon_{C1} = [(\Omega_2^2 + (N-1)\Omega_1^2)/4\delta$, and $\varepsilon_{C2} = [2\Omega_2^2 + (N-2)\Omega_1^2]/4\delta$ are the lowest order light-shifts of the states $|A\rangle$, $|C_1\rangle$ and $|C_2\rangle$ respectively, and $\Omega \equiv \sqrt{N}\Omega_1\Omega_2/2\delta$ is the Raman Rabi frequency. We can work out the LSB conditions with this Hamiltonian. By making the light shifts in the states $|A\rangle$ and $|C_1\rangle$ equal and the shift in $|C_2\rangle$ highly detuned from them, we can eliminate the excitation to $|C_2\rangle$.

The states $|A\rangle$ and $|C_1\rangle$ are resonant when $\Delta = \varepsilon_{C1} - \varepsilon_A \approx (\Omega_2^2 - \Omega_1^2)/4\delta$. Upon subtraction of a suitably chosen term $(\varepsilon_A + \Delta/2)$ from the diagonal term in the Hamiltonian and the approximation that $N \gg 1$, we get

\[
\tilde{H} = \hbar \begin{bmatrix}
0 & \Omega/2 & 0 \\
\Omega/2 & 0 & \Omega/\sqrt{2} \\
0 & \Omega/\sqrt{2} & \Delta_B
\end{bmatrix},
\]

where the blockade shift is defined as $\Delta_B \equiv (\varepsilon_{C2} - \varepsilon_{C1}) - (\varepsilon_{C1} - \varepsilon_A)$. This quantity vanishes for the first order values of the light shifts $\varepsilon_A$, $\varepsilon_{C1}$, and $\varepsilon_{C2}$ shown above, so that there is no blockade effect. However, to second order approximation, the blockade shift is $\Delta_B = -(\Omega_1^4 + \Omega_2^4)/(8\delta^3)$. If we operate under condition where $\Delta_B \gg \Omega/\sqrt{2}$, the transition to $|C_2\rangle$ becomes inconsequentially small and the ensemble of atoms oscillates between the collective states $|A\rangle$ and $|C_1\rangle$. 
Figure 2.3. Exact numerical solution of the evolution of the states using the LSB parameters (in units of $\Gamma$): $\Omega_1 = 0.001$, $\Omega_2 = 100$, $N = 2500$, $\delta = 1000$ and $\Delta = 2.497$. The plot is for $5\pi$ oscillations. The vertical axis is the population of the indicated collective state.

We have also determined numerically, for $N = 2500$, the evolution of the population for the six collective states in the truncated system, using the Hamiltonian of Eq. (2.3), without resorting to adiabatic elimination. The results are illustrated in Fig. 2.3 for a set of parameters that satisfy the LSB condition identified above. As can be seen from this figure, nearly all the population stays between levels $|A\rangle$ and $|C_1\rangle$, undergoing Rabi oscillations between them. The residual excitations of the other four states are very small, and can be made smaller by using weaker Rabi frequencies. Note that we have ignored the decay of the $|g\rangle$ states (at the rate of $\Gamma$), which is a valid approximation for $\delta \gg \Gamma$. 
2.4. Limitations of the Original Model for Light Shift Blockade

In the preceding section, we showed that the numerical simulation of the truncated system appears to validate the LSB process. For a large value of $N$, this result is still an approximation. However, the system can be modeled exactly for very small values of $N$. In particular, if we choose $N = 2$, there are only 6 collective states altogether. Thus, it is possible to check without truncation whether the LSB process holds in this case. Referring back to Fig. 2.2, the complete set of collective states for $N = 2$ consists of $|A\rangle$, $|G_1\rangle$, $|C_1\rangle$, $|G_{1,1}\rangle$, $|C_2\rangle$ and $|G_2\rangle$. We determined the evolution of this system numerically, starting with the system being in the $|A\rangle$ state. The results are illustrated in Fig. 2.4. In Fig. 2.4a, we show the population of the collective states under the approximation that the state $|G_2\rangle$ can be neglected completely, since $\delta \gg \sqrt{N_1} \Omega_1$ and $\delta \gg \Omega_2$, corresponding to very small populations in states $|G_1\rangle$ and $|G_{1,1}\rangle$. As can be seen, the result is consistent with LSB, since the maximum population of $|C_2\rangle$ is very small. In Fig. 2.4b, we relax this approximation, and keep the state $|G_2\rangle$ in the system. This produces an apparently surprising result. The population in $|C_2\rangle$ can now reach almost unity for some interaction time. Thus, the LSB process is strongly violated. It should be noted that the maximum population of $|G_2\rangle$ is negligible (Fig. 2.4a), so that ignoring the excitation to $|G_2\rangle$ seems to be a reasonable one. Yet, the relaxation of this approximation modifies the population dynamics in a very significant way.

In order to understand this behavior, it is instructive first to consider the process of collective excitation more explicitly. Specially, it can be shown that, for excitation by semi-classical fields, and in the absence of interaction between the atoms, the general quantum state of an ensemble is always given by the outer (tensor) product of the quantum
states of the individual atoms \[45\]. The collective states representation of the evolution of such a system is merely an alternative way of describing the process. To illustrate this explicitly, let us consider a case involving two-level atoms, with \(|a\rangle\) and \(|c\rangle\) being the lower and higher energy levels respectively.

Let us denote by \(|\psi_i\rangle\) the quantum state of the \(i\)-th atom. Then, the total quantum state of the system, \(|\Psi\rangle\), is given by: \(|\Psi\rangle = N \prod_{i=1}^{N} |\psi_i\rangle\). Thus, if we write \(|\psi_i\rangle = \alpha_i |a_i\rangle + \beta_i |c_i\rangle\), then \(|\Psi\rangle = \prod_{i=1}^{N} (\alpha_i |a_i\rangle + \beta_i |c_i\rangle)\). For simplicity, let us assume that \(N = 2\). We then get: \(|\Psi\rangle = (\alpha_1 |a_1\rangle + \beta_1 |c_1\rangle) (\alpha_2 |a_2\rangle + \beta_2 |c_2\rangle)\). Consider the product state basis which is spanned by \(|a_1a_2\rangle, |a_1c_2\rangle, |c_1a_2\rangle\) and \(|c_1c_2\rangle\). The total state can thus be written as

Figure 2.4. Numerical solution of the evolution of the collective states of two atoms. Here, \(\Omega_1 = 0.001, \Omega_2 = 100, N = 2500, \delta = 1000\) and \(\Delta = 2.497\) (in units of \(\Gamma\)). (a): Collective states of two atoms when \(|G_2\rangle\) is eliminated. (b): Collective states of two atoms with the full Hamiltonian.
\[
|\Psi\rangle = \alpha_1 \alpha_2 |a_1 a_2\rangle + \alpha_1 \beta_2 |a_1 c_2\rangle + \beta_1 \alpha_2 |c_1 a_2\rangle + \beta_1 \beta_2 |c_1 c_2\rangle
\]

(2.6)

Consider next the complete collective state basis spanned by \(|a_1 a_2\rangle, |+\rangle = (|a_1 c_2\rangle + |c_1 a_2\rangle)/\sqrt{2}, |−\rangle = (|a_1 c_2\rangle - |c_1 a_2\rangle)/\sqrt{2}, and |c_1 c_2\rangle\). This basis is simply related to the product state basis by a 45 rotation in the plane of \(|a_1 c_2\rangle\) and \(|c_1 a_2\rangle\), so that the rotation matrix can be written as

\[
R = \begin{pmatrix}
1 & 0 & 0 & 0 \\
0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\
0 & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} & 0 \\
0 & 0 & 0 & 1
\end{pmatrix}
\]

(2.7)

Thus, the total state in the collective state basis can be written as

\[
|\Psi\rangle_c = R |\Psi\rangle = \begin{pmatrix}
\alpha_1 \alpha_2 \\
(\alpha_1 \beta_2 + \beta_1 \alpha_2)/\sqrt{2} \\
(\alpha_1 \beta_2 - \beta_1 \alpha_2)/\sqrt{2} \\
\beta_1 \beta_2
\end{pmatrix}
\]

(2.8)
Similarly, we can represent the Hamiltonian in these different bases. In the rotating wave picture, the Hamiltonian for a single atom can be expressed as

\[
H_1 = \hbar \begin{bmatrix}
0 & \Omega/2 \\
\Omega/2 & -\delta \\
\end{bmatrix},
\]

where \( \Omega \) is the Rabi frequency and \( \delta = \omega - (\omega_c - \omega_a) \) is the detuning of the laser frequency from the resonance frequency of the two states. When there are two atoms, the Hamiltonian in the basis of states \( |a_1 a_2\rangle, |a_1 c_2\rangle, |c_1 a_2\rangle \) and \( |c_1 c_2\rangle \) is

\[
H = H_1 \otimes I_2 + I_1 \otimes H_2
\]

where \( I_i \) is the identity matrix and \( H_i \) is the Hamiltonian for the \( i \)-th atom. For example,

\[
\langle a_1 a_2 | H | c_1 a_2 \rangle = \langle a_1 a_2 | (H_1 \otimes I_2) | c_1 a_2 \rangle + \langle a_1 a_2 | (I_1 \otimes H_2) | c_1 a_2 \rangle
\]

\[
= \langle a_1 | H_1 | c_1 \rangle \langle a_2 | H_1 | a_2 \rangle + \langle a_1 | I_1 | c_1 \rangle \langle a_2 | H_2 | a_2 \rangle
\]

\[
= \langle a_1 | H_1 | c_1 \rangle = \Omega_1 / 2
\]

Thus, the Hamiltonian can be written as

\[
H = \hbar \begin{bmatrix}
0 & \Omega_2/2 & \Omega_1/2 & 0 \\
\Omega_2/2 & -\delta & 0 & \Omega_1/2 \\
\Omega_1/2 & 0 & -\delta & \Omega_2/2 \\
0 & \Omega_1/2 & \Omega_2/2 & -2\delta
\end{bmatrix},
\]
where the Rabi frequencies are assumed to be real. Under a 45° rotation in the plane of $|a_1c_2\rangle$ and $|c_1a_2\rangle$, the new Hamiltonian in the basis $|a_1a_2\rangle, |+\rangle, |-\rangle, |c_1c_2\rangle$ is

$$H' = R^{-1}HR$$

(2.11)

$$H' = \hbar \begin{bmatrix} 0 & \frac{\Omega_1 + \Omega_2}{2\sqrt{2}} & -\frac{\Omega_1 - \Omega_2}{2\sqrt{2}} & 0 \\
\frac{\Omega_1 + \Omega_2}{2\sqrt{2}} & -\delta & 0 & \frac{\Omega_1 + \Omega_2}{2\sqrt{2}} \\
-\frac{\Omega_1 - \Omega_2}{2\sqrt{2}} & 0 & -\delta & \frac{\Omega_1 - \Omega_2}{2\sqrt{2}} \\
0 & \frac{\Omega_1 + \Omega_2}{2\sqrt{2}} & -\frac{\Omega_1 - \Omega_2}{2\sqrt{2}} & -2\delta \end{bmatrix}.$$  

For $\Omega = \Omega_1 = \Omega_2$, the asymmetric state, $|-\rangle$, is decoupled from the other states, and the Hamiltonian becomes

$$H' = \hbar \begin{bmatrix} 0 & \sqrt{2}\Omega/2 & 0 & 0 \\
\sqrt{2}\Omega/2 & -\delta & 0 & \sqrt{2}\Omega/2 \\
0 & 0 & -\delta & 0 \\
0 & \sqrt{2}\Omega/2 & 0 & -2\delta \end{bmatrix}.$$  

(2.12)

The Hamiltonian in Eqn. (2.12) describes the situation where only symmetric collective states are excited.

This is also evident by noting that the general collective state can now be expressed as $|\Psi\rangle_c = \alpha^2 |aa\rangle + \sqrt{2}\alpha\beta |+\rangle + \beta^2 |cc\rangle$, where $\alpha = \alpha_1 = \alpha_2$ and $\beta = \beta_1 = \beta_2$ (since $\Omega_1 = \Omega_2$). The form of this state shows clearly that it is impossible to suppress excitation to the $|cc\rangle$ state while still exciting the $|+\rangle$ state. Thus, the degree of excitation of a given collective state is related to the degree of excitation of all other collective states. While the three-level system we are considering is more complicated in the details, this fundamental rule still holds. As such, under this set of conditions (i.e. semiclassical laser
field, and no interaction between the atoms) it is not possible to block the excitation to state $|C_2\rangle$ while allowing for excitation of state $|C_1\rangle$. The result shown in Fig. 2.4b is merely a manifestation of this constraint. The subtle error that led us to the previous conclusion about the realizability of LSB was the approximation that the role of $|G_2\rangle$ is negligible. This approximation was entirely logical in a general sense, but turns out, rather surprisingly, not to be valid.

Of course, if the laser field is treated quantum mechanically, by considering it as a superposition of Fock states, the quantum state of the atoms and the photons are inherently entangled. As such, the state of the ensemble cannot be expressed as a product of the states of each atom. Under such a situation, it should in principle be possible to achieve the blockade effect. However, such a blockade works in a clean manner only when the numbers of photons are limited to a few. As discussed earlier, our objective is to achieve a blockade when the laser field has a mean photon number much larger than unity, i.e. the semi-classical limit. In this limit, the only way to achieve a blockade is to allow for interaction between the atoms. Here we describe a scheme where interactions between Rydberg excited levels are used to achieve the LSB effect.

### 2.5. Rydberg Assisted LSB of Two Atoms

We modify the lambda scheme of a single atom by adding a Rydberg level $|r\rangle$ and an intermediate level $|d\rangle$, which is coupled to $|r\rangle$ and $|c\rangle$, but not to $|a\rangle$, as illustrated in Fig. 2.5a. We denote as $\hbar\omega_j$ the energy of the state $|j\rangle$, for $j = a, g, c, d$ and $r$. The Rabi frequencies are denoted as $\Omega_1$, $\Omega_2$, $\Omega_3$ and $\Omega_4$ for the $a \rightarrow g$, $g \rightarrow c$, $c \rightarrow d$ and $d \rightarrow r$ transitions, respectively. For convenience, we also define the relevant detunings as
\[ \delta_1 = \omega_1 - (\omega_b - \omega_a), \quad \delta_2 = \omega_2 - (\omega_b - \omega_c), \quad \delta_3 = \omega_3 - (\omega_d - \omega_c) \] and \[ \delta_4 = \omega_4 - (\omega_r - \omega_d). \]

As before, the average detuning for the \( \Lambda \)-transition is defined as \( \delta = (\delta_1 + \delta_2)/2 \), and the corresponding two photon detuning is defined as \( \Delta = \delta_2 - \delta_1 \). We also define as \( \delta_r = \delta_3 + \delta_4 \) to be the two photon detuning for the ladder transition \( c \rightarrow d \rightarrow r \). After making the usual dipole and rotating wave approximations and upon making the rotating wave transformation, the Hamiltonian in the basis of states \( |a\rangle, |g\rangle, |c\rangle, |d\rangle \) and \( |r\rangle \) can
be expressed as

\[
H_{1R} = \hbar \begin{bmatrix}
\Delta & \Omega_1/2 & 0 & 0 & 0 \\
\Omega_1/2 & -\delta + \Delta/2 & \Omega_2/2 & 0 & 0 \\
0 & \Omega_2/2 & 0 & \Omega_3/2 & 0 \\
0 & 0 & \Omega_3/2 & -\delta_3 & \Omega_4/2 \\
0 & 0 & 0 & \Omega_4/2 & -\delta_r 
\end{bmatrix}.
\]

(2.13)

To illustrate the basic concept, we consider first the collective states of only two atoms, with a distance \( r_{12} \) which is assumed to be comparable to the characteristic distance scale of interatomic Rydberg interaction.

For simplicity, we consider first the symmetric collective states of two atoms, as illustrated in Fig. 2.5b, where we have adopted the compact notation that, for example, \( |AA\rangle = |aa\rangle \), \( |CC\rangle = |cc\rangle \), \( |AC\rangle = (|ac\rangle + |ca\rangle) / \sqrt{2} \) and so on. Since the Hamiltonian for the two atoms now contains the interaction between the two atoms, the general quantum
state of the total system can no longer be written as a product between the quantum states of individual atoms. As such, it should now be possible to produce the LSB effect. Specifically, note that the dipole-dipole interaction between the atoms when they are both excited to the Rydberg state will shift the energy of the $|RR\rangle$ state compared to its value when the atoms are far apart. Since there is an asymmetry in the degree to which the $|r\rangle$ state is coupled to $|a\rangle$ and $|c\rangle$, the shift in the energy of $|RR\rangle$ will affect differently the light shifts experienced by $|AA\rangle$, $|AC\rangle$ and $|CC\rangle$. This is precisely what is needed for realizing LSB. In what follows, we derive analytically, under adiabatic elimination approximation, the parameters needed for realizing the optimal LSB condition. We then verify the results via exact numerical calculation. This is followed by a derivation of the condition needed for optimal LSB for an arbitrary value of $N$, the number of atoms in the ensemble.

As can be seen from Fig. 2.5b, there are fifteen symmetric collective states for two atoms. In order to establish an approximate analytical result (which would then serve...
as a guide for choosing parameters for exact numerical calculation), we first simplify the picture by reducing the 5-level system for each atom (see Fig. 2.6a) to an effective 3-level system (see Fig. 2.6b) via eliminating adiabatically two of the intermediate states, \(|g\rangle\) and \(|d\rangle\), that are highly detuned. Once this is done, the effective Hamiltonian for each atom, in the basis of \(|a\rangle\), \(|c\rangle\) and \(|r\rangle\), can be expressed as

\[
H'_{1R} = \hbar \begin{bmatrix}
\Delta + \varepsilon_a & \Omega_{ac}/2 & 0 \\
\Omega_{ac}/2 & \varepsilon_c & \Omega_{cr}/2 \\
0 & \Omega_{cr}/2 & -\delta_r + \varepsilon_r
\end{bmatrix},
\]

where \(\Omega_{ac} = \Omega_1\Omega_2/2\delta\) is the Raman-Rabi frequency of transition \(|a\rangle \rightarrow |c\rangle\), and \(\Omega_{cr} = \Omega_3\Omega_4/2\delta_3\) is the two-photon Rabi frequency of transition \(|c\rangle \rightarrow |r\rangle\), while \(\varepsilon_a = \Omega_1^2/4\delta\), \(\varepsilon_c = \Omega_2^2/4\delta + \Omega_3^2/4\delta_3\) and \(\varepsilon_r = \Omega_4^2/4\delta_3\) are the light shifts of states \(|a\rangle\), \(|c\rangle\) and \(|r\rangle\) respectively. If we define two new parameters \(\Delta_{ac} = \Delta + \varepsilon_a - \varepsilon_c\) and \(\Delta_{cr} = \delta_r + \varepsilon_c - \varepsilon_r\), these become the effective, relevant detunings between the levels. Then we can rewrite the single atom Hamiltonian in the basis of \(|a\rangle\), \(|c\rangle\) and \(|r\rangle\) as

\[
H'_{1R} = \hbar \begin{bmatrix}
\Delta_{ac} & \Omega_{ac}/2 & 0 \\
\Omega_{ac}/2 & 0 & \Omega_{cr}/2 \\
0 & \Omega_{cr}/2 & -\Delta_{cr}
\end{bmatrix}.
\]

If the distance between the two atoms, \(r_{12}\), is much larger than the scale of Rydberg interaction, the combined Hamiltonian in the basis of the nine product states (\(|a_1a_2\rangle\), \(|a_1c_2\rangle\), \(|a_1r_2\rangle\), \(|c_1a_2\rangle\), \(|c_1c_2\rangle\), \(|c_1r_2\rangle\), \(|r_1a_2\rangle\), \(|r_1c_2\rangle\), \(|r_1r_2\rangle\)) can be written as \(H_T = H'_{1R} \otimes I_2 + I_1 \otimes H'_{2R}\), and the 81 elements of \(H_T\) can be easily calculated in the same manner as used.
in deriving Eqn. (10). When transformed to the collective state picture, the asymmetric
states become decoupled, just as before, and we are left with a six state system spanned
by \(|AA\rangle, |AC\rangle, |CC\rangle, |AR\rangle, |CR\rangle\) and \(|RR\rangle\) (using the compact notation introduced in
Fig. 2.5b), which are shown in Fig. 2.7, and the Hamiltonian can be expressed as

\[
H_T' = \hbar \begin{bmatrix}
2\Delta_{ac} & \frac{\sqrt{2}}{2} \Omega_{ac} & 0 & 0 & 0 & 0 \\
\frac{\sqrt{2}}{2} \Omega_{ac} & \Delta_{ac} & \frac{\sqrt{2}}{2} \Omega_{ac} & \frac{1}{2} \Omega_{cr} & 0 & 0 \\
0 & \frac{\sqrt{2}}{2} \Omega_{ac} & 0 & 0 & \frac{\sqrt{2}}{2} \Omega_{cr} & 0 \\
0 & \frac{1}{2} \Omega_{cr} & 0 & \Delta_{ac} - \Delta_{cr} & \frac{1}{2} \Omega_{ac} & 0 \\
0 & 0 & \frac{\sqrt{2}}{2} \Omega_{cr} & \frac{1}{2} \Omega_{ac} & -\Delta_{cr} & \frac{\sqrt{2}}{2} \Omega_{cr} \\
0 & 0 & 0 & 0 & \frac{\sqrt{2}}{2} \Omega_{cr} & -2\Delta_{cr}
\end{bmatrix}. 
\]

When the distance \(r_{12}\) becomes comparable to the characteristic distance scale for in-
teratomic Rydberg interaction, the Hamiltonian for the collective states, \(H_{TR}'\), is the same
as \(H_T'\) except for the last diagonal element. Specifically,
\[
\langle RR| H_{TR}' |RR\rangle = \langle RR| H_T' |RR\rangle - V_r = -2\Delta_{cr} - V_r, 
\]
where \(V_r\) represents the dipole-dipole interaction between two atoms. Thus, we can write

\[
H_{TR}' = H_T' - V_R |RR\rangle \langle RR|. 
\]

The various terms of \(H_{TR}'\) are illustrated schematically in Fig. 2.7.

When we allow \(\Delta_{cr} \gg \Delta_{ac}, \Omega_{ac}, \Omega_{cr}\), the upper levels \(|AR\rangle, |CR\rangle\) and \(|RR\rangle\) can be
adiabatically eliminated. The reduced Hamiltonian in the basis of \(|AA\rangle, |AC\rangle\) and \(|CC\rangle\)
Figure 2.8. Evolution of population using the simplified two-atom picture in Fig. 2.6. Figure (a) represents the case when the dipole-dipole interaction is not present \((V_r = 0)\). Figure (b) represents the case when the dipole-dipole interaction is present \((V_r = 16)\).

is

\[
\hat{H}_{TR} \approx \hbar \begin{bmatrix}
2\Delta_{ac} & \frac{\sqrt{2}}{2}\Omega_{ac} & 0 \\
\frac{\sqrt{2}}{2}\Omega_{ac} & \Delta_{ac} + \frac{\Omega_{cr}}{2} \cdot v & \frac{\sqrt{2}}{2}\Omega_{ac} + \frac{\sqrt{2}}{2}\Omega_{cr} \cdot \frac{uv}{1-2vw} \\
0 & \frac{\sqrt{2}}{2}\Omega_{ac} + \frac{\sqrt{2}}{2}\Omega_{cr} \cdot \frac{uv}{1-2vw} & \Omega_{cr} \cdot \frac{v}{1-2vw}
\end{bmatrix},
\]

where, for simplicity, we have defined \(u = \Omega_{ac}/2\Delta_{cr}\), \(v = \Omega_{cr}/2\Delta_{cr}\), \(w = \Omega_{cr}/2(2\Delta_{cr} + V_r)\), and we have assumed that \(\Omega_{cr} \gg \Omega_{ac}\). In order to make the levels \(|AA\rangle\) and \(|AC\rangle\) resonant, we enforce the condition that \(\Delta_{ac} = \Omega_{cr} \cdot v/2\), which leads to \(\Omega_{cr}^2 = 4\Delta_{ac}\Delta_{cr}\). When
the energy levels are all reduced by $2\Delta_{ac}$, Eqn. (2.18) becomes

\begin{equation}
\tilde{H}_{TR}' = \hbar \begin{pmatrix}
0 & \frac{\sqrt{2}}{2} \Omega_{ac} & 0 \\
\frac{\sqrt{2}}{2} \Omega_{ac} & 0 & \frac{\sqrt{2}}{2} \Omega_{ac} + \frac{\sqrt{2}}{2} \Omega_{cr} \cdot \frac{uv}{1-2vw} \\
0 & \frac{\sqrt{2}}{2} \Omega_{ac} + \frac{\sqrt{2}}{2} \Omega_{cr} \cdot \frac{uv}{1-2vw} & \Delta_B
\end{pmatrix},
\end{equation}

where $\Delta_B \equiv \Omega_{cr} \cdot 2vw/(1-2vw)$ is the blockade shift. When $\Delta_B$ is much larger than the coupling between the states $|AC\rangle$ and $|CC\rangle$, we are able to block the excitation to state $|CC\rangle$ and achieve LSB. This can be achieved under the condition where $V_r + 2\Delta_{cr} \ll \Omega_{cr}/2\Omega_{ac}\Delta_{cr}$. When these conditions are met, we achieve resonance between states $|AA\rangle$ and $|AC\rangle$, blocking excitation to state $|CC\rangle$.

In order to verify the validity of this conclusion, we have simulated the evolution of the three-level system of two atoms (i.e. the system shown in Fig. 2.6b), using the 6 $\times$ 6 collective state Hamiltonian, $H_{TR}'$ (Eqn. (2.17)), which included the effect of Rydberg interaction, but without making use of the adiabatic elimination of states $|AR\rangle$, $|CR\rangle$ and $|RR\rangle$. The parameters we have used are $\Omega_{ac} = 0.00002$, $\Omega_{cr} = 1$, $\Delta_{ac} = -0.031129$ and $\Delta_{cr} = -8$ (in units of $\Gamma$), consistent with the requirement of achieving LSB. The result of this simulation is shown in Fig. 2.8. Fig. 2.8a represents the case when the Rydberg-interaction parameter, $V_r$ is set to zero. In this case, the maximum amplitude of $|CC\rangle$ reaches unity. When $V_r = 16$, the maximum amplitude of $|CC\rangle$ is nearly zero, and the system oscillates between $|AA\rangle$ and $|AC\rangle$, as shown in Fig. 2.8b. It should also be noted that under this blockade condition, the oscillation frequency between levels $|AA\rangle$ and $|AC\rangle$ is increased by $\sqrt{2}$. The upper levels $|AR\rangle$, $|CR\rangle$ and $|RR\rangle$ are minimally excited regardless of whether interaction is present or not. This justifies the adiabatic elimination
Figure 2.9. Evolution of population using the full two-atom picture in Fig. 2.6b when the dipole-dipole interaction is not present ($V_r = 0$).

of these states employed in deriving the $3 \times 3$ reduced Hamiltonian for the collective states, shown in Eqn. (2.18).

The parameters used in the evolution of the simplified two-atom Hamiltonian can be used to extract the values of parameters necessary for the exact two-atom 15-level system shown in Fig. 2.5b. We choose the parameters as follows: $\Omega_1 = 0.0004$, $\Omega_2 = 0.8$, $\delta = -8$, $\Delta = -0.0199$, $\Omega_3 = 20$, $\Omega_4 = 320$, $\delta_3 = -3200$, $\delta_4 = 3200$. Notice that here we make the choice that $\Delta \simeq (\Omega_2^2 - \Omega_1^2)/4\delta$ in order to produce full Rabi oscillations between $|AA\rangle$ and $|AC\rangle$. The results of the plots with and without the Rydberg interaction are shown in Fig. 2.9. Despite the fact that 15 levels are present, only the levels $|AA\rangle$, $|AC\rangle$
Figure 2.10. Evolution of population using the full two-atom picture in Fig. 2.6b when the dipole-dipole interaction is present ($V_r = 16$).

and $|CC\rangle$ are populated while the excitations to the other states remain under 1%. As was the case with the simplified Hamiltonian, the presence of the Rydberg interaction ($V_r = 16$) suppresses the excitation to level $|CC\rangle$ so that an effective two-level system is generated, as illustrated in Fig. 2.10.

2.6. Rydberg Assisted LSB in $N$-atom ensembles

This process can be generalized for $N$ atoms. Referring back to Fig. 2.6, we recall first that adiabatic elimination of states $|g\rangle$ and $|d\rangle$ reduces the system to three levels (Fig. 2.6b). The first six collective states involving these single atom states, for $N$-atoms,
are as follows

\[ |A\rangle \equiv |a_1, a_2, \ldots, a_N\rangle, \]

\[ |C_1\rangle \equiv \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |a_1, a_2, \ldots, c_j, \ldots, a_N\rangle, \]

\[ |C_2\rangle \equiv \frac{1}{\sqrt{N C_2}} \sum_{j,k(k\neq k)}^{N C_2} |a_1, a_2, \ldots, c_j, \ldots, c_k, \ldots, a_N\rangle, \]

(2.20)

\[ |R_1\rangle \equiv \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |a_1, a_2, \ldots, r_j, \ldots, a_N\rangle, \]

\[ |R_{1,1}\rangle \equiv \frac{1}{\sqrt{2^{N C_2}}} \sum_{j,k(k\neq k)}^{2^{N C_2}} |a_1, a_2, \ldots, r_j, \ldots, c_k, \ldots, a_N\rangle, \]

\[ |R_2\rangle \equiv \frac{1}{\sqrt{N C_2}} \sum_{j,k(k\neq k)}^{N C_2} |a_1, a_2, \ldots, r_j, \ldots, r_k, \ldots, a_N\rangle. \]

Of course, there are many more collective states. However, our goal is to find the condition where the system oscillates between \( |A\rangle \) and \( |C_1\rangle \), with negligible excitation to the remaining collective states. If we can show that the excitation to states \( |C_2\rangle, |R_1\rangle, |R_{1,1}\rangle \) and \( |R_2\rangle \) are negligible, then it follows that the excitation to all other higher energy collective states is also negligible. Thus, it is justified to limit our consideration to only these six states.

With the single atom Hamiltonian in the basis of \( |a\rangle, |c\rangle \) and \( |r\rangle \) shown in Eqn. (2.15), the Hamiltonian formed with states \( |A\rangle, |C_1\rangle, |C_2\rangle, |R_1\rangle, |R_{1,1}\rangle \) and \( |R_2\rangle \) can be written
as

\[
H_{NR'} = \hbar \begin{bmatrix}
2\Delta_{ac} & \sqrt{N} \Omega_{ac} & 0 & 0 & 0 & 0 \\
\sqrt{N} \Omega_{ac} & \Delta_{ac} & \sqrt{2(N-1)} \Omega_{ac} & \Omega_{cr} & 0 & 0 \\
0 & \sqrt{2(N-1)} \Omega_{ac} & 0 & 0 & \sqrt{2} \Omega_{cr} & 0 \\
0 & \Omega_{cr} & 0 & \Delta_{ac} - \Delta_{cr} & \sqrt{N-1} \Omega_{ac} & 0 \\
0 & 0 & \sqrt{2} \Omega_{cr} & \sqrt{N-1} \Omega_{ac} & -\Delta_{cr} & \sqrt{2} \Omega_{cr} \\
0 & 0 & 0 & 0 & \sqrt{2} \Omega_{cr} & -2\Delta_{cr} - V_r \\
\end{bmatrix}.
\]

Under the condition that \(\Delta_{cr} \gg \Delta_{ac}, \sqrt{N} \Omega_{ac}, \Omega_{cr}\), for large \(N\), this reduces to

\[
\widetilde{H}_{NR'} = \hbar \begin{bmatrix}
0 & \sqrt{2} \Omega_{ac} & 0 & 0 & 0 \\
\sqrt{2} \Omega_{ac} & 0 & \sqrt{2} \Omega_{ac} + \Omega_{cr} & \frac{uv\sqrt{2(N-1)}}{1-(N-1)u^2-2vw} \\
0 & \sqrt{2} \Omega_{ac} & \sqrt{2} \Omega_{ac} + \Omega_{cr} & \frac{uv\sqrt{2(N-1)}}{1-(N-1)u^2-2vw} & \Delta_B \\
\end{bmatrix}.
\]

in the basis of \(|A\rangle\) and \(|C_1\rangle\) and \(|C_2\rangle\), where the first two levels were made resonant by choosing \(\Delta_{ac} = (\Omega_{cr}/2) \cdot v (1 - 2vw)/(1 - (N - 1) u^2 - 2vw)\). The blockade shift is now \(\Delta_B \equiv \Omega_{cr} \cdot 2vw/(1 - (N - 1) u^2 - 2vw)\). Note that when \(N = 2\), the Hamiltonian, the detuning, and the blockade shift are equivalent to the calculations made earlier for the two-atom case. The conditions necessary to block the excitation to state \(|C_2\rangle\) are \(\Omega_{cr} \gg \sqrt{N} \Omega_{ac}\) and \(w \gg \sqrt{N} u\), which again occur when \(V_r \rightarrow -2\Delta_{cr}\), just as in the case of \(N = 2\).

Fig. 2.11 shows the populations of the six collective states of Eqn. \(2.20\) under the LSB conditions found for 1000 atoms. The parameters are \(\Omega_{ac} = 0.00002/\sqrt{1000}\), \(\Omega_{cr} = 1\), \(\Delta_{ac} = -0.031129\), \(\Delta_{cr} = -8\) and \(V_r = 16\) (in units of \(\Gamma\)). As can be seen, states \(|A\rangle\) and
Figure 2.11. Evolution of population in the six lowest energy states of Hamiltonian in Eqn. (2.20) for $N = 1000$, with the same conditions as Fig. 2.8 except $\Omega_{ac}$ here is $\sqrt{N}$ smaller, and the dipole-dipole interaction $V_r = 16\Gamma$.

Figure 2.12. Evolution of population in the six lowest energy states of Hamiltonian in Eqn. (2.20), with the same conditions as Fig. 2.11 except the dipole-dipole interaction $V_r = 16000\Gamma$.

$|C_1\rangle$ are resonant, and population in state $|C_2\rangle$ is very small. With so little excitation into $|C_2\rangle$, the Rydberg assisted LSB guarantees the suppression of the higher excitations, thereby validating the use of a truncated Hamiltonian in Eqn. (2.20).
So far, we have shown that the Rydberg assisted LSB works for $V_r = 16\Gamma$, where $\Gamma$ is the decay rate of the state $|g\rangle$. Consider, for example, the specific case of $^{87}\text{Rb}$ atoms. In this case, $\Gamma \simeq 6\text{MHz}$, so that $V_r \simeq 96\text{MHz}$, which corresponds to an interatomic distance of $\sim 10\mu\text{m}$. We envision a scenario where the collective ensemble would be confined to a sphere with a diameter $\sim 10\mu\text{m}$, realizable, for example, by loading atoms from a MOT into a FORT (far-off resonance trap), containing about $10^3$ atoms. For some pair of atoms, the interatomic distance would be smaller than $10\mu\text{m}$. It is well known that $V_r$ scales approximately as $r^{-3}$, where $r$ is the interatomic distance between a pair of atoms for $r < 10\mu\text{m}$ [46]. Thus, for $r = 1\mu\text{m}$, $V_r \simeq 16000\Gamma \simeq 96 \times 10^3\text{MHz}$. We show in Fig. 2.12 that the Rydberg assisted LSB works for this value of $V_r$ for $N = 1000$ atoms.

2.7. Conclusion

The light shift imbalance induced blockade in an atomic ensemble had been studied previously, in which the difference in the light shifts produced in collective state energy levels leads to a condition where the system remains confined to a superposition of the ground and the first excited states. The significance of this result for quantum computing was discussed in Ref [47]. Upon further investigation into the nature of collective states, we found that the light shift imbalance alone is not enough to produce a blockade. By introducing Rydberg interaction, and using the technique of adiabatic elimination, we are able to establish the conditions under which the blockade can be achieved. Numerical simulations confirm the validity of this result.

The ensemble-based qubits realized in this manner can be used to implement a controlled-NOT (CNOT) gate, which is a universal gate for quantum computing, using a variation
of the Pellizzari scheme [36]. The details of the process for realizing a CNOT gate in this way, using $^{87}$Rb atoms are essentially the same as what was presented in Ref [43]. Many such gates can be linked to one another, via nearest neighbor quantum coupling, to realize an elementary quantum computer (EQC). The size of an EQC, contained inside a single vacuum chamber, is likely to be limited to a number of the order of ten. However, as shown in Ref [43], many such EQCs can be linked via optical fiber, using photons to transport quantum information, thus making this approach scalable. Of course, it is also possible to realize a CNOT gate between single atoms, caught in FORTs, by making use of Rydberg interactions [48]. However, it is very difficult to load a single atom consistently in a FORT. In contrast, the approach proposed here is relatively insensitive to the actual number of atoms held in the FORT. Thus, this approach may prove to be a more viable alternative for scalable quantum computing using neutral atoms.
CHAPTER 3

Off-resonant Raman-Rabi excitation based collective state atomic clock

3.1. Introduction

It is well known that the width of the fringes, observed as a function of the detuning, in a pulsed excitation of an atomic transition, is limited by the inverse of the interaction time. This effect is routinely observed in systems such as microwave or Raman atomic clocks [49, 50, 51, 52, 53]. It is also well known that the effective interaction time can be extended by employing Ramsey’s technique of separated field excitations [54]. In that case, the transit time limited linewidth is determined by the inverse of the time delay between the two fields. The temporal profile of the field envelope seen by the atoms is a pair of square pulses, each with a duration $T_1$, separated by $T_2$. For a conventional clock (CC), the Ramsey technique produces a sync function with a width of $\sim T_1^{-1}$, modulated by a sinusoid with a fringe width of $\sim T_2^{-1}$, all centered at the carrier frequency.

The width of these fringes can be reduced by making use of entanglement, as demonstrated by Wineland et al. using trapped ions [55]. Consider, for example, a situation where the use of entanglement allows one to couple the ground state of three particles to a state where all three particles are in the excited state, representing a collective excitation. This corresponds to an effective increase in the transition frequency by a factor of three. As such, the detuning for a single atom gets tripled for this collective excitation, so that
the width of the Ramsey fringe gets reduced by a factor of three. However, realizing such a scheme for a large number of particles is beyond the capability of current technology.

Here, we describe a scheme that produces Ramsey fringes that are narrower by a factor of more than $10^3$ for parameters that are readily accessible, without making use of entanglement. While the concept can be applied to other types of atomic clocks, as described later, the specific experiment we propose is an optically off-resonant Raman atomic clock using ensembles of $N$ cold atoms. The clock transition is detected by measuring one of the collective states rather than measuring individual atomic states. The fringes observed as a function of the Raman (i.e. two photon) detuning is found to be $\sim \sqrt{N}$ times narrower than the transit time limited width that would be seen by measuring individual atomic states, as is the case with the CC. For the current state of the art of trapped atoms, the value of $N$ can easily exceed $10^6$, so that a reduction of fringe width by a factor of more than $10^3$ is feasible.

The reduction in the width of the fringe, especially by such a large factor, strongly violates the conventional transit time limit of spectroscopic resolution. However, we show, via a detailed analysis of the standard quantum limit and the Heisenberg limit, that, indeed, this violation of the conventional transit-time limit is allowed, and is within the constraint of the more fundamental uncertainty principle of quantum mechanics. We also show that under certain conditions, frequency fluctuation of the COSAC can be significantly smaller, by as much as a factor of 10, than that for a fluorescence detection based conventional clock employing the same transition and same atomic flux. The ultra-narrow resonances produced in this process may also open up the possibility of exploring
novel ways of implementing spin-squeezing techniques for further improvement in clock
stability [56, 7, 9, 57].

3.2. Clock

The discussion can be generalized to $N$ atoms that are all excited by the same field. We assume that there are no overlaps between the wavefunctions of the atoms and there is no interaction among them [1]. The evolution of each atom under these assumptions can be described individually, and the total quantum state is simply the outer (tensor) product of individual quantum states [44, 45]. However, the interaction can also be described equivalently using a basis of collective states [44, 1]. The Hilbert space of $N$ two level atoms is spanned by $2^N$ states. Thus, when transformed to the collective state basis, there are also $2^N$ collective states. For identical Rabi frequencies and resonant frequencies, however, only the generalized symmetric states [44], of which there are only $(N+1)$, are relevant, and the rest of the $(2^N - N - 1)$ states become decoupled. The case where inhomogeneity of the Rabi frequencies and different Doppler shifts experienced by different atoms are taken into account is presented at the end of this section. We also note that if different atoms see different phase factors from the excitation fields, these factors can be absorbed into the definition of the generalized symmetric states [44].

The simplified symmetric states, known as the conventional Dicke states [1], represent the case where it is assumed that the mean separation between the atoms is much less than the wavelength corresponding to the two level transition (which, for the co-propagating off resonant Raman excitation, is $\sim (k_1 - k_2)^{-1}$). While this constraint is not necessary for the concept proposed here [44], it is easier to describe the process initially under this
constraint. The observables computed remain correct when this constraint is not met. Some of these Dicke states are as follows: $|E_0\rangle \equiv |111...1\rangle$, $|E_1\rangle \equiv \sum_{i=1}^{N} |11...2_i...1\rangle/\sqrt{N}$, $|E_2\rangle \equiv \sum_{i,j\neq i}^{N} |11...2_i...2_j...1\rangle/\sqrt{N}C_2$, $|E_3\rangle \equiv \sum_{i,j,k}^{N} |11...2_i...2_j...2_k...1\rangle/\sqrt{N}C_3$, and $|E_N\rangle \equiv |222...2\rangle$ where $N!/n!(N - n)!$. For instance, $|E_2\rangle$ is the Dicke state with two atoms in $|2\rangle$ and the rest in $|1\rangle$. Any two atoms can be in $|2\rangle$ with equal probability, with $N!C_2 = N(N-1)/2$ such possible combinations.

The Hamiltonian in the basis of the symmetric collective states is $H = \sum_{k=0}^{N} [-k\hbar\delta |E_k\rangle\langle E_k|] + \sum_{k=0}^{N-1} [(h\Omega_{k+1}|E_k\rangle\langle E_{k+1}| + H.c.]$ where $\Omega_{k+1} = \sqrt{N-k}\sqrt{k+1}\Omega$ is the Rabi frequency between collective states [44, 1]. The states are separated by $\hbar\delta$ in energy and couple at different rates. For instance, $\Omega_1 = \Omega_N = \sqrt{N}\Omega$, $\Omega_2 = \Omega_{N-1} = \sqrt{2(N-1)}\Omega$, etc. The middle states have the strongest coupling rate of $\Omega_{N/2} = N\Omega$ and the end states couple most weakly.

The final state of the system at the end of the second $\pi/2$ pulse can be derived by using either the collective state picture or, equivalently, the single atom picture. For a large value of $N$, carrying out the calculation in the collective states basis is numerically cumbersome and analytically intractable. However, we can find the state trivially by using the single atom picture and then determining the coefficients of the collective states.
by simple projection, given the definition of the \((N + 1)\) generalized symmetric collective states. As such, the final state of the system is \(|\psi\rangle = \prod_{i=1}^{N}(W_{\pi/2}^0 W_0^{\delta T_2} W_{\pi/2}^0)|1\rangle_i\). In the basis of the generalized symmetric collective states, this becomes:

\[
|\psi\rangle = \left(-ie^{i\theta}\right)^N \sum_{k=0}^{N} \sqrt{N} C_k (\sin \theta)^{N-k} (\cos \theta)^k |\tilde{E}_k\rangle
\]

The population of the state \(|\tilde{E}_N\rangle\) at the end of the separated field experiment is

\[
P_C^N \equiv |\langle \tilde{E}_N |\psi\rangle|^2 = (\cos \theta)^{2N}
\]

which is simply \((P_2)^N\). This quantity, \(P_C^N\), represents the probability of finding the whole system in the state \(|E_N\rangle\) whereas \(P_2\) represents the probability of finding each atom in state \(|2\rangle\). In a conventional experiment, the population of atoms in state \(|2\rangle\) is measured, for example, by collecting fluorescence produced by coupling \(|2\rangle\) to an auxiliary state. The resulting signal is proportional to \(P_2\), independent of the number of atoms.

The experiment that we propose, to be described shortly, produces a signal that is proportional to \(P_C^N\). When Eq. (3.2) is plotted for various values of \(N\) (Fig. 3.1 (b)), it is evident that the linewidth of the fringe as a function of \(\theta\) decreases as \(N\) increases. The value of the linewidth, defined as the full width half maximum (FWHM), is given by \(\Gamma(N) = 2 \arccos (2^{-1/2N})\). The derivative of \(\Gamma(1)/\Gamma(N)\) with respect to \(N\), for large \(N\), approaches the value of 0.8899 + \(O(N^{-3/2})\), which we have verified with a linear fit to \(\Gamma(1)/\Gamma(N)\). To a good approximation, \(\Gamma(N)/\Gamma(1) \approx 1/\sqrt{N}\). Noting that \(\theta = \delta T_2/2\), \(\Gamma(1) \approx \pi/T_2\) is understood to be the transit time limited linewidth. Then \(\Gamma(N) = \Gamma(1)/\sqrt{N} = \pi/(T_2 \sqrt{N})\) is a violation of the transit time limit, which is discussed
in Section 3.6 along with the physical interpretation of what occurs in the collective atomic clock system.

3.3. Simulations

3.3.1. Effect of velocity distribution

A two level atomic system $|\psi\rangle$ interacts with light fields and evolves as $|\psi(t' + t)\rangle = W_{\Omega t}^{\delta t}|\psi(t')\rangle$. The two levels in the proposed scheme are, for example, the hyperfine ground states of an alkali atom such as $^{85}\text{Rb}$. After the $\pi/2$-dark-$\pi/2$ sequence, the system is in state $|\psi\rangle = W_{\pi/2}^{\delta T_2}W_0^{\delta T_2}W_{\pi/2}^{\delta T_2}|1\rangle$. Unlike in Section 1.1.2, we here do not make the approximation that $\delta \ll \Omega$. Then the signal we expect to see for a single atom is proportional to $P_2 = |\langle 2|\psi\rangle|^2 = |\langle 2|W_{\pi/2}^{\delta T_2}W_0^{\delta T_2}W_{\pi/2}^{\delta T_2}|1\rangle|^2$, and the collective state signal is

$$S_{col} = \prod_{i=1}^{N} |\langle 2|W_{\pi/2}^{\delta T_2}W_0^{\delta T_2}W_{\pi/2}^{\delta T_2}|1\rangle|^2$$

(3.3)

We assume that the density of atoms in the trap is fixed at $\rho_A = 10^9$ mm$^{-3}$, so that the width of the atomic ensemble, which has a Gaussian spatial distribution, varies with the number of atoms. With $N = 2 \times 10^6$ atoms in the trap, the size of the cigar-shaped ensemble is 1 mm in length in the direction of the Raman beams, and $\sim 50$ $\mu$m in diameter in the other two directions.

When an atom with velocity $v$ interacts with a field with frequency $\omega$ propagating in the direction of the atom, the frequency of the field is shifted by $\delta_D = v\omega/c$. The Maxwell Boltzmann velocity distribution is $\rho_{MB}(v, T) = \sqrt{m_a/(2\pi kT)} exp^{-m_a v^2/(2kT)}$ where $m_a$ is the atomic mass and $T$ is the temperature. We assume the temperature to be given by
Figure 3.2. Collective state signal at the end of the Ramsey field experiment for various number of atoms, with parameters $\Omega = 5 \cdot 10^5 \text{ s}^{-1}$ and $T_2 = 3 \cdot 10^{-5} \text{ s}$. Plotted are the ideal signal (dashed line), $S_{\text{col}}$, and the reduced signal (solid line), $S_{\text{Dop}}$, where the effect of Doppler shift is taken into account.

the Doppler cooling limit, so that $T_{\text{MOT}} = \Gamma_{\text{Rb}} \hbar/(2k) = 138 \mu \text{K}$ for $^{87}\text{Rb}$. The average velocity is then $v_{av} \sim 18.3 \text{ cm/s}$, with a corresponding Doppler shift of $\delta_{D_{av}} = 4.18 \text{ Hz}$. Under these conditions, the signal is

$$S_{\text{Dop}} = \Pi_{v'=-5v_{av}}^{5v_{av}} \left| \langle 2 | W_{\pi/2}^\dagger(\delta+\delta_D(v'))T_2 W(\delta+\delta_D(v'))T_2 W_{\pi/2}^\dagger(\delta+\delta_D(v'))T_2 |1 \rangle \right| \rho_{\text{MB}}(v',T_{\text{MOT}}) \right]$$

where we take into account velocities that are up to five times the $v_{av}$. Plotted in Fig. 3.2 are the signals $S_{\text{col}}$ and $S_{\text{Dop}}$ for various $N$ values, with $T_2 = 3 \cdot 10^{-5} \text{ s}$ and $\Omega = 5 \cdot 10^6 \text{ s}^{-1}$. The Doppler effect decreases the overall signal while having virtually no effect on its width. It decreases exponentially as $N$ increases. However, for the given choice of temperature and $N = 2 \cdot 10^6$, the reduced signal is $S_{\text{Dop}} \sim 0.9S_{\text{col}}$. Of course, the signal can be improved if the temperature is reduced below the Doppler cooling limit.
3.3.2. Effect of field inhomogeneity

Consider next the effect of the inhomogeneity in the laser field amplitude. We assume that the atomic ensemble has a Gaussian spread with a width of $\omega_A$: $\rho_N(\gamma) = \rho_0 e^{-\left(\gamma^2/\omega_A^2\right)}$. The width considered in this section is in the direction perpendicular to the propagation direction of the Raman beams, since the atoms spread in the propagation direction of the beams see the same fields. Each of the two laser fields that produce the Raman-Rabi excitation is also assumed to have a Gaussian profile with a width of $\omega_L > \omega_A$. Since the Raman-Rabi frequency is proportional to the product of the Rabi frequencies for each of these lasers, it follows that the Raman-Rabi frequency is also a Gaussian with a width of $\omega_L$: $\Omega(\gamma) = \Omega_0 e^{-\left(\gamma^2/\omega_L^2\right)}$. The peak value of $\Omega$ (i.e., $\Omega_0$) is chosen so that the atoms at the center ($r = 0$) experience a perfect $\pi/2$-pulse for an interaction time of $T_1$. Ignoring the
effect of the Doppler spread in the velocity, the COSAC signal is then given by

\[ S_\Omega = \Pi_{\tau=-w_{\rho}}^{w_{\rho}} | \langle 2 | W_{\Omega_1(r)T_1} W_{\Omega_2(r)T_2} | 1 \rangle |^2 \rho_N(r) \]

The signals for various ratios of \( w_L/w_A \) are plotted in Fig. 3.3 for \( N = 2 \cdot 10^6 \) and density of \( \rho_A = 10^9 \text{ mm}^{-3} \). The signal affected by the inhomogeneous fields can reach the peak value of the ideal signal when \( \omega_L/\omega_A = 50 \). Since \( w_A = 50 \mu\text{m} \) in our system, \( w_L = 2.5 \) mm for the Raman beams is sufficiently large enough to achieve this goal.

### 3.3.3. Effect of spontaneous emission

In the analysis of the COSAC, we have used a model in which the intermediate state is adiabatically eliminated. However, the actual population of this state is approximately \( \Omega_1^2/\Delta^2 \) with \( \Omega_1 \sim \Omega_2 \). In the time that it takes for a \( \pi = \Omega/T_1 \simeq \Omega_1^2/(2\Delta T_1) \) pulse (or two \( \pi/2 \) pulses) to occur, we can estimate that the number of spontaneous emissions that occur per atom is \( (\Omega_1^2/\Delta^2) \Gamma T_1 \simeq 2\pi \Gamma/\Delta \). For \( \Delta = 200 \Gamma \), this, number is about \( 3 \times 10^{-2} \), and increases by a factor of \( N \) for an ensemble of \( N \) atoms. (Note that there is no enhancement of the rate of spontaneous emission due to superradiant effects, since we are considering a dilute ensemble). As a result, the signal for both the CC and the COSAC would deviate from the ideal one. The actual effect of spontaneous emission on the CC can be taken into account by using the density matrix equation for a three level system. However, in this case, it is not possible to ascribe a well defined quantum state for each atom. This, in turn, makes it impossible to figure out the response of the COSAC, since our analysis for the COSAC is based on using the direct product of the quantum state of each atom. For a large value of \( N \), it is virtually impossible to develop
a manageable density matrix description of the system directly in terms of the collective states. However, it should be possible to evaluate the results of such a density matrix based model for a small value of \( N \) (< 10, for example). In the near future, we will carry out such a calculation and report the findings.

For the general case of large \( N \), one must rely on an experiment (which, in this context, can be viewed as an analog computer for simulating this problem) to determine the degree of degradation expected from residual spontaneous emission. It should be noted that the deleterious effect of spontaneous emission, for both the CC and the COSAC, can be suppressed to a large degree by simply increasing the optical detuning while also increasing the laser power. This is the approach used, for example, in reducing the effect of radiation loss of atoms in a far off resonant trap (FORT).

### 3.3.4. Effect of fluctuation in number of atoms

For both the CC and the COSAC, the signal is collected multiple times and averaged to increase the signal to noise ratio (SNR). However, the number of atoms can fluctuate from shot to shot. When \( N \) fluctuates by \( \Delta N \), the signal in the CC changes by the same amount while the linewidth does not change. It is easy to see this from the classical signal, \( S_{CC} = N \cos^2 \theta \). Changing \( N \) by \( \Delta N \) will change the signal, but the FWHM, which occurs at \( S_{CC} = N/2 \), will not change. A more thorough approach for expressing the classical and quantum noise of the CC and the COSAC is covered in Section 3.5-A. In this section, we focus on how the fluctuation in the number of atoms from shot to shot affects the signal of the COSAC.
Figure 3.4. (left) Collective state signal (solid line) at the end of the Ramsey field experiment for $N = 2 \cdot 10^6$ atoms; $\Omega = 5 \cdot 10^6 \text{ s}^{-1}$; $T = 3 \cdot 10^{-5}$ s. The dashed curves show the signal for $N + \Delta N$ (narrower) and $N - \Delta N$ (wider), where $\Delta N/N = 0.01$. (right) Plot of $\Delta \Gamma/\Gamma$ as a function of $\Delta N/N$.

Fig. 3.4 (left) is the plot of a collective signal with $N = 2 \cdot 10^6$. The dashed red lines represent the case in which $\Delta N/N = 0.01$. Increasing the number of atoms by $\Delta N$ decreases the linewidth, and decreasing the number of atoms by $\Delta N$ increases the linewidth. However, the peak of the signal remains at unity. This is in contrast to the results from velocity distribution and field inhomogeneity. We calculate the change in the COSAC linewidth by noting that its FWHM is approximately $\Gamma(N) = \Gamma(1)/\sqrt{N}$. The width of the uncertainty in $\Gamma(N)$, as a result of fluctuation in $N$, is $\Delta \Gamma(N) = \Gamma(1)/\sqrt{N} - \Delta N - \Gamma(1)/\sqrt{N} + \Delta N$, so that the fractional fluctuation is $\Delta \Gamma(N)/\Gamma(N) = (1 - \Delta N/N)^{-1/2} - (1 + \Delta N/N)^{-1/2} = \Delta N/N + 0.625(\Delta N/N)^3 + O[(\Delta N/N)^7]$. For small $\Delta N/N$, the fractional change in FWHM is $\Delta \Gamma(N)/\Gamma(N) \simeq \Delta N/N$ to a good approximation. Fig. 3.4 (right) shows this correspondence for $N = 2 \cdot 10^6$. However, the plot is equivalent for any $N$, since the fractional change in FWHM is only dependent on $\Delta N/N$. 
3.4. Experiment and detection scheme for realizing a COSAC

Before proceeding further, we describe the experimental approach that can be used to measure $P_N^C$, as summarized in Fig. 3.5. For concreteness, and without loss of generality, we consider $^{87}\text{Rb}$ as the atomic species. By making use of the necessary D2 line transitions, we start by trapping atoms in a magneto-optical trap (MOT), and transferring them into a more localized dipole trap, cooled down to the Doppler cooling limit of $T_D = h\Gamma/(2k_B) = 138 \mu K$ [58, 59, 60, 61]. After capturing about $2 \cdot 10^6$ atoms in a cigar shaped cloud with a diameter of $\sim w_A = 50 \mu m$ and length of 1 mm, the atoms are released and optically pumped into the $|F = 1\rangle$ state by applying a beam that is resonant with $5^2S_{1/2}, |F =
Figure 3.6. Initialization of the system involves first optically pumping the atoms into \(|F = 1\rangle\) state by applying a laser field that is resonant with \(5^2S_{1/2}, |F = 2\rangle \rightarrow 5^2P_{3/2}, |F' = 2\rangle\) transition. Afterwards, as is depicted here, a \(\pi\) polarized beam that is resonant with \(5^2S_{1/2}, |F = 1\rangle \rightarrow 5^2P_{1/2}, |F' = 1\rangle\) transition is applied. Because the \(|F = 1, m_F = 0\rangle \rightarrow |F' = 1, m_{F'} = 0\rangle\) transition is forbidden for the D1 line, the atoms are eventually pumped into \(|F = 1, m_F = 0\rangle\).

\[2\rangle \rightarrow 5^2P_{3/2}, |F' = 2\rangle\) transition of rubidium D2 line. Furthermore, a \(\pi\) polarized beam that is resonant with \(5^2S_{1/2}, |F = 1\rangle \rightarrow 5^2P_{1/2}, |F' = 1\rangle\) transition of rubidium D1 line is applied, as depicted in Fig. 3.6. Because the \(|F = 1, m_F = 0\rangle \rightarrow |F' = 1, m_{F'} = 0\rangle\) transition is forbidden for the D1 line, the atoms will finally be pumped into \(|F = 1, m_F = 0\rangle\) level. It is possible, with the imperfections that are inadvertently present in the system, that there might be some residual atoms left in \(|F = 1, m_F = -1\rangle\) and \(|F = 1, m_F = 1\rangle\). We avoid the detection of these residual atoms by making use of the fact that the Zeeman shifts of levels in \(|F = 1\rangle\) and \(|F = 2\rangle\) are in opposite directions, which will be discussed in more detail after we outline the null measurement scheme. Once the initialization of atoms into \(|F = 1, m_F = 0\rangle\) state is complete, a bias magnetic field of \(\sim 2\) G, generated with a pair of Helmholtz coils, is turned on in the \(\hat{z}\) direction. While the atoms are in free fall, we turn on a pair of co-propagating right circularly polarized \((\sigma_+)\) Raman beams in the \(\hat{z}\) direction. One of these beams is tuned to be \(\sim 3.417\) GHz red detuned from the \(|F = 1\rangle \rightarrow |F' = 1\rangle\) transition (D1 manifold), and the other is tuned to be \(\sim 3.417\) GHz red detuned from the \(|F = 2\rangle \rightarrow |F' = 1\rangle\) transition (D1 manifold).
Figure 3.7. In the detection zone, we probe the population of state $|E_N\rangle$ by applying field $\omega_1$ and detecting Stokes photons produced during the Raman transition. In the bad cavity limit, the atomic system will not reabsorb the photon that has been emitted during the Raman process, such that the transition from $|E_k\rangle$ to $|E_{k+1}\rangle$ will occur, but not vice versa.

The second Raman beam is generated from the first one via an acousto-optic modulator (AOM), for example. The AOM is driven by a highly stable frequency synthesizer (FS), which is tuned close to $\sim 6.835$ GHz corresponding to the frequency difference between the $|F = 1\rangle$ and $|F = 2\rangle$ states in the $5^2S_{1/2}$ manifold.

These beams excite off-resonant Raman transitions between $|F = 1, m_F = m\rangle$ and $|F = 2, m_F = m\rangle$ levels, for $m = 1, 0, -1$. Since the system is initialized in $|F = 1, m_F = 0\rangle$, the $\sigma_+$ Raman transitions through the excited states $|F' = 1, m_{F'} = 1\rangle$ and $|F' = 2, m_{F'} = 1\rangle$ couple the initial state to $|F = 2, m_F = 0\rangle$. Hence, the energy levels $|1\rangle$ and $|2\rangle$ from the previously discussed $\Lambda$ scheme correspond to hyperfine ground states $|F = 1, m_F = 0\rangle$ and $|F = 2, m_F = 0\rangle$, respectively. The resulting four level system, with the two excited states, can be reduced to a two level system in the same manner as the $\Lambda$ system by adiabatically eliminating the excited states together. The resulting two level system has a coupling rate that is the sum of the two Raman Rabi frequencies, one involving the $|F' = 1, m_{F'} = 1\rangle$ state, and the other involving the $|F' = 2, m_{F'} = 1\rangle$ state. The laser power at $\omega_1$ and $\omega_2$ are adjusted to ensure that the light shifts of levels $|1\rangle$ and $|2\rangle$ are matched.
In the first interaction zone, the co-propagating Raman beams interact with the atomic ensemble for a duration of $\Omega T_1 = \pi/2$. After waiting for a time $T_2$, chosen such that $T_2 \gg T_1$, we pulse the Raman beams again, in place, to interact with the atomic ensemble for another duration $\Omega T_1 = \pi/2$. The Raman beams can be pulsed in place as long as the width of the beams is much larger than that of the free-falling, thermally expanding atomic cloud.

After these excitations, we probe the population in one of the collective states, $|E_N\rangle$, where all the individual atoms are in state $|2\rangle$, by a method of zero photon detection. For illustrative purposes, let us consider first a situation where the atomic ensemble is contained in a single mode cavity with mode volume $V$, cavity decay rate $\gamma_c$, and wavevector $k_2 = \omega_2/c$. The cavity is coupled to the atomic transition $|2\rangle \rightarrow |3\rangle$ with coupling rate $g_c = |e\langle r|E/\hbar$, where $|e\langle r|$ is the dipole moment of the atom and the field of the cavity is $E = \sqrt{2\hbar\omega_2/\epsilon_0 V}$. If we then send a probe beam, an off-resonant classical laser pulse with frequency $\omega_1$, the presence of the cavity will allow Raman transitions to occur between the collective states $|E_k\rangle$ and $|E_{k+1}\rangle$ with the coupling rates $\Omega'_{k+1} = \sqrt{N-k}\sqrt{k+1}\Omega'$ where $\Omega' = \Omega g_c/2\Delta$. The schematic of the interaction is shown in Fig. 3.7.

In the bad cavity limit where $\gamma_c \gg \sqrt{N}\Omega'$, the Raman transitions will still occur. However, the atomic system will not reabsorb the photon that has been emitted during the process, such that the transition from $|E_k\rangle$ to $|E_{k+1}\rangle$ will occur, but not vice versa. The electric field of such a photon is $E = \sqrt{2\hbar\omega_2/(\epsilon_0 Ac\tau)}$, where $A$ is the cross sectional area of the atomic ensemble, $c$ is the speed of light, and $\tau$ is the duration of the photon. This limit applies in our case, which has no cavity. In this limit, the stimulated Raman
scattering is an irreversible process that can be modeled as a decay with an effective decay rate that is singular to each $|E_j\rangle$ state. The decay rate from state $|E_1\rangle$ is $\gamma_0 = 4NL|g_c\Omega_1|^2/(\Delta^2c) = N\gamma_{sa}$ where $\gamma_{sa} = 16L\Omega^2/c$ [62] is the decay rate for a single atom. The value of $g_c$ is given by $|\langle e(r)\rangle| \cdot E$. The effective decay rates for the other states can be calculated following the same logic as $\gamma_j = (j + 1)(N - j)\gamma_{sa}$.

When photons are scattered through stimulated Raman scattering in the detection process, the resonant optical density (OD) determines the degree to which the emission occurs in the direction of propagation of the probe beam [62]. Specifically, the fraction of photons that are not emitted in the direction of the probe is give by $1/OD$. Thus, $(1 - 1/OD)$ determines the effective collection efficiency of the detection process. The OD depends on the density of atoms $n$, the diameter of the atomic ensemble $w_A$, and the resonant scattering cross section $\sigma \simeq (\lambda/2)^2$, as $\rho = \sigma nL$. For the rubidium-87 D1 line wavelength, $\lambda \sim 795$ nm, and a cigar shaped trap with $N = 2 \cdot 10^6$ atoms, a diameter of 50 $\mu$m, and a length of 1 mm, we find that the resonant optical density is $\rho \sim 300$. The beam consisting of the probe and the emitted photons is sent to a high speed detector, which produces a dc voltage as well as a signal at the beat frequency of $\sim 6.835$ GHz. The phase of this beat frequency signal is unknown. As such, the total signal is sent in two different paths, one to be multiplied by the FS signal and another to be multiplied by the FS signal shifted in phase by 90 degrees. Each of these signals is squared, then combined and sent through a low pass filter (LPF) to extract the dc voltage that is proportional to the number of scattered photons. A voltage reading above a predetermined threshold value will indicate the presence of emitted photons during the interrogation period. The interrogation period is set to $\gamma_0 T = 10$ where $\gamma_0 = \gamma_{N-1} = N\gamma_{sa}$ is the slowest decay rate,
to ensure that even the longest lived state has a chance to decay almost completely. If no photon emission occurs and the voltage reads below the threshold, this indicates that the atoms are all in $|2\rangle$ and the collective state of the system is $|E_N\rangle$. For any other collective state, at least one photon will be emitted. For a given value of $\delta$, this process is repeated $m$ times (where the choice of $m$ would depend on the temporal granularity of interest). The fraction of events corresponding to detection of no photons would represent the signal for this value of $\delta$. The process is now repeated for a different value of $\delta$, thus enabling one to produce the clock signal as a function of $\delta$. Usual techniques of modulating the detuning and demodulating the signal can be used to produce the error signal for stabilizing the FS, thus realizing the COSAC.

With an off-resonant beam of electric field $E_1 = E_{10}e^{i\omega_1 t}$ (read beam) that induces a Raman transition from one hyperfine ground state to another, the photon that is produced has a field of the form $E_2 = E_{20}e^{i(\omega_2 t + \phi)}$ with a single photon Rabi frequency. The photon emitted with stimulated emission, in free space with high optical density, both fields travel in the same direction, in the propagation direction of the read beam. Hence, placing a detector along this direction ensures capture of the emitted photon. The signal observed by this detector is

$$S = I = |E_1 + E_2|^2$$

$$= A + \frac{B}{2} [e^{i(\Delta\omega t - \phi)} + e^{-i(\Delta\omega t - \phi)}]$$

$$= A + B \cos (\Delta\omega t - \phi)$$

(3.6)

where $A = |E_{10}|^2 + |E_{20}|^2$, $B/2 = |E_{10}||E_{20}|$, and $\Delta\omega = \omega_2 - \omega_1 \sim 3.03 \text{GHz}$. 

Meanwhile, the frequency synthesizer or the voltage controlled oscillator that produces the Raman beams has field $E_3 = E_{30} \cos (\Delta \omega t + \psi)$ after it undergoes frequency doubling. We split up this signal, and one path passes through a 90 degree phase shifter, so that the field here is $E_4 = E_{30} \sin (\Delta \omega t + \psi)$.

The signal from the detector is split up, so that a part of it combines (multiplies) with $E_3$, and the other part with $E_4$. For one path, then, the signal is

\[(3.7) \quad S_1 = C [A + B \cos (\Delta \omega t - \phi)] \cos (\Delta \omega t + \psi)\]

and the signal through the other path is

\[(3.8) \quad S_2 = C [A + B \cos (\Delta \omega t - \phi)] \sin (\Delta \omega t + \psi)\]

where the overall factor of $1/2$ is ignored, and $C = E_{30}$.

Each path is split up into two, and recombined with itself, so that the signals become:

\[(3.9) \quad S'_1 = C^2 [A + B \cos (\Delta \omega t - \phi)]^2 \cos^2 (\Delta \omega t + \psi)\]

and

\[(3.10) \quad S'_2 = C^2 [A + B \cos (\Delta \omega t - \phi)]^2 \sin^2 (\Delta \omega t + \psi)\]

When the two signals are added together, the resulting signal is:

\[(3.11) \quad S'_{1+2} = C^2 \left( A^2 + B^2 \cos^2 (\Delta \omega t - \phi) + 2AB \cos (\Delta \omega t - \phi) \right)\]
As can be seen, the dependence on the arbitrary phase from the voltage controlled oscillator, $\psi$, is eliminated by this process (the squaring of each signal is necessary for this to occur). We can place a low pass filter in front of this signal, so that only the DC portion of it passes through. Using the fact that the time average of $\cos^2(\theta t) = 1/2$, the final signal becomes:

$$S_F = C^2 \left( A^2 + \frac{B^2}{2} \right)$$

(3.12)

$$= |E_{30}|^2 \left[ (|E_{10}|^2 + |E_{20}|^2)^2 + \left( \frac{|E_{10}| |E_{20}|}{2} \right)^2 \right]$$

From this information, we can try to deduce the threshold above which a photon emitted from the Raman process has been detected. For simplicity, we substitute $a \equiv |E_{10}|^2$, $b \equiv |E_{20}|^2$, and $c \equiv |E_{30}|^2$ so that:

$$\frac{S_F}{c} = a^2 + b^2 + 4ac \equiv d$$

(3.13)

Solving for the signal that we want, $b = -2a + \sqrt{3a^2 + d}$. The signal is positive only when $d > a^2$, but this is always true, as can be seen from Eq. (3.13). Therefore, by monitoring the initial signal amplitude $a$, the signal amplitude from the voltage controlled oscillator, $c$, and the detected signal, $S_F$, we can obtain the desired detection of $b$.

As noted earlier, it is possible that a small fraction of the detected signal might be due to the residual atoms that were not optically pumped to $|F = 1, m_F = 0\rangle$ initially. The $\sigma_+$ polarized Raman probe is applied to $|F = 1\rangle$ level, and the residual atoms in $|F = 1, m_F = -1\rangle$ and $|F = 1, m_F = 1\rangle$ can also see the excitation. However, the bias magnetic field of 2 Gauss lifts the degeneracy of the energy levels. Moreover, since $g_F = -1/2$
for $|F = 1\rangle$ and $g_F = 1/2$ for $|F = 2\rangle$, the energy levels shift in opposite directions such that the Raman signals for the transitions involving $m_F = -1$ and $m_F = 1$ are detuned from resonance. Each will be shifted by $\delta_z = -m(g_{F=2} - g_{F=1})\mu_B B/\hbar = -1.4$ [MHz/Gauss] · $m_F B$ where $B = 2$ Gauss. Therefore, these transitions will not be a part of the detection, which only involves looking at 6.835 GHz beat frequency between the probe and the spontaneously generated photon.

In the particular implementation of the COSAC considered here, we have used off-resonant Raman transition. However, effects such as residual light shifts can limit the stability of such a clock. The ground states can also be coupled directly by using a microwave pulse, which has the advantage of being free from differential light shifts. Thus, the COSAC can also be realized by using a traveling wave microwave pulse sequence for the separated Ramsey field experiment [63], as long as the detection pulse remains the same. Since the Hamiltonian for light-shift balanced off-resonant Raman excitation, with the excited state eliminated adiabatically, is formally identical to that of microwave excitation [20], the basic behavior of the COSAC would be identical for a microwave version.

### 3.5. Performance of the COSAC compared to that of the CC

In order to compare the performance of the COSAC to that of the comparable CC, we examine the stability of the clocks by investigating the fluctuation that has both quantum mechanical and classical components, or $\delta f|_{\text{total}} = (\Delta S_{QM} + \Delta S_{\text{class}})/(\partial S/\partial f)$, where $S(f)$ is the signal and $f$ is the detuning of the clock away from its center value. Because the signal depends on the frequency, the fluctuations in a clock are not necessarily constant,
and there is not a single value of the SNR to compare unless we compare the two clocks at a particular value of the frequency. Instead, the fluctuations must be compared as a function of $f$ for completeness. In this section, we discuss the quantum fluctuation due to quantum projection noise, $\Delta P = \sqrt{P(1 - P)}$ [56], where $P$ is the population of the state to be measured, the classical noise in the long term regime, and the effects of detector efficiency and the collection efficiency. The ratios of the frequency fluctuations in the CC to the frequency fluctuations in the COSAC show that the two clocks perform comparably around the signal at $f = 0$ if the clocks have perfect collection efficiency. However, the traditional fluorescence detection based clock suffers from collection efficiency issues that the collective clock is immune to. For the CC, a resonant beam probes the clock state, generating spontaneously emitted photons. The collection efficiency of such a system is limited by the solid angle of the detection system. On the other hand, the COSAC collects the fluorescence of photons through coherent Raman scattering, which enables large collection efficiency that can be close to unity for sufficiently high resonant optical density (as noted earlier). As such, for the same number of atoms detected per unit time, the COSAC is expected to perform better than the fluorescence detection based CC by as much as a factor of 10. This is discussed in greater detail in subsection C of this section.

### 3.5.1. Effects of quantum and classical noise

In order for the COSAC to be useful, it must perform at least as well as, or better than, the CC, and for that, we must compare the two clocks’ stability in the short term and the long term regimes. The stability of a clock can be measured by investigating the frequency fluctuation that has both quantum mechanical and classical components.
Before comparing the stabilities of the COSAC and the CC, it is instructive first to review briefly the stability of a CC.

For concreteness, we consider an off-resonant Raman-Ramsey clock as the CC. The population of the detected state $|2\rangle$ at the end of the second pulse is given by $P_2 = \cos^2(fT_2/2)$, where $T_2$ is the separation period of the two $\pi/2$-pulses and $f$ is the deviation of the clock frequency away from its ideal value, expressed in radial units (i.e. rad/s rather than Hz). The signal is detected by probing the desired state for a duration of time. If $\tilde{N}$ is the number of atoms per unit time and $\tau$ is the interrogation period, the net signal is $S_{sa} = \tilde{N}\tau P_2 = \tilde{N}\tau \cos^2(fT_2/2)$. For the sake of comparison, we allow the number of atoms per trial in the COSAC signal, $N$, multiplied by the number of trials, $m$, to equal $\tilde{N}\tau$. Therefore, we can write $S_{sa} = mN \cos^2(fT_2/2)$. The quantum mechanical variance of this quantity is $\Delta S_{QM,sa} = \sqrt{mN/2} \sin(fT_2)$, where the derivation is made by noting that the fluctuations in $mN$ is $\sqrt{mN}$ [56], and the projection noise in a single two level atomic system is $\Delta P_2 = \sqrt{P_2(1-P_2)}$ [56]. (It should be noted that the fluctuation in $mN$ is also a manifestation of this projection noise, as discussed in detail in [56].) When the probability of finding the population in this state is unity or nil, the projection noise vanishes; on the other hand, it is largest at $P_2 = 1/2$. Calculating the slope from the signal, we find that $\partial S_{sa}/\partial f = -[mN/(2\gamma_{sa})] \sin(fT_2)$, where $\gamma_{sa} = 1/T_2$ is the linewidth.

Assuming perfect quantum efficiency for the detection process, the frequency fluctuation can be written as $\delta f|_{total} = |(\Delta S_{QM} + \Delta S_{class})/(\partial S/\partial f)|$, which can be regarded as noise ($\Delta S$), both quantum and classical, over the Spectral Variation of Signal ($\partial S/\partial f$), or SVS. In what follows, we consider first the effect of quantum noise only. Thus, the
quantum frequency fluctuation (QFF) for a CC can be expressed as

\[
\partial f_{QM,CC} \equiv \frac{\Delta S_{QM,sa}}{\left(\frac{\partial S_{sa}}{\partial f}\right)} = \frac{\gamma_{sa}}{\sqrt{mN}}
\]  

It should be noted that while both \( \Delta S_{QM} \) and \( (\partial S/\partial f) \) depend on \( f \), their ratio is a constant, which is merely an accident due to the fact that the signal is cosinusoidal. However, this accidental cancellation has led to an apparently simple perception of the QFF as being simply the ratio of the linewidth \( (\gamma_{sa}) \) to the SNR, where the SNR is understood to be \( \sqrt{mN} \). This expression for the SNR, in turn, follows from thinking about the signal as being \( S' = mN \) and noise \( N' \) as being \( \sqrt{mN} \), so that \( \text{SNR} \equiv S'/N' = \sqrt{mN} \). However, it should be clear from the discussion above that the signal is not given by \( mN \), and noise is not given by \( \sqrt{mN} \); rather, they both depend on \( f \).

In cases where frequency fluctuation is not a constant (as will be the case for the COSAC), we can no longer measure the stability of the clock in terms of a constant \( \gamma \)/SNR. Instead, it is necessary to carry out the full calculation of the frequency fluctuation as a function of frequency. Thus, we will adopt the convention that the net frequency fluctuation, \( \delta f \), should be thought of as the ratio of the noise to the SVS. This approach should be adopted universally for all metrological devices. Of course, for devices where the relevant quantity is not the frequency, the definition should be adapted accordingly. For example, in an interferometer that measures phase, the relevant quantity can be expressed as follows: net phase fluctuation is the ratio of the noise to the Angular Variation of Signal (AVS).
Following this convention, we can now examine the net frequency fluctuation of the COSAC and compare it to that of the CC. We will first compare their quantum fluctuations, which is relevant in the short term regime, and then the classical fluctuations, which dominates the long term regime. The collective state signal for \( m \) trials is \( S_{col} = m P^C_N = m \cos^{2N} (fT_2/2) \) and the projection noise is \( \Delta P^C_N = \sqrt{P^C_N (1 - P^C_N)} \) for a single trial and \( \Delta P^C_N = \sqrt{m} \sqrt{P^C_N (1 - P^C_N)} \) for \( m \) trials, so that the total quantum mechanical noise in the signal is

\[
\Delta S_{QM, col} = \sqrt{m} \cos^N (fT_2/2) \sqrt{1 - \cos^{2N} (fT_2/2)}
\]

and the SVS is

\[
\frac{\partial S_{col}}{\partial f} = -(mN/\gamma_{sa}) \sin (fT_2/2) \cos^{2N-1} (fT_2/2)
\]

Therefore, the frequency fluctuation in the COSAC due solely to quantum noise can be expressed as:

\[
\delta f_{QM, COSAC} = \left| \frac{\gamma_{sa}}{N \sqrt{m}} \sqrt{\frac{1 - P^C_N}{P^C_N} \cot \left( \frac{fT_2}{2} \right)} \right|
\]

where \( P^C_N \) is a function of \( f \). Thus, unlike in the case of the CC, the frequency fluctuation is not a constant, and depends strongly on \( f \).

We consider first the limiting case of \( f \to 0 \). Using Taylor expansion, it is easy to see that

\[
\delta f_{QM, COSAC} \simeq \frac{\gamma_{sa}}{\sqrt{mN}}
\]
which is the same as that of the CC, given in Eq. (3.14). This can be understood physically by noting that while the fringe width becomes much narrower for the COSAC, the SNR also decreases due to the fact that a single observation is made for all N atoms in a given trial.

The QFF for the COSAC, given in Eq. (3.17), is smallest as $f \to 0$ and increases as $f$ moves away from resonance. The ratio of the QFF for the CC, given in Eq. (3.14), to that of the COSAC, given in Eq. (3.17), is plotted as a function of $f$ in Fig. 3.8 (left) for $T_2 = 10^{-4}$ s, $m = 1000$ and $N = 2 \cdot 10^6$. Here, the vertical bars indicate the FWHM of the COSAC signal. It is clear from this plot that the QFF for the COSAC increases significantly as we move away from resonance. However, since a servo will keep the value of $f$ confined to be close to zero, the frequency stability of the COSAC, under quantum noise limited operation, should be very close to that of the CC, assuming that all the other factors remain the same.
The classical frequency fluctuation (CFF), \( \delta f_{\text{class}} = \Delta S_{\text{class}} / (\partial S / \partial f) \), is the limiting factor in the long term stability. While the quantum fluctuation is dominated by quantum projection noise, the classical noise is dominated by noise in the electronics employed to generate the clock signal. Since the pieces of equipment used in the development of both the COSAC and CC suffer from similar noise issues, the variance \( \Delta S \) is expected to be of the same order of magnitude for both clocks. On the other hand, the SVS, \( (\partial S / \partial f) \), is not the same, as was shown previously. The ratio of the SVS of the COSAC to the SVS of the CC is

\[
\frac{\partial S_{\text{col}} / \partial f}{\partial S_{\text{sa}} / \partial f} = \cos^2\left(\frac{f T_2}{2}\right) \cos^2\left(\frac{N f T_2}{2}\right) = \frac{P_C}{P_2}
\]

and is plotted in Fig. 3.8 (right). With \( \Delta S_{\text{class, col}} \sim \Delta S_{\text{class, sa}} \), the ratio of the CFF of the COSAC to the CFF of the CC can be written

\[
\frac{\delta f_{\text{class,COSAC}}}{\delta f_{\text{class,CC}}} \simeq \frac{\cos^2\left(\frac{f T_2}{2}\right)}{\cos^2\left(\frac{N f T_2}{2}\right)}
\]

Similar to the ratio of the two clocks in QFF, Eq. (3.20) is smallest as \( f \to 0 \) and increases as \( f \) moves away from resonance. Thus, with respect to both quantum and classical sources of noise, the COSAC must be operated near \( f \simeq 0 \) for optimal performance.

We have investigated the effects of quantum and classical noise by deriving the expression for fluctuation in frequency. However, as was shown in the first section, the signal is also a function of other experimental variables; and in general, the fluctuations in any of these can be expressed as

\[
\partial A \equiv \left| \frac{\Delta S_{\text{QM}}(A) + \Delta S_{\text{class}}(A)}{\partial S(A) / \partial A} \right|
\]
where $A$ is the variable whose fluctuation is of interest, and the signal $S$ is expressed in terms of $A$.

### 3.5.2. Effect of detector efficiency

We recall briefly that in the COSAC detection scheme, a laser with a frequency corresponding to one leg of the Raman transition interacts with the atoms, which are in the quantum state $|\psi\rangle = c_N |E_N\rangle + \sum_{j=0}^{N-1} c_j |E_j\rangle$. Interaction between this field, the atoms, and the free space vacuum modes on the other leg would lead to production of photons unless $c_N = 1$ and $c_j = 0$ for all $j$. These photons are detected using a heterodyning technique, as described previously. The voltage output of the heterodyning system is proportional to the amplitude of the electric field corresponding to the photons.

In general, one or more photons are produced as $|E_j\rangle$ decays to $|E_{j+1}\rangle$ and subsequent states. The time needed for these photons to be produced depends on the vacuum and probe field induced Raman transition rates between $|E_j\rangle$ and $|E_{j+1}\rangle$. If one assumes perfect efficiency for detecting each of these photons, and waits for a time long compared to the inverse of the weakest of these transition rates, then the detection of no photons implies that the system is in state $|E_N\rangle$. In practice, we can choose a small threshold voltage at the output of the heterodyning system as an indicator of null detection. Thus, any signal below this threshold would be viewed as detection of the quantum system in the $|E_N\rangle$ state, and all signals above this threshold would be discarded. The number of events below this threshold for $m$ trials carried out with all the parameters of the experiment unchanged, is the derived signal for the COSAC. After collecting data for all the values of detuning that is of interest, the result would ideally yield the plot of the COSAC signal.
\(S_{\text{col}} = |c_N|^2\), averaged over \(m\) trials. However, with a fractional detector efficiency and finite detection period, the signal would deviate from the ideal result.

Consider first the effect of the detection period. Given the decay rate of the off-resonant Raman process, \(\gamma_j = (j + 1)(N - j)\gamma_{sa}\) as described previously, the probability that \(|E_j\rangle\) will produce zero photons during the measurement period \(\tau\) is \(P_{0,j} = e^{-\gamma_j \tau}\). Thus, the total probability of zero photon emission (which should vanish ideally for any \(c_j \neq 0\)) is given by \(P_0 = \sum_{j=0}^{N-1} |c_j|^2 e^{-\gamma_j \tau}\). The collective state signal, \(S_{\text{col}}\), is the total probability of finding zero photons during \(\tau\), and can be expressed as \(S_{\text{col}} = |c_N|^2 + \sum_{j=0}^{N-1} |c_j|^2 e^{-\gamma_j \tau}\). Noting that \(\gamma_N = 0\), we can rewrite this compactly as \(S_{\text{col}} = \sum_{j=0}^{N} |c_j|^2 e^{-\gamma_j \tau}\). The lower and upper bounds of \(S_{\text{col}}\) can be established by considering the strongest and the weakest effective decay rates. The strongest decay rate occurs for the middle state, \(\gamma_{N/2} = (N/2)(N/2 + 1) \approx (N^2/4)\gamma_{sa}\), where \(N \gg 1\) approximation has been made. With the substitution of the largest decay rate for each \(|E_j\rangle\) into the equation for \(S_{\text{col}}\), the lower bound is set by

\[
(3.22) \quad S_{\text{LB}} = |c_N|^2 + (1 - |c_N|^2) e^{-N^2/4\gamma_{sa}\tau}
\]

Likewise, with the substitution of the weakest decay rate for each \(|E_j\rangle\), \(\gamma_0 = \gamma_{N-1} = N\gamma_{sa}\), into \(S_{\text{col}}\), the upper bound is set by

\[
(3.23) \quad S_{\text{UB}} = |c_N|^2 + (1 - |c_N|^2) e^{-N\gamma_{sa}\tau}
\]

The signal produced in time \(\tau\) will then lie somewhere between the lower and the upper bounds.
Consider next the effect of non-ideal detection efficiency of the heterodyning scheme. To be concrete, let us define as $\eta$ the efficiency of detecting a single photon. In practice, this parameter will depend on a combination of factors, including the quantum efficiency of the high-speed photodetector and the overlap between the probe laser mode and the mode of the emitted photon, as well as the resonant optical depth of the ensemble, as discussed earlier. For the COSAC, it should be noted that we are interested in knowing only whether one or more photons have been detected, and not in the actual number of photons. When more photons are emitted, the detector will have a better chance of observing a non-zero signal, and hence distinguish zero photon emission from the rest with more certainty. For example, if three photons are emitted during the interrogation time, then four different outcomes are possible:

- All three photons are detected, with probability $\eta^3$;
- Two of the photons are detected, with probability $\eta^2(1-\eta)$; this can occur for any two of the photons, so the multiplicity is 3;
- One photon is detected, with probability $\eta(1-\eta)^2$ and multiplicity of 3.
- No photons are detected, with probability $\epsilon^3$ where $\epsilon \equiv 1-\eta$

The sum of these probabilities is 1. The probability that at least 1 photon is detected is thus $(1-\epsilon^3)$. For any state $j \neq N$, the probability of detecting at least 1 photon is therefore $(1-\epsilon^{N-j})$.

Moreover, we must also consider how the effective detection efficiency is influenced by the fact that the collective states decay at different rates. Specifically, the $j$th level for $j < N$ might produce $N-j$ photons, $N-j-1$ photons, down to no photons, depending on the length of the measurement time and the effective decay rate. If the system is in
the state $|E_{N-3}\rangle$, for example, it can produce up to 3 photons but with probabilities that change over the course of the detection period. For a given time $\tau$, $|E_{N-3}\rangle$ evolves into a sum of the states $|E_{N-3}\rangle \rightarrow \sum_{k=N-3}^{N} a_{jk}(\tau)|E_k\rangle$, where the coefficient $a_{jk}(\tau)$ depends on the effective decay rate that is specific to each state, and changes as the states evolve in time. The detector efficiency can be inserted to show the true probability of detecting a non-zero signal, keeping in mind that no photon is produced if the ensemble remains in state $|E_{N-3}\rangle$, 1 photon is produced via evolution of the ensemble to state $|E_{N-2}\rangle$, and so on. Then the probability of at least one photon being produced during a period of $\tau$ is

$$P_{N-3} = \sum_{k=N-3}^{N} \left(1 - \varepsilon^{k-N+3}\right) |\alpha_{jk}(\tau)|^2$$

Thus, the total probability of detecting at least one photon is:

$$P = \sum_{j=0}^{N-1} |c_j|^2 \sum_{k=j}^{N} \left(1 - \varepsilon^{k-j}\right) |\alpha_{jk}(\tau)|^2$$

The probability of seeing no photon is

$$S_{col} = 1 - P = 1 - \sum_{j=0}^{N-1} |c_j|^2 \sum_{k=j}^{N} \left(1 - \varepsilon^{k-j}\right) |\alpha_{jk}(\tau)|^2$$

The numerical analysis for a large number of atoms is tedious and scales as at least $(N - 1)!$ for the COSAC. However, we can take the worst case scenario to serve as the upper bound for the signal. The worst case occurs when only a single photon is produced as a result of $|E_j\rangle$ decaying to only the $|E_{j+1}\rangle$ state, so that the index of the second summation stops at $k = j + 1$. In this case, we can write $|a_{j,j+1}(\tau)| = (1 - e^{-\gamma\tau})$ and the
signal becomes

\begin{equation}
S_{\text{col}} = |c_N|^2 + \varepsilon \left(1 - |c_N|^2\right) + \eta \sum_{j=0}^{N-1} |c_j|^2 e^{-\gamma_j \tau}
\end{equation}

Now, using the approach we employed in arriving at equations Eq. (3.22) and Eq. (3.23), we now consider the strongest and the weakest decay rates for single photon production to arrive at the lower and upper bounds of the zero photon count signal:

\begin{equation}
S_{\text{LB}} = 1 - \eta \left(1 - |c_N|^2\right) \left(1 - e^{-\frac{N^2}{4} \gamma_{sa} \tau}\right)
\end{equation}

\begin{equation}
S_{\text{UB}} = 1 - \eta \left(1 - |c_N|^2\right) \left(1 - e^{-N \gamma_{sa} \tau}\right)
\end{equation}

Plots in Fig. 3.9 are of the ideal signal (under infinite detection time and \( \eta = 1 \)), the lower bound, and the upper bound for various values of \( \tau \) and \( \eta \) for \( N = 2 \cdot 10^6 \), \( T_2 = 3 \cdot 10^{-5} \) s, and \( \gamma_{sa} = 10^4 \) s\(^{-1} \). As can be seen, the detector efficiency and measurement time do
not affect the peak value of the amplitude. As the signal trails off for non-zero detuning, however, the difference increases. The decrease in $\eta$ affects both $S_{UB}$ and $S_{LB}$ similarly, whereas the effect of the decrease in $\tau$ is more evident in $S_{UB}$. With the given parameters, the interrogation period of $\tau = 10^{-4}$ s and detector efficiency of $\eta = 0.99$ yields almost ideal signal. A somewhat lower value of $\eta$ (e.g. 0.70) still yields a signal that is nearly ideal near zero detuning, which is the desired operating regime for the COSAC, as pointed out earlier.

If we set $\gamma_{sa}\tau = 1$, the signal depends on $\eta$ as

\begin{equation}
S_{col} \simeq 1 - \eta \left[ 1 - \cos^{2N} (fT_2/2) \right]
\end{equation}

for large $N$ and $m = 1$. Hence, we can calculate the QFF for the COSAC to see how it depends on the detector efficiency, and how it compares to the CC. For the CC, it is straightforward to show that with $S_{sa} = \eta N \cos^2 (fT_2/2)$, the quantum mechanical noise in the signal is $\Delta S_{sa} = \sqrt{\eta N} \cos (fT_2/2) \sin (fT_2/2)$ and the SVS is $|\partial S_{sa}/\partial \delta| = (\eta N/\gamma_{sa}) \cos (fT_2/2) \sin (fT_2/2)$, so that the QFF is $\delta f_{Q,CC} = \gamma_{sa}/\sqrt{\eta N}$. It is also straightforward to calculate the QFF for the COSAC. The total quantum mechanical noise in the COSAC signal in Eq. (3.30) is:

\begin{equation}
\Delta S_{Q,col} = \sqrt{\eta} \cos^N (fT_2/2) \sqrt{1 - \cos^{2N} (fT_2/2)}
\end{equation}

and the SVS is

\begin{equation}
\partial S_{col}/\partial f = - (\eta N/\gamma_{sa}) \sin (fT_2/2) \cos^{2N-1} (fT_2/2)
\end{equation}
Thus, the QFF in the COSAC is:

\[
\delta f_{QM,COSAC} = \left| \frac{\gamma_{sa}}{\sqrt{N} \sqrt{\eta}} \sqrt{1 - \frac{P_C}{P_N}} \cot \left( \frac{fT_2}{2} \right) \right|
\]

which approaches \( \gamma_{sa}/\sqrt{\eta N} \) as \( f \to 0 \). Assuming that the detector efficiencies of the COSAC and the CC can be essentially the same, they do not affect the ratio of the two QFFs.

### 3.5.3. Effect of collection efficiency

We consider next the effect of the collection efficiency, \( \beta \). The signal, for both the COSAC and CC, is directly proportional to \( \beta \). Thus, it is easy to see, using Eqs. (3.14) and (3.17), that

\[
\zeta \equiv \frac{\delta f_{QM,COSAC}}{\delta f_{QM,CC}} = \left[ \frac{1}{\sqrt{N}} \sqrt{1 - \frac{P_C}{P_N}} \cot \left( \frac{fT_2}{2} \right) \right] \sqrt{\frac{\beta_{CC}}{\beta_{COSAC}}}
\]

where \( \beta_{CC} (\beta_{COSAC}) \) is the collection efficiency of the CC (COSAC).

As noted above, the quantity written in the square bracket in Eq. (3.34) approaches unity as \( f \to 0 \). Thus, in this limit, we see that the ratio of the QFF for the COSAC to that of the CC would depend on the ratio of the collection efficiencies of the detection processes. As discussed previously, for a high enough resonant optical density (10\(^3\) in the example we are considering) the coherent stimulated Raman scattering based detection method used for the COSAC process has a collection efficiency that is close to unity, or \( \beta_{COSAC} \approx 1 \). As for the CC, the fluorescence is typically collected from the spontaneous
emission process, which emits photons in a dipolar radiation pattern. We can estimate typical values of $\beta_{CC}$ by considering, for example, a CC that makes use of cold atoms released from a MOT. For a lens placed at a distance of 5 cm, with a diameter of 2.5 cm, ignoring the dipolar pattern of radiation for simplicity, and assuming it to be uniform in all directions, this system yields a value of $\beta_{CC} \approx r^2/(4d^2) = 1/16$ corresponding to $\zeta \sim 0.25$. In a typical CC, various geometric constraints make it difficult to achieve a value of $\beta_{CC}$ much larger than this. In fact, in cases where the total volume occupied by the CC has to be constrained in order to meet the user requirements, the value of $\beta_{CC}$ is typically 1%, which would correspond to $\zeta \sim 0.1$. Thus, the near unity collection efficiency of the COSAC can lead to an improvement of the clock stability by as much as a factor of 10, compared to a typical CC that makes use of fluorescence detection.

Absorption is another way of detecting the signal in a CC. However, many practical issues must be taken into account if absorption is to be used. First, the fluctuation in the clock frequency is affected by additional noise contributed by the laser used in absorption. Let us assume that the observation time window is $\tau$, and the number of photons in the probe beam, before absorption, is $N_P$, and the probe is in a Coherent state. We also assume that the number of atoms passing through the detection process in this time window is $N_A$, and the linewidth of the resonance is $\Gamma$. If the detection process produces an absorption by a fraction of $\alpha$ (i.e., $\alpha = 1$ represents perfect absorption of the laser beam), and the detector has a quantum efficiency of $\eta$, then the resulting fluctuation in the clock frequency can be expressed as:

$$\delta \omega_{ABS} = \Gamma \left( \frac{1}{\sqrt{\eta \alpha N_A}} + \frac{1}{\sqrt{\eta \alpha N_P}} \right)$$
Here, the first term inside the parenthesis represents the quantum projection noise of the atoms, and the second term represents the shot noise of the photons (which can be thought of as the quantum projection noise of photons). The validity of this expression can be easily established by considering various limits. Consider first the ideal case where $\xi \equiv \eta \alpha = 1$. For $N_P \gg N_A$, the additional noise from the laser can be neglected, and we get the fundamental noise limit due to the quantum projection noise of atoms. On the other hand, if $N_A \gg N_P$, the quantum projection noise from the atoms can be neglected, and the process is limited by the shot-noise of the laser. In general, the parameter $\xi$ represents the overall quantum efficiency of the detection process. The corresponding expression for detection via fluorescence is $\delta \omega_{FLU} = \Gamma(\eta \rho N_A)^{-1/2}$, where again $\eta$ is the quantum efficiency of the detector, and $\rho$ is the fraction of fluorescence falling on the detector.

The contribution from the second term in Eq. (3.35) shows that the intensity of the laser beam used in absorption must be made strong enough in order to make the effect of this term negligible compared to the first term. However, since the absorption process is nonlinear and saturates for a strong laser beam, increasing the laser intensity often decreases the effective value of $\alpha$. For example, consider an ensemble of $2 \cdot 10^6$ atoms with a linear resonant optical density of 300, which can be realized (as we have shown above) for an ensemble confined to a cigar shaped ensemble with a diameter $\sim 50 \, \mu \text{m}$. For a weak probe, the value of $\alpha$ is unity. However, as the probe power is increased, the value of $\alpha$ decreases dramatically. This can be seen by considering a situation where the value of $N_P$ is $10^9$, for example. Since the atomic transition used for absorption is not closed (i.e., not cyclic), the ensemble can only absorb a number of photons that is of the
order of $2 \cdot 10^6$. Thus, the maximum value of $\alpha$ would be only about 0.002. Furthermore, if the area of the laser beam ($A_L$) is much larger than the area of the atomic ensemble ($A_A$), then the value of $\alpha$ can never exceed the value of $A_A/A_L$. We are not aware of any publication reporting a cold atom clock that makes use of absorption for detecting the atoms, possibly because of these constraints and considerations. Nonetheless, as a matter of principle, an absorption process can certainly be used to reduce the quantum frequency fluctuation below what is observed in fluorescence detection systems, under proper choice of parameters.

3.6. Physical Interpretation of Linewidth Reduction and Its Relevance to the Transit Time Limit

As we have shown, the fact that the linewidth in a COSAC is narrower by a factor of $\sqrt{N}$ can be proven mathematically. However, it is instructive to discuss the physical mechanism that leads to this narrowing. Furthermore, it is also important to address the issue of why the violation of the conventional notion of the transit time limit does not contradict the fundamental laws of quantum mechanics.

3.6.1. Physical interpretation of line narrowing

We consider a simple picture of an oscillator and a probe in order to understand the physical explanation as to why the linewidth of a COSAC narrows by $\sqrt{N}$. A clock is essentially an oscillator oscillating at some frequency $\omega$. In order to ascertain that the oscillator has not drifted, the oscillator frequency is mapped into light and interacts with a two level atom, with the ground state $|1\rangle$ and the excited state $|2\rangle$, and a transition
frequency \( \omega_0 \). If \( \omega \) does not match \( \omega_0 \), an error signal proportional to \( \delta = \omega - \omega_0 \) is produced to correct for the difference. Now consider for a moment that we can create a two state superposition of \( N \) atoms such that they are all either in the ground state or the excited state. In other words, \( |\psi\rangle = C_0 |E_0\rangle + C_N |E_N\rangle \) where \( |E_0\rangle = |111...11\rangle \) and \( |E_N\rangle = |222...22\rangle \). The energy difference between these two states is \( N\omega_0 \). The oscillator frequency is still \( \omega \), but when a light field with \( N \) photons is compared with such a two level system, the difference in energy is \( N\delta = N\omega - N\omega_0 \). If it were possible to produce an error signal that is proportional to this energy difference without degrading the effective signal to noise ratio (or, more accurately, the ratio of noise to the SVS, as discussed in Section V-A), the resulting clock would be \( N \)-fold more accurate. This is functionally equivalent to the clock transition frequency being enhanced by a factor of \( N \).

However, this clean two level superposition of collective states is virtually impossible to achieve with a collection of \( N \) non-interacting atoms and a single field since there is no electric dipole moment to excite the \( |E_N\rangle \) state directly from the \( |E_0\rangle \) state. What occurs instead is that all the states between these get excited as well, as illustrated in Fig. 3.10. If we consider only the excitations from state \( |E_0\rangle \), there are \( N \) possible transitions that can occur, so that the error signal includes the set of all the possible detunings, \( \delta, 2\delta, 3\delta, ... N\delta \). In other words, there are effectively \( N \) different sensors running at the same time. All the other states also act as sensors as they interact with the others. It turns out, as we have proven mathematically in Section 2.2, that the error signal becomes proportional to \( \sqrt{N}\delta \), corresponding to an effective detuning of \( \sqrt{N}\delta \). This is functionally equivalent to the clock transition frequency being enhanced by a factor of \( \sqrt{N} \).
Figure 3.10. Collective state energy levels, separated by $\omega_0$, are excited by a field of frequency $\omega$. All the states from $|E_0\rangle$ to $|E_N\rangle$ are excited, and participate in producing an effective clock transition frequency proportional to $\sqrt{N}$.

In the Ramsey fringe experiment, the error signal that is generated occurs as a result of the phase difference between the interacting states. A detailed picture can be viewed in Fig. 3.11. Consider first a single two level atom, initially in state $|1\rangle_A$, going through the Ramsey fields. In the Jaynes-Cummings model, when a field with $m$ photons interacts with an atom, the $\pi/2$-pulse will produce the quantum state $|\psi\rangle = |1\rangle_A|m\rangle_\nu - i|2\rangle_A|m-1\rangle_\nu$.

The energy of state $|2\rangle_A|m-1\rangle_\nu$ is lower than that of state $|1\rangle_A|m\rangle_\nu$ by $\hbar \delta$. In the second zone, these two composite states evolve freely for a time $T_2$ and accumulate different phases. State $|1\rangle_A$, with energy 0 remains the same, whereas $|2\rangle_A$ with energy $\omega_0$ evolves as $e^{i\omega_0 T_2}$. The field with $m$ photons evolve as $e^{im\omega T_2}$ whereas the field with $m-1$ photons evolve as $e^{i(m-1)\omega T_2}$. Thus, the quantum state of the total system at the end of the dark zone is

\[
|\psi\rangle = e^{im\omega T_2}|1\rangle_A|m\rangle_\nu - ie^{i\omega_0 T_2}e^{i(m-1)\omega T_2}|2\rangle_A|m-1\rangle_\nu
\]

The net accumulated phase difference in the two states is $e^{i\delta T_2}$. The third zone where another $\pi/2$-pulse occurs produces interference between the two states, so that when
interrogation occurs, the signal produced is in the form of Ramsey fringes that oscillate at frequency $\delta$. Therefore, the energy difference between the two composite states determines the oscillation frequency of the Ramsey fringes. Alternatively, if one were to plot the signal as a function of the dark zone time, $T_2$, the width of the fringe is given by the inverse of this energy difference. If the same calculation is carried out now for a two state system where the ground state is $|E_0\rangle_A|m\rangle$ and the excited state is $|E_N\rangle_A|m-N\rangle$, where $|E_0\rangle_A$ and $|E_N\rangle_A$ are the collective states of $N$ atoms, then the energy difference is $N\delta$ and the width of the fringe as a function of $T_2$ would be $1/(N\delta)$ and the width of the Ramsey fringe as a function of $\delta$ will be $(T_2^{-1}/N)$.

As mentioned earlier, such a two level system of collective states for a large value of $N$ is virtually impossible to realize for non-interacting atoms. Instead, for $N$ atoms, the first Ramsey zone produces a superposition of all the states from $|E_0\rangle_A$ to $|E_N\rangle_A$. In the second zone, each of the collective states $|E_k\rangle_A$ accumulates a phase factor of $e^{i(\delta T_2)k}$ with respect to the state $|E_0\rangle_A$. When the atoms pass through the third zone, each of these collective states interferes with one another and contributes to the total population of $|E_N\rangle_A$. It is the collection of these interferences among all the collective states that produces the narrowed linewidth.

We have verified this interpretation explicitly for two atoms. The collective states in this case are (where the subscript $A$ has been dropped) $|E_0\rangle$, $|E_1\rangle$, and $|E_2\rangle$. After they accumulate different phases in the second zone, each of them contributes to the final state $|E_2\rangle$ by amount $\chi_0 = 1/4$, $\chi_1 = e^{i\delta T}/2$, and $\chi_2 = e^{2i\delta T}/4$ respectively. The total signal is $S_{col} = |\langle E_2|E_2\rangle|^2 = \cos^4(\delta T_2/2)$. This comes about because $S_{col} = |\chi_0 + \chi_1 + \chi_2|^2 = |\chi_0 + \chi_1|^2 + |\chi_1 + \chi_2|^2 + |\chi_0 + \chi_2|^2 - (\chi_0^2 + \chi_1^2 + \chi_2^2)$. In other words, it is as though
Figure 3.11. Ramsey fringe experiment of a two level atom, in the Jaynes Cummings model, involves states \(|1\rangle_A |m\rangle_\nu\) and \(|2\rangle_A |m-1\rangle_\nu\) where the state with subscript \(A\) represents the atomic state, and subscript \(\nu\) represents the Ramsey field. The phase difference of the two levels at the end of the experiment is \(e^{i\delta T_2}\), and the signal produced would oscillate at frequency \(\delta\); If a two level system existed in which the ground state were the collective state \(|E_0\rangle_A |m\rangle_\nu\) and the excited state were the collective state \(|E_N\rangle_A |m - N\rangle_\nu\), the phase accumulation between the two states at the end of the Ramsey fringe experiment would be \(e^{iN\delta T_2}\), and the oscillation frequency would be \(N\delta\).

\(|E_0\rangle\) and \(|E_1\rangle\) interfered together to produce Ramsey fringes at frequency \(\delta\), \(|E_1\rangle\) and \(|E_2\rangle\) interfered together to produce Ramsey fringes at frequency \(\delta\), and \(|E_0\rangle\) and \(|E_2\rangle\) interfered together to produce Ramsey fringes at frequency \(2\delta\); the signal observed is the addition of all these Ramsey fringes minus an overall factor (see Fig. 3.12), which is due to the fact that the actual process is a simultaneous interference between the three states.

### 3.6.2. Violation of the conventional notion of the transit time limit

The narrowing of the COSAC fringe as given by \(\Gamma(N) = \Gamma(1)/\sqrt{N} = \pi/(T_2\sqrt{N})\) violates the conventional transit time limit, which constrains the fringe width to be at least \(\sim 1/T_2\).

This is a manifestation of the uncertainty relation \(\Delta f \cdot \Delta t \geq 1\), which apparently follows from the Heisenberg uncertainty principle of \(\Delta E \cdot \Delta t \geq \hbar\). However, when we properly define \(\Delta f\) as the uncertainty in the fringe width – in the case of the Ramsey technique considered here – and \(\Delta t\) as the total observation time, we can derive the uncertainty
relations more systematically and show that despite the fact that the conventional transit
time limit is violated, the Heisenberg uncertainty principle is not violated.

First, consider a single atom that undergoes the Ramsey fringe experiment. The
uncertainty in the fringe width is $\Delta f = (1/T_2)$, where $T_2$ is the separation period between
the two $\pi/2$ pulses. When the experiment is repeated $m$ times, it is as though the
separation period expands $m$-fold, so that the effective observation time is in fact $\Delta t = mT_2$, and the uncertainty in the fringe width is $\Delta f = (1/T_2)/\sqrt{m}$ in the standard quantum
limit (SQL) and $\Delta f = (1/T_2)/m$ in the Heisenberg limit (HL). Hence, the product $\Delta f \cdot \Delta t$
yields $\sqrt{m}$ in the SQL and 1 in the HL. Note that as $m \to 1$, the SQL approaches the
HL, which is the more fundamental limit.

Next, consider $N$ atoms in the same Ramsey fringe experiment during a single trial.
Since each atom, in its individual state, is considered separately from the rest, having $N$
atoms is equivalent to running $N$ trials simultaneously. The effective observation time in this case is $\Delta t = NT_2$, and the uncertainties in the fringe width are $\Delta f = (1/T_2)/\sqrt{N}$ in the SQL and $\Delta f = (1/T_2)/N$ in the HL. Moreover, if the experiment is repeated $m$ times, the effective observation time increases to $\Delta t = mNT_2$, and the uncertainties in the fringe width are $\Delta f = (1/T_2)/\sqrt{mN}$ in the SQL and $\Delta f = (1/T_2)/(mN)$ in the HL. Thus, we find that the uncertainty relations for $N$ atoms and $m$ trials are $\Delta f \cdot \Delta t = \sqrt{mN}$ in the SQL and $\Delta f \cdot \Delta t = 1$ in the HL.

Consider next the COSAC case, containing $N$ atoms, and repeated $m$ times. As we have shown in Section 3.5, the frequency fluctuation in the COSAC is $\Delta f = 1/(T_2\sqrt{mN})$ for ideal detection efficiency. It may not be obvious what the effective observation time is for this case. However, given the fact that, under ideal detection efficiency, the COSAC is equivalent to the case of $N$ atoms repeated $m$ times, we are led to conclude that the effective observation time is $\Delta t = T_2mN$. As such, we get $\Delta f \cdot \Delta t = \sqrt{mN}$, which is the SQL in this case. In the HL, we could get $\Delta f \cdot \Delta t = 1$. Thus, we see that when the frequency uncertainty and the observation times are interpreted properly, the COSAC signal does not violate the fundamental quantum limit.

3.7. Conclusion

We have described an atomic clock with a significant reduction in the Ramsey fringe linewidth, by a factor of $\sqrt{N}$, by measuring the amplitude of a collective state with a heterodyne detection scheme. We have shown that the reduction occurs due to multipath interference among the collective states, and does not violate the fundamental quantum limit. The performance of the COSAC has been compared to that of the CC by analyzing
quantum and classical fluctuations in frequency. When the effects of detector efficiency and collection efficiency are considered, it can be seen that the COSAC may perform 10 times better than a typical CC employing fluorescence detection.
CHAPTER 4

Coherent population trapping based collective state atomic clock

The demand for an accurate optical atomic clock and interferometer has continued ever since frequency combs from femtosecond lasers have been able to count high optical frequencies with precision. Besides their importance in frequency and time standards, atomic clocks and interferometers’ applications range from telecommunications and navigation to test of fundamental physics.

We have developed a new clock system in which we measure the population of atoms in a single collective state. In [13], we have shown that such a fundamentally new system has its strengths – reduction in linewidth by $\sqrt{N}$, where $N$ is the number of atoms; null measurement scheme involving optically dense atomic cloud leads to increased quantum efficiency in fluorescence measurement – and challenges – various inhomogeneities in the system may lead to a more pronounced loss in signal to noise ratio. It also leads to a deeper understanding of frequency fluctuation, that signal to noise ratio is not always a function of frequency, and transit time limit, that although the $\sqrt{N}$ reduction in linewidth occurs for collective state atomic clock (COSAC) for the same Ramsey fringe separation time as the conventional clock, and appears to be a strong violation of the conventional transit time limit, it nevertheless does not violate the Heisenberg uncertainty principle. Moreover, we have shown in [reference] that observation of collective states may lead to
better detection of spin squeezed states, which is necessary to achieve uncertainty level under the standard quantum limit.

In this paper, we extend the idea of COSAC to include basis states of the coherent population transfer (CPT) process \([64, 65, 66, 67]\). By applying the CPT technique, we can avoid the light shift issue and the necessity for an accurate \(\pi\) pulse time encountered in the off-resonant Raman setup. In Section 4.1 we describe the CPT process and its application to atomic clocks. In Section 4.2 we define the collective states in terms of CPT basis states, and discuss their application to atomic clocks. In Section ??, we elaborate on the null measurement scheme using an off-resonant Raman excitation in high optical density. Lastly, in Section ??, we conclude with a summary and an outlook.

### 4.1. CPT process

The CPT based atomic clock employs Raman interactions in \(\Lambda\)-type three level atomic system, shown in Fig. 4.1a. For traditional descriptions of CPT based atomic clocks, see \([68]\) The long lived ground states, \(|a\rangle\) and \(|b\rangle\), interact separately with the short lived excited state, \(|e\rangle\), via electric dipole interactions. Field of frequency \(\omega_1\), with detuning \(\delta_1\), couples \(|a\rangle\) and \(|e\rangle\), and field of frequency \(\omega_2\), with detuning \(\delta_2\), couples \(|b\rangle\) and \(|e\rangle\). In the \(\Lambda\)-type scheme, the ground states do not directly interact with each other. The strengths
of these interactions are given by Rabi frequencies \( \Omega_1 \equiv \mu_{ae} \cdot E_1 / \hbar \) and \( \Omega_2 \equiv \mu_{be} \cdot E_2 / \hbar \), where \( \mu \) is the dipole moment operator of the atom. After dipole approximation and rotating wave approximation, the Hamiltonian for this system is:

\[
H = \sum_{i=a,b,e} \epsilon_i \sigma_{ii} - \frac{\hbar}{2} \left( \Omega_1 e^{i(\omega_1 t - \phi_1)} \sigma_{ae} + \Omega_2 e^{i(\omega_2 t - \phi_2)} \sigma_{be} + H.c. \right)
\]

where \( \epsilon_i \) is the energy of state \( |i\rangle \), \( \sigma_{ij} \equiv |i\rangle \langle j| \), \( \phi_i \) for \( i = [1, 2] \) (\( j = [1, 2] \)) are the phases of the fields, and the Rabi frequencies are taken to be real. The Hamiltonian can be written in the rotating wave basis for convenience, in which the Hamiltonian is time-independent and the states evolve in time. The basis states in this frame are \( |\tilde{a}\rangle \equiv \exp[\frac{-i}{\hbar}(\omega_1 t + \phi_1)]|a\rangle \), \( |\tilde{b}\rangle \equiv \exp[\frac{-i}{\hbar}(\omega_2 t + \phi_2)]|b\rangle \), and \( |\tilde{e}\rangle \equiv |e\rangle \), and the Hamiltonian is:

\[
\tilde{H} = \frac{\hbar}{2} \left[ \Delta (\sigma_{\tilde{a}\tilde{a}} - \sigma_{\tilde{b}\tilde{b}}) - (2\delta + i\Gamma)\sigma_{\tilde{e}\tilde{e}} \right] - \frac{\hbar}{2} (\Omega_1 \sigma_{\tilde{a}\tilde{e}} + \Omega_2 \sigma_{\tilde{b}\tilde{e}} + H.c.)
\]

where \( \Gamma \) is the decay rate from state \( |\tilde{e}\rangle \). The equation of motion for the density matrix can be written as \( \dot{\rho} = (i/\hbar)[\tilde{\rho}, \tilde{H}] + \tilde{L}_{\tilde{e}\tilde{e}} \), where the source matrix, \( \tilde{L} = \Gamma_{aa} \sigma_{\tilde{a}\tilde{a}} + \Gamma_{eb} \sigma_{\tilde{b}\tilde{e}} \).

Next, we partially diagonalize the Hamiltonian by rotating it by \( R = \cos \theta (\sigma_{aa} + \sigma_{bb}) + \sin \theta (\sigma_{ab} - \sigma_{ba}) + \sigma_{ee} \), where \( \sin \theta = \Omega_1 / \Omega \) and \( \cos \theta = \Omega_2 / \Omega \), and \( \Omega \equiv \sqrt{\Omega_1^2 + \Omega_2^2} \). The basis states in this frame, as depicted in Fig. 4.1b, are the coherent superpositions of the ground states, \( |-\rangle = \cos \theta |\tilde{a}\rangle - \sin \theta |\tilde{b}\rangle \) and \( |+\rangle = \cos \theta |\tilde{a}\rangle + \sin \theta |\tilde{b}\rangle \), and the excited state \( |e\rangle = |\tilde{e}\rangle \). The equation of motion in this basis is given by \( \dot{\rho}_R = (i/\hbar)[\rho_R, H_R] + L_R \rho_{ee} \).

The rotated Hamiltonian is:

\[
H_R = \frac{\hbar}{2} \left[ C \Delta (\sigma_{--} - \sigma_{++}) - (2\delta + i\Gamma)\sigma_{ee} \right] + \frac{\hbar}{2} (S \Delta \sigma_{--} - \Omega \sigma_{++} + H.c.)
\]
where \( C \equiv \cos 2\theta \) and \( S \equiv \sin 2\theta \), and the rotated source matrix is \( L_R = (\Gamma/2)[(1 + Cd)\sigma_- + (1 - Cd)\sigma_+ + Sd(\sigma_{--} + \sigma_{++})] \), where \( d = (\Gamma_{ea} - \Gamma_{eb})/\Gamma \).

In this frame, \( |\rangle \), or the dark state, is decoupled from the excited state, \( |e \rangle \). Interaction with fields \( \omega_1 \) and \( \omega_2 \) couples only the bright state \( |+\rangle \) to \( |e \rangle \), and drives the transition between them. Spontaneous emission, however, drives the transition from \( |e \rangle \) to both \( |--\rangle \) and \( |+\rangle \). When \( d \neq 0 \), or \( \Gamma_{ea} \neq \Gamma_{eb} \), the source matrix also contributes to the coherence terms in the Hamiltonian. For simplicity, we only consider the case \( d = 0 \) for the rest of the paper, so that \( \Gamma_{ea} = \Gamma_{eb} = \Gamma/2 \). Thus, the interaction with \( \omega_1 \) and \( \omega_2 \) will optically pump the system into the dark state.

In the case that \( \Gamma \gg S\Delta, \Omega \), the excited state will come into equilibrium with the ground states rapidly at rate \( \Gamma \) while the ground states change at a time scale much greater than \( 1/\Gamma \). Hence, we can adiabatically eliminate \( |e \rangle \). For \( |\Psi \rangle = c_-(t)|\rangle + c_+(t)|+\rangle + c_e(t)|e \rangle \), we can make the substitution

\[
(4.4) \quad c_e \simeq i[\Omega/(\Gamma - 2i\delta)]c_+
\]

Hence, the system can be reduced to just two states: the dark state \( |--\rangle \) and the damped bright state

\[
(4.5) \quad |+\rangle_d \equiv |+\rangle + i\frac{\Omega}{\Gamma - 2i\delta}|e \rangle
\]
The wave function is now $|\Psi\rangle = c_- |\rangle - \rangle + c_+ |+\rangle_d$, and the Hamiltonian in the new basis states is:

$$H = \frac{\hbar}{2} \left( \begin{array}{cc} C\Delta & S\Delta \\ S\Delta & -(C\Delta - 2\beta) - 2i\alpha \end{array} \right)$$

(4.6)

where $\alpha$ is the effective damping rate given by,

$$\alpha \equiv \frac{1}{2} \frac{\Omega^2 \Gamma}{\Gamma^2 + 4\delta^2} = \left| \frac{\Omega}{\Gamma - 2i\delta} \right|^2 \frac{\Gamma}{2}$$

(4.7)

and $\beta$ is the additional light shift between the states, given by,

$$\beta \equiv \frac{\Omega^2 \delta}{\Gamma^2 + 4\delta^2} = \left| \frac{\Omega}{\Gamma - 2i\delta} \right|^2 \delta$$

(4.8)

The effective coupling rate between $|\rangle - \rangle$ and $|+\rangle_d$ is $S\Delta$, and the decay from $|+\rangle_d$ to $|\rangle -$ occurs at rate $\alpha$. The Hamiltonian can be made symmetric by subtracting $\hbar \beta/2$ from the diagonal elements:

$$H = \frac{\hbar}{2} [-\beta' \sigma_{--} + (\beta' - 2i\alpha)\sigma_{++d} + S\Delta \sigma_{+-} + H.c.]$$

(4.9)

where $\sigma_{++d} = |+\rangle_d \cdot d \langle +|$ and $\beta' \equiv \beta - C\Delta$. Hence, the energy difference between $|\rangle -$ and $|+\rangle_d$, or the ac Stark shift, is $\beta'$.

The traditional CPT based clock utilizes atoms that are either in vapor or trapped with laser and magnetic fields, that interact with either a single laser field, or two pulses of laser fields that are separated in time. They initially populate both ground states equally. In the previous case, atoms interact with co-propagating laser fields of frequency $\omega_1$ and $\omega_2$ that are on resonance. The interaction drives the transition from $|+\rangle$ to $|\rangle -$,
until the population in \(|+\rangle\) is depleted in time scale larger than \(1/\alpha\). When \(\Delta \neq 0\), the states are coupled at rate \(S\Delta\). Plotting \(\rho_{ee}\) as a function of \(\Delta\), the integrated signal over the interaction time is that of the electromagnetically induced transparency (EIT) effect \([69, 70, 71, 72]\).

In the latter case, there is Raman interaction between the atoms and the laser fields for time scale \(T_1 \gg 1/\alpha\) to optically pump the atoms into the dark state. During the next period \(T_2\), the Raman interaction is turned off, so that the only interaction present is the Rabi flopping between \(|-\rangle\) and \(|+\rangle\) at rate \(S\Delta\). In the language of the Bloch vector model, the Bloch vector at the end of the first interaction zone is \(\mathbf{R}(T_1) = \hat{\mathbf{e}}_3\). Since \(\mathbf{R}\) rotates at rate \(S\Delta\) around \(\hat{\mathbf{e}}_1\), the Bloch vector at the end of the dark zone is \(\mathbf{R}(T_1 + T_2) = \sin(S\Delta T_2)\hat{\mathbf{e}}_2 + \cos(S\Delta T_2)\hat{\mathbf{e}}_3\). In the next interaction zone, the atoms interact with the Raman fields with the same phases as in the first zone. Again, there will be decay to the dark zone at the time scale \(T_3 \gg 1/\alpha\). Since the signal observed from fluorescence is \(S_f = \Gamma \rho_{ee} = 2\alpha \rho_{++}\) from Eq. (4.4), and \(\rho_{++} = (1 - R_3)/2\), the fluorescence at time \(t\) is

\[
(4.10) \quad \quad S_{f,+}(T_1 + T_2 + t) = \alpha[1 - \cos(S\Delta T_2)]e^{-\alpha t}
\]

Thus, the integrated signal at time \(T_3\) is

\[
(4.11) \quad \quad \bar{S}_{f,+}(T_1 + T_2 + T_3) = \int_0^{T_3} S_f(T_1 + T_2 + t) dt \approx \sin^2 \left(\frac{S\Delta T_2}{2}\right)
\]

In other words, the (integrated) signal, as a function of \(\Delta\), will produce Ramsey fringes. This is the signal proportional to the population in the bright state. Alternatively, the
signal that one would observe for the population in the dark state is

\begin{equation}
\bar{S}_{f,-}(T_1 + T_2 + T_3) \simeq \cos^2(S\Delta T_2/2)
\end{equation}

4.2. Collective state CPT clock

The discussion can be generalized to \( N \) atoms that are all excited by the same field. We assume that there are no overlaps between the wavefunctions of the atoms and there is no interaction among them \([1]\). The evolution of each atom under these assumptions can be described individually, and the total quantum state is simply the outer (tensor) product of individual quantum states \([44, 45]\). However, the interaction can also be described equivalently using a basis of collective states \([44, 1]\). The Hilbert space of \( N \) two level atoms is spanned by \( 2^N \) states. Thus, when transformed to the collective state basis, there are also \( 2^N \) collective states. For identical Rabi frequencies and resonant frequencies, however, only the generalized symmetric states \([44]\), of which there are only \((N+1)\), are relevant, and the rest of the \((2^N - N - 1)\) states become decoupled. The case where inhomogeneity of the Rabi frequencies and different Doppler shifts experienced by different atoms are taken into account is presented at the end of this section. We also note that if different atoms see different phase factors from the excitation fields, these factors can be absorbed into the definition of the generalized symmetric states \([44]\).

The simplified symmetric states, known as the conventional Dicke states \([1]\), represent the case where it is assumed that the mean separation between the atoms is much less than the wavelength corresponding to the two level transition (which, for the co-propagating off resonant Raman excitation, is \( \sim (k_1 - k_2)^{-1} \)). While this constraint is not necessary for the concept proposed here \([44]\), it is easier to describe the process initially under this
constraint. The observables computed remain correct when this constraint is not met. Some of these Dicke states are as follows: \( |E_0\rangle \equiv |---\cdots-\rangle \), \( |E_1\rangle \equiv \sum_{i=1} |---\cdots+i\cdots-\rangle /\sqrt{N} \), \( |E_2\rangle \equiv \sum_{i,j\neq i} |---\cdots+i\cdots+j\cdots-\rangle /\sqrt{NC_2} \), \( |E_3\rangle \equiv \sum_{i,j,k} |---\cdots+i\cdots+j\cdots+k\cdots-\rangle /\sqrt{NC_3} \), and \( |E_N\rangle \equiv |++\cdots+\rangle \) where \( NC_n = N!/(n!(N-n)!) \). For instance, \( |E_2\rangle \) is the Dicke state with two atoms in \( |+\rangle \) and the rest in \( |-\rangle \). Any two atoms can be in \( |+\rangle \) with equal probability, with \( NC_2 = N(N-1)/2 \) such possible combinations.

The Hamiltonian in the basis of the symmetric collective states, excluding the \( \alpha \) term, is \( H = \hbar \sum_{k=0}^{N} \left[ \beta' (2k - N)/2 - i\alpha k \right] |E_k\rangle \langle E_k| + \sum_{k=0}^{N-1} \left[ \hbar C'_k |E_k\rangle \langle E_{k+1}| + H.c. \right] \)

where \( C'_{k+1} = \sqrt{N-k} \sqrt{k+1} C \Delta \) is the Rabi frequency between collective states \( [44, 1] \).

The states are separated by \( \hbar/\beta' \) in energy and couple at different rates. For instance, \( C'_1 = C'_N = \sqrt{N} C \Delta \), \( C'_2 = C'_{N-1} = \sqrt{2(N-1)} C \Delta \), etc. The middle states have the strongest coupling rate of \( C'_{N/2} = NC \Delta \) and the end states couple most weakly.

The final state of the system during the second pulse can be derived by using either the collective state picture or, equivalently, the single atom picture. For a large value of \( N \), carrying out the calculation in the collective states basis is numerically cumbersome and analytically intractable. However, we can find the state trivially by using the single atom
picture and then determining the coefficients of the collective states by simple projection, given the definition of the \((N + 1)\) generalized symmetric collective states.

\[
\bar{S}_{+,C} = \sin^{2N}(S\Delta T_2/2)
\]

which is simply \(\bar{S}_{f,+}\). This quantity, \(\bar{S}_{+,C}\), represents the probability of finding the whole system in the state \(|E_N\rangle\) whereas \(\bar{S}_{f,+}\) represents the probability of finding each atom in state \(+\rangle\). In a conventional experiment, the population of atoms in state \(+\rangle\) is measured, for example, by collecting fluorescence produced by coupling \(+\rangle\) to an auxiliary state. The resulting signal is \(\bar{S}\), independent of the number of atoms. The experiment that we propose, to be described shortly, produces the signal \(\bar{S}_{-,C} = \cos^{2N}(S\Delta T_2/2)\). When \(\bar{S}_{-,C}\) is plotted for various values of \(N\) (Fig. 4.2 (b)), it is evident that the linewidth of the fringe as a function of \(\theta\) decreases as \(N\) increases.

Hence, the basis states for the CPT process can also be used to build a COSAC. The same inhomogeneities present in the off-resonant Raman process based COSAC are also present here, and the experimental setup and the detection scheme remain the same. The difference lies in the pulse widths, and that the experiment consists of an interaction zone, a dark, free precession zone, and a detection zone. In the detection zone, instead of using CPT beams, the same off-resonant excitation is performed to ensure that the photons emitted are stimulated and that they travel in the same propagation direction as the read beam. Lastly, one must be careful in dealing with the CPT pulses; a slight difference in phase of the read beam, with respect to the write beam, would result in unwanted mixing between the dark state and the bright state, resulting in tarnished signal.
CHAPTER 5

Experimental considerations

5.1. Optical system

The purpose of the experiment is to detect the collective state $|E_N\rangle$, first with a single $\pi$ pulse, and then with an experiment involving separated Ramsey fields. These experiments involve knowing the precise width(s) of the applied Raman pulse(s). Also important is the pulse width of the detection zone, which is qualitatively different from the experimental pulses since only a single pulse is applied and the signal that we’re interested in is the photons emitted from the stimulated Raman adiabatic passage. In order to calculate the experimental parameters, we need to first take a look at the energy levels in rubidium.

The $D_2$ lines in Rubidium 85 have two hyperfine ground states $5^2S_{1/2}$, $F = 2$ and $F = 3$, which are $3.0357324390(60)$ GHz apart [24]. The atoms in these states can be optically excited to the $5^2P_{3/2}$ states with hyperfine levels $F' = 1, 2, 3, 4$. Initially, the atoms are in a magneto optical trap (MOT) that uses the $F = 3$ to $F' = 4$ closed transition (red detuned by 10 MHz) with the repump beam that optically pumps the atoms from $F = 2$ to the excited states. For more information on laser cooling and trapping, some helpful references are [73, 74, 59, 60, 58, 75]. Fig. 5.2 shows the paths of the beams. Turning off the repump beam first and then the trap beam initializes the atoms in $F = 2$ state for the clock experiment. However, there is no direct transition from $F' = 4$ to
Figure 5.1. The energy diagram of rubidium 85 D2 line. The energy levels are $|a\rangle \equiv |F = 2, m_f = 0\rangle$, $|b\rangle \equiv |F = 3, m_f = 0\rangle$, $|c\rangle \equiv |F' = 2, m_{f'} = 1\rangle$, $|d\rangle \equiv |F' = 3, m_{f'} = 1\rangle$, and $|e\rangle \equiv |F' = 4, m_{f'} = 1\rangle$. The states $|a\rangle$ and $|b\rangle$ are the lower legs of the $\Lambda$ transition, and the states $|c\rangle$ and $|d\rangle$ form an effective single excited level in the $\Lambda$ transition. The state $|e\rangle$ is brought into discussion because the Raman beams of frequency $\omega_1$ and $\omega_2$ are brought from a laser that is locked 90 MHz away from the $|b\rangle \rightarrow |e\rangle$ transition. The values of $\delta_c$ and $\delta_d$ are discussed in the main text.

$F = 2$. This is in fact an off-resonant transition from $F' = 3$ to $F = 2$, detuned by $\delta' = 110.640(68) \text{ MHz} = 18.238 \Gamma$ (10 MHz less than the difference in energy between $F' = 3$ and $F' = 4$). The decay rate of the D2 line is $2\pi \cdot 6.0666(18) \text{ MHz}$. The probability of populating the state $F' = 3$ is $\rho_{3'3'} = \Omega'^2/(2\Omega'^2 + \Gamma^2 + 4\delta')$, where $\Omega'$ is the Rabi frequency of the MOT beams. Taking the saturation limit, $\Omega'^2 = \Gamma^2/2$, we find $\rho_{3'3'} = 3.75 \cdot 10^{-4}$. Since there are two ground states, the rate of optically pumping a single atom to state $F = 2$ is $\Gamma' \simeq \Gamma/2$, so that the total optical pumping rate is $R_{op} \simeq \rho_{3'3'} \cdot \Gamma/2 = 2\pi \cdot 1.14$ kHz. The time that it requires for this to occur is $T_{op} \sim \text{880 } \mu\text{s}$. After this time period, all the atoms are optically pumped to $F = 2$.

However, we can shorten this time by several orders of magnitude by using a beam that is resonant to $|b\rangle \rightarrow |d\rangle$ transition. Then $\rho_{3'3'} \sim \frac{1}{2}$ and $R_{op} \sim \Gamma/4 = 2\pi \cdot 1.5166(5)$ MHz, so that the optical pumping period is $T_{op} \sim 0.7 \mu\text{s}$. This is done by placing a 30
MHz AOM in the path of a beam that is already red shifted from $F' = 4$ transition by 90 MHz, and downshifting the beam further.

5.2. Raman Rabi frequency calculation

The beam that is used for the clock experiment comes from the MOT laser, so that it is initially locked to $F = 3$ to $F' = 4$ transition. A beamsplitter takes this beam and sends it into two different paths, each of which contains an acousto optic modulator (AOM).
Figure 5.3. Optical paths of the off-resonant Raman beams. Laser 1, AOM 1, D1, D2, and Rb are the same as in the previous figure. The Raman beams come from the same laser as the MOT beams. The Raman beams go through polarizers, marked P in the figure, and then through AOM 4 and AOM 5, which are Brimrose GPF-1500-200-630 models. They have efficiency of well over 15% when they are at their best. A non-polarizing beam splitter, NPBS, combines the two Raman beams, dumps half the power, and send the other half through a fiber coupler, FC, to ensure that the beams are co-propagating. They then go through a tapered amplifier (after an Electro Optic Tech Isolator not drawn in the figure), and then through a switching IntraAction 40 MHz AOM, AOM 6. After a two lens system to enlarge the beams, they combine with the MOT-z beam and go through the cell.

One AOM upshifts the beam by 1.517866 GHz and the other downshifts it by 1.517866 GHz. The AOMs are driven by a single frequency synthesizer that stabilizes the frequency to the kHz range, and keeps it from drifting away from 1.517866 GHz. The Λ scheme in the clock experiment involves $|F = 2, m_F = 0\rangle \equiv |a\rangle$ and $|F = 3, m_F = 0\rangle \equiv |b\rangle$ as the lower states. These states are coupled to the excited states $|F' = 2, m_{F'} = 1\rangle \equiv |c\rangle$ and
Figure 5.4. The dipole matrix elements of relevant levels. The transitions, $|a\rangle \rightarrow |c\rangle$, $|a\rangle \rightarrow |d\rangle$, $|b\rangle \rightarrow |c\rangle$, and $|b\rangle \rightarrow |d\rangle$, are directly involved in the Raman transition. However, the other two are also necessary to calculate the effective light shift. For simplicity, the dipole matrix elements from $|a\rangle$ are to the left side of their respective transition lines, and those from $|b\rangle$ are to the right.

<table>
<thead>
<tr>
<th>Label</th>
<th>Transition $(\sigma^+)$</th>
<th>Dipole matrix element</th>
<th>$I_{\text{sat}}$ [mW/cm$^2$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>$</td>
<td>F = 2, m_F = 0\rangle$ to $</td>
<td>F' = 2, m_{F'} = 1\rangle$</td>
</tr>
<tr>
<td>2</td>
<td>$</td>
<td>F = 2, m_F = 0\rangle$ to $</td>
<td>F' = 3, m_{F'} = 1\rangle$</td>
</tr>
<tr>
<td>3</td>
<td>$</td>
<td>F = 3, m_F = 0\rangle$ to $</td>
<td>F' = 2, m_{F'} = 1\rangle$</td>
</tr>
<tr>
<td>4</td>
<td>$</td>
<td>F = 3, m_F = 0\rangle$ to $</td>
<td>F' = 3, m_{F'} = 1\rangle$</td>
</tr>
</tbody>
</table>

Table 5.1. Transitions involved in the collective clock experiment, their dipole matrix elements, and the corresponding saturation intensities.

$|F' = 3, m_{F'} = 1\rangle \equiv |d\rangle$ by co-propagating $\sigma^+$ Raman beams detuned from $F = 3$ to $F' = 4$ transition by 1.518 GHz (described in the previous paragraph). The states $F' = 1$ and $F' = 4$ are not involved in the Raman transition. Those that are involved are listed in Table 5.1 with their properties.

Using alphabet subscripts to denote the levels and numerical subscripts to denote the transitions, the Hamiltonian in the basis of states $|a\rangle$, $|b\rangle$, $|c\rangle$, and $|d\rangle$ after dipole
approximation and rotating wave approximation is:

\( H_0 = \hbar \begin{pmatrix} \omega_a & 0 & -\frac{\Omega_1}{2}e^{i\omega_1 t} & -\frac{\Omega_2}{2}e^{i\omega_2 t} \\ 0 & \omega_b & -\frac{\Omega_3}{2}e^{i\omega_3 t} & -\frac{\Omega_4}{2}e^{i\omega_4 t} \\ -\frac{\Omega_1}{2}e^{i\omega_1 t} & -\frac{\Omega_3}{2}e^{i\omega_3 t} & \omega_c & 0 \\ -\frac{\Omega_2}{2}e^{i\omega_2 t} & -\frac{\Omega_4}{2}e^{i\omega_4 t} & 0 & \omega_d \end{pmatrix} \)

Rotating wave transformation can be performed on this Hamiltonian to yield it in the rotated bases of \(|a\rangle, |b\rangle, |c\rangle, \) and \(|d\rangle\):

\( H_{\text{rot}} = \hbar \begin{pmatrix} \frac{\delta}{2} & 0 & -\frac{\Omega_1}{2} & -\frac{\Omega_2}{2} \\ 0 & -\frac{\delta}{2} & -\frac{\Omega_3}{2} & -\frac{\Omega_4}{2} \\ -\frac{\Omega_1}{2} & -\frac{\Omega_3}{2} & -\delta_c & 0 \\ -\frac{\Omega_2}{2} & -\frac{\Omega_4}{2} & 0 & -\delta_d \end{pmatrix} \)

where \( \delta_c \equiv (\delta_{ac} + \delta_{bc})/2 \) is the average detuning from the lower states to state \(|c\rangle\), \( \delta_d \equiv (\delta_{ad} + \delta_{bd})/2 \) is the average detuning from the lower states to state \(|d\rangle\), and the two photon detuning is \( \delta = \delta_{ac} - \delta_{bc} = \delta_{bd} - \delta_{ad} \). The physical values of \( \delta_c \) and \( \delta_d \) can be calculated by noting that the laser is locked to 90 MHz downshifted from the \(|b\rangle\) to \(|c\rangle \equiv |F' = 4, m_{f'} = 1\rangle\) transition (with a 90 MHz AOM in the saturated absorption spectroscopy setup) and detuned by \( \Delta = 1.5178662 \) GHz by the Raman AOMs. The energy difference between \(|c\rangle\) and \(|e\rangle\) is \( \omega_{ce} = 183.042(29) \) MHz, and between \(|d\rangle\) and \(|e\rangle\) is \( \omega_{de} = 120.640(68) \) MHz. Then the laser frequency \( \omega_1 \equiv \omega_{ac} + \delta_{ac} = \omega_{be} + \Delta - 90 \) MHz, where we can substitute \( \omega_{be} = \omega_{ac} + \omega_{ce} - \omega_{ab} \). The value of \( \Delta \) is set such that \( \omega_{ab} = 2\Delta \). We can finally see that, when \( \delta = 0 \), \( \delta_{ac} = -1.424 \) GHz. Carrying out the
algebra in this manner for the other detunings reveal that \(\delta_c = \delta_{ac} = \delta_{bc} = -1.424\) GHz and \(\delta_d = \delta_{ad} = \delta_{bd} = -1.487\) GHz.

With the conditions \(\delta_c, \delta_d \gg \Omega_i\) where \(i = 1, 2, 3, 4\), The Hamiltonian in Eq. (5.2) can be reduced to:

\[
(5.3) \quad H_{\text{red}} = \hbar \left( \frac{\delta}{2} + \frac{\Omega_1^2}{4\delta_c} + \frac{\Omega_2^2}{4\delta_d} + \frac{\Omega_1\Omega_3^*}{4\delta_c} + \frac{\Omega_2\Omega_4^*}{4\delta_d} \right)
\]

in the basis of states \(|a\rangle\) and \(|b\rangle\). Thus, the states \(|c\rangle\) and \(|d\rangle\) can be effectively grouped into a single state that provides the adiabatic passage of stimulated off-resonant Raman transition. The quantity that provides the strength, or the rate, of the coupling between the states \(|a\rangle\) and \(|b\rangle\) is \(\Omega_R \equiv \Omega_1\Omega_3^*/4\delta_c + \Omega_2\Omega_4^*/4\delta_d\). Using the definition \(I_i/I_{\text{sat},i} = 2\Omega_i^2/\Gamma^2\), we can rewrite this quantity as:

\[
(5.4) \quad \Omega_R = \frac{\Gamma^2}{8} \left( \frac{1}{\delta_c\sqrt{I_{\text{sat},1}I_{\text{sat},3}}} + \frac{1}{\delta_d\sqrt{I_{\text{sat},2}I_{\text{sat},4}}} \right) I \approx 2\pi \cdot 627 \text{ [Hz]} \cdot I
\]

where the intensity \(I = I_i\) for all \(i = 1, 2, 3, 4\). The experimental pulse with width \(\pi/2\) is achieved when \(\Omega_R \tau = \pi/2\), or \(I \cdot \tau = 3.987 \cdot 10^{-4}\) mW s/cm\(^2\). The pulse period \(\tau\) is limited by the fact that the atoms released from the MOT will expand thermally while free falling. The most probable velocity with which the atoms expand thermally is given by \(v = \sqrt{2k_B T/m}\). The lower limit on the temperature of the MOT is given by the Doppler cooling limit: \(k_B T_D = \hbar \Gamma/2\) where \(T_D\) is the temperature at which the rate of heating (due to spontaneous emission) equals the rate of cooling. For rubidium 85, \(T_D = 146 \mu\text{K}\). However, the temperature of the MOT depends on other factors such as the detuning of the MOT lasers (we detune our lasers by 10 MHz), the magnetic field gradient produced...
by the anti-Helmholtz coils (our field gradient is about 10-15 G/cm), and the pressure of
the vacuum chamber. If we use the ideal case of the Doppler limit, the average velocity of
released atoms is \( v = 0.168 \) m/s. Table 5.2 shows some values of the height to which the
atoms will fall for a given time period: \( h = vt + \frac{gt^2}{2} \). However, it is to be noted that the
atoms in the center of the trap are colder than the atoms in the periphery, by the nature
of the MOT. Then we can estimate the fall of atoms from the center of the MOT to be
\( h = \frac{gt^2}{2} \), and the expansion of the spatial profile of the MOT to increase by \( vt \).

The experiment has to be carried in a concise enough time such that the width of the
expanding collection of atoms and the height that the atoms fall are contained within the
width of the Raman beams. The width of the Raman beams cannot be made arbitrarily
large since the power falls as the square of the radius. For a 1 ms experiment, the Raman
beams must be more than 1.36 mm in width to account for the size of the MOT (about 1
mm) and the symmetric width around it of 0.173 mm. (In fact, there is a more stringent
reason to expand the Raman beams by at least five times the size of the MOT, which
involves the effect of inhomogeneous field on the collective state signal, as is discussed
elsewhere).

<table>
<thead>
<tr>
<th>( \tau ) [ms]</th>
<th>height [mm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.001</td>
<td>0.0002</td>
</tr>
<tr>
<td>0.01</td>
<td>0.0017</td>
</tr>
<tr>
<td>0.1</td>
<td>0.0169</td>
</tr>
<tr>
<td>1</td>
<td>0.1732</td>
</tr>
<tr>
<td>2</td>
<td>0.3562</td>
</tr>
<tr>
<td>5</td>
<td>0.9641</td>
</tr>
<tr>
<td>8</td>
<td>1.6602</td>
</tr>
<tr>
<td>10</td>
<td>2.1733</td>
</tr>
</tbody>
</table>

Table 5.2. Height to which the atoms fall (due to freefall and thermal expansion) after they are released from the MOT after various time intervals.
The frequency synthesizer is limited to 1 kHz step in frequency. In order to smoothly plot the signal as a function of frequency, the Raman Rabi frequency should be at least \( \Omega_R = 2\pi \cdot 10 \text{ kHz} \). Table 5.3 lists various values of \( \Omega_R \), the intensity, \( I \), the effective value of power, \( P = \pi r^2 I \), for the MOT size of \( r = 1 \text{ mm} \) (which is probably larger than what we have in the lab), the actual value of power we need for a beam of radius \( r = 3 \text{ mm} \), and the period of a single zone \( \pi \) pulse.

In the experiment, the Raman beams are collimated and sent through a fiber coupler, and then through a free space tapered amplifier (TA). The TA produces about 560 mW of power. About 60 mW is from the amplifier itself, and the two legs of the Raman beams have even powers of about 250 mW. However, they go through an AOM and lose a lot of power. More importantly, too strong of a Raman transition would require pulses that are much smaller than 1 \( \mu \text{s} \), which is difficult to achieve in the laboratory. Therefore, we allow the intensity per beam to be about 16 mW/cm\(^2\). Then \( \Omega_R \sim 20 \text{ kHz} \) in our system, and the period of the \( \pi \) pulse is \( \tau = 25 \mu \text{s} \).

### 5.3. Calculation of light shift

In trying to detect the signal from the single zone experiment, and, later on, the three zone experiment, we need to be aware of systematic imperfections. One of these is the light shift. When a two level atomic system – with ground state \( |1\rangle \) and excited state \( |2\rangle \) – with energy separation, \( \omega_0 \), interacts with a light field of frequency \( \omega \), the Hamiltonian is simply,

\[
H_{\text{red}} = \hbar \begin{pmatrix}
0 & \Omega_0/2 \\
\Omega_0/2 & -\delta_0 - i\Gamma/2
\end{pmatrix}
\]
For a given Raman Rabi frequency, $\Omega_R$, desired, the values of intensity in each Raman beam, $I$, the effective power for the MOT ($r=1$ mm), the actual power needed (for a $r = 3$ mm beam), and the $\pi$-pulse period are tabulated.

Table 5.3.

<table>
<thead>
<tr>
<th>$\Omega_R$ [kHz]</th>
<th>$I$ [mW/cm$^2$]</th>
<th>$P$ [mW] ($r=1$ mm)</th>
<th>$P$ [mW] ($r=3$ mm)</th>
<th>$\tau$ [$\mu$s]</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>15.9</td>
<td>0.5</td>
<td>4.5</td>
<td>50</td>
</tr>
<tr>
<td>50</td>
<td>79.7</td>
<td>2.5</td>
<td>22.6</td>
<td>10</td>
</tr>
<tr>
<td>100</td>
<td>159.5</td>
<td>5.0</td>
<td>45.1</td>
<td>5</td>
</tr>
<tr>
<td>150</td>
<td>239.2</td>
<td>7.5</td>
<td>67.6</td>
<td>3.33</td>
</tr>
<tr>
<td>200</td>
<td>319.0</td>
<td>10.0</td>
<td>90.2</td>
<td>2.5</td>
</tr>
<tr>
<td>250</td>
<td>398.7</td>
<td>12.5</td>
<td>112.7</td>
<td>2</td>
</tr>
<tr>
<td>300</td>
<td>478.5</td>
<td>15.0</td>
<td>135.3</td>
<td>1.67</td>
</tr>
<tr>
<td>350</td>
<td>558.2</td>
<td>17.5</td>
<td>157.8</td>
<td>1.43</td>
</tr>
<tr>
<td>400</td>
<td>638.0</td>
<td>20.0</td>
<td>180.4</td>
<td>1.25</td>
</tr>
<tr>
<td>450</td>
<td>717.7</td>
<td>22.5</td>
<td>202.9</td>
<td>1.11</td>
</tr>
<tr>
<td>500</td>
<td>797.4</td>
<td>25.1</td>
<td>225.5</td>
<td>1</td>
</tr>
<tr>
<td>1000</td>
<td>1594.9</td>
<td>50.1</td>
<td>450.9</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Table 5.4. Possible transitions from $|F = 2, m_F = 0\rangle$.

<table>
<thead>
<tr>
<th>Label</th>
<th>Transition ($\sigma+$)</th>
<th>Dipole matrix element</th>
<th>$I_{sat}$ [mW/cm$^2$]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1'</td>
<td>$</td>
<td>F' = 1, m_{F'} = 1\rangle$</td>
<td>$\sqrt{1/20}$</td>
</tr>
<tr>
<td>2'</td>
<td>$</td>
<td>F' = 2, m_{F'} = 1\rangle$</td>
<td>$\sqrt{7/36}$</td>
</tr>
<tr>
<td>3'</td>
<td>$</td>
<td>F' = 3, m_{F'} = 1\rangle$</td>
<td>$\sqrt{4/45}$</td>
</tr>
</tbody>
</table>

Table 5.5. Possible transitions from $|F = 3, m_F = 0\rangle$.

where $\delta_{0} = \omega_{0} - \omega$, and $\Omega_{0}$ is the Rabi frequency. Applying the Schrödinger equation,

$$
\dot{C}_1 = -i \frac{\Omega_{0}}{2} \bar{C}_2
$$

$$
\dot{C}_2 = -i \left[ \frac{\Omega_{0}}{2} \bar{C}_1 - (\delta_{0} + i \frac{\Gamma}{2}) \bar{C}_2 \right]
$$

(5.6)
where $\tilde{C}_1$ and $\tilde{C}_2$ are the coefficients in $|\Psi\rangle = \tilde{C}_1|1\rangle + \tilde{C}_2|2\rangle$.

Decoupling the differential set of equations, we see that:

\begin{equation}
\dot{\tilde{C}}_1 = -i \left[ \frac{\Omega_0^2 \delta_0}{4\delta_0^2 + \Gamma^2} - i \frac{\Omega_0^2 \Gamma}{2(\delta_0^2 + \Gamma^2)} \right] \tilde{C}_1
\end{equation}

The first term, $\beta = \Omega_0^2 \delta_0/(4\delta_0^2 + \Gamma^2)$ is the light shift due to the interaction with light. The second term, $\alpha = \Omega_0^2 \Gamma/[2(\delta_0^2 + \Gamma^2)]$ is the effective decay rate. Because there are three possible transitions from $|F = 2, m_F = 0\rangle$ to the excited states (see Table 5.4), the $\beta$ terms from all of them are added up to give the value $\beta_2 = \beta_{1'}, \beta_2', \beta_3'$. Likewise, for the three transitions from $|F = 3, m_F = 0\rangle$ (see Table 5.5), the light shifts are added up to yield $\beta_3 = \beta_{4'}, \beta_5', \beta_6'$. For a positive detuning, the lower energy levels shift down, and for negative detuning, they shift up. If $\beta_2 > \beta_3$, the two photon detuning is $\delta < 0$. Therefore, the peak frequency for the Raman Rabi transition will decrease. For example, working with $\Omega_R = 20$ kHz will shift the Raman peak from the initial value of $\Delta = 1.517866$ GHz to $\Delta_{\text{new}} = 1.517866$ GHz +29 kHz.

5.4. Magnetic coils

The MOT coils, which are in anti-Helmhotlz configuration, and the bias coils, which are in Helmholtz configuration, are magnetic wires wound around custom made aluminum coil holders. After using a Python code to ascertain that the coils that we build will not be physically possible, we have information on how many wires we need to wind per layer, how many layers we need to wind, what current is needed for a given radius of each coil and distance between the coils, and what magnetic field profile will be. Fig. 5.5 is the picture of the coils used for our final MOT design. For the MOT coils, which are the
Table 5.6. Calculations of light shifts for various $\Omega_R$. The overall light shifts are approximately an order of magnitude smaller than the values of $\Omega_R$.

<table>
<thead>
<tr>
<th>$\Omega_R$ [kHz]</th>
<th>$\beta_2$ [kHz]</th>
<th>$\beta_3$ [kHz]</th>
<th>$\beta$ [kHz]</th>
</tr>
</thead>
<tbody>
<tr>
<td>10</td>
<td>20.4</td>
<td>19.0</td>
<td>-1.5</td>
</tr>
<tr>
<td>50</td>
<td>102.1</td>
<td>94.8</td>
<td>-7.3</td>
</tr>
<tr>
<td>100</td>
<td>204.1</td>
<td>189.5</td>
<td>-14.6</td>
</tr>
<tr>
<td>150</td>
<td>306.2</td>
<td>284.3</td>
<td>-21.9</td>
</tr>
<tr>
<td>200</td>
<td>408.2</td>
<td>379.0</td>
<td>-29.2</td>
</tr>
<tr>
<td>250</td>
<td>510.3</td>
<td>473.8</td>
<td>-36.5</td>
</tr>
<tr>
<td>300</td>
<td>612.3</td>
<td>568.6</td>
<td>-43.8</td>
</tr>
<tr>
<td>350</td>
<td>714.4</td>
<td>663.3</td>
<td>-51.1</td>
</tr>
<tr>
<td>400</td>
<td>816.4</td>
<td>758.1</td>
<td>-58.3</td>
</tr>
<tr>
<td>450</td>
<td>918.5</td>
<td>852.9</td>
<td>-65.6</td>
</tr>
<tr>
<td>500</td>
<td>1020.6</td>
<td>947.6</td>
<td>-72.9</td>
</tr>
<tr>
<td>1000</td>
<td>2041.1</td>
<td>1895.2</td>
<td>-145.9</td>
</tr>
</tbody>
</table>

Figure 5.5. Coils for the experiment. The top ones, with radius of $r = 2.8$ cm, are of the bias coils. They provide about 2.5 G at the center when separated in distance by 6 cm and 0.5 A of current is provided. The bottom ones, with radius of $r = 1.5$ cm, are of the MOT coils. They provide about 12 G/cm at the center when separated in distance by 4 cm and 2 A of current is provided.

small ones in the figure, the inner radius is 1.5 cm. There are 7 turns per layer and 7 layers. For the distance between the coils of 4 cm, and current of 2 A, we are able to
Figure 5.6. Magnetic field test conducted on the coils to make sure that
the coils behave according to the theoretical simulations conducted before
building them. The left plots are of the MOT coils and the right are of the
bias coils. In the top plots, the distance between the coils was varied while
current stayed fixed. In the bottom plots, the distance was fixed while the
current was varied.

achieve magnetic field gradient of 12 G in the center. For the bias coils, which are the
large ones in the figure, the inner radius is 2.8 cm. There are also 7 turns per layer and
7 layers. For the distance between the coils of 6 cm, and current of 0.5 A, we are able to
achieve magnetic field of over 2.6 G in the center. The switching circuits for the coils are
depicted in Fig. 5.7 and the actual picture of the switch for the bias coils is in Fig. 5.8
When we were working with the second metal vacuum chamber, Dr. Frank Narducci informed us of one experimental setback in working with metal chambers - that the magnetic field of the MOT takes a long time to die down, and if we perform the Raman Rabi pulse before it dies down by twice the decay time of the MOT field, we will not be able to see a clear Raman signal. He taught us a quick way to test out the duration of the residual field of the MOT: First, take a piece of magnetic wire, and wind it about 5 times around the center of the chamber. They should be wound in the same direction as one of the MOT coils (it should not be perpendicular). Take the two ends and file them off. Then take a BNC cable and cut off one end, exposing the signal end and the ground. Join one end of the magnetic wire to the signal end of the BNC cable, and another end of the wire to the ground of the BNC cable. Next, connect the BNC cable to an oscilloscope.
Connect the MOT coils to a signal generator to turn the current on and off at around 10 Hz. Connect the signal generator to the scope and use it as a trigger. See how long it takes for the voltage in the magnetic wire to die out. The voltage corresponds to the current present in the wire, which corresponds to the induced magnetic field still present in the chamber. As can be seen in Fig. 5.9, when we ran the above test on our chamber, we found that it takes around 10 ms for the voltage to die down to about 1/3 of its original value when the current through MOT coils was turned off. It took about 30 ms for the field to die off completely. Hence, in order to ensure that we get a clean signal, about 60 ms of waiting time after the MOT magnetic fields are turned off in order to proceed with
the Raman pulse. However, it is very difficult to keep the atoms around for that long with simply the optical molasses.

This was the single most important reasoning behind our migration to a glass cell system. With a glass cell, the signal dies out a lot quicker than what is seen here. In fact, a test with the coils revealed that the 30 ms decay time shorted to about 5 ms, so that 10 ms was all that was needed before the Raman beams could be turned on. The next section covers all the transitions we made over the years.

![Figure 5.9](image.png)

Figure 5.9. Magnetic field decay test performed with the MOT coils in the metal chamber, before we migrated to the glass cell. It can be seen that, even with lower current, the decay time does not shorten all that much; it remains at around 30 ms. The eddy current in the metal chamber is to blame.
5.5. Improvements

Figure 5.10. The first vacuum chamber that we ran experiments on in the lab.

The first metal chamber that we began working with in the lab was extremely large and covered most of one optics table. In Fig. 5.10, the chamber without its original chirp slowing arm is shown. The feedthroughs at the top of the main experimental chamber are there to supply current to the MOT coils inside, as well as to supply water to cool them. The ion pump in the picture is VacIon Plus 300 and pumps 300 l/s. The roughing pumps were two diffusion pumps and mechanical pumps. The chirp slowing arm had a rubidium oven that was heated up with a Variac and went through collimators and a cold finger (cooled with liquid nitrogen). After the arm was taken out, we added a large viewport, and added a feedthrough with getters on another part of the chamber. Once we got the MOT going, we did an atom launch experiment for an atomic fountain. Although we were successful, we decided that a smaller system was desirable for many reasons and stopped work in this chamber.
Figure 5.11. The second vacuum chamber that required a smaller, 150 l/s ion pump and had the MOT coils fit outside the chamber.

Figure 5.12. Another view of the second vacuum chamber

The next chamber was another metal chamber that was initially built as a test chamber. We performed single Raman pulse test after Renpeng Fang and Daniel Villalon built the Labview program to take data from scratch, and we figured out the pulse sequence. For this test, the magneto optic trap collected cold atoms for 1.9 s. Then the MOT coils
are off while the repump and MOT beams are still on and optical molasses keep atoms in place for 10 ms. During this period, the alignment of the beams is very important to keep as many atoms as in place as possible. Then we start the optical pumping cycle. The repump beam is turned off to initialize the atoms to \( |F = 2\rangle \). Because the MOT beams are detuned by 110 MHz from \( |F = 2\rangle \) to \( |F' = 3\rangle \) transition, the optical pumping takes around 1 ms, unless the Rabi frequency is very large. Then we turn off the MOT beam and the bias coils are turned on. (The bias coils may need to be turned on before this point since they may not switch on fast enough, but it turns out that you can still get a good result without the Raman coils by just using the Earths magnetic field.) For the Raman beams of about 28 mW between them, the \( \pi \) pulse period was about 4 µs. The results are plotted in Fig. 5.13 for a single run, and in Fig. 5.14 for averaged data, with 10 averages per point, for two different bias coil strengths. Even for averaged data, the results were extremely noisy, and the various transitions were hardly resolved. We were certain at this point that the residual field from the MOT coils were to blame.

![Figure 5.13. Single run result of single Raman pulse experiment in the MOT chamber depicted in Fig. 5.11.](image)
In an effort to get a clean signal, we felt inclined to scrap the vacuum chamber and start anew in a glass cell. The first glass chamber, we struggled with for several months due to our working with faulty flanges and our inexperience working with 1.33” flanges; we learned that we may not use as much torque to tighten the bolts as on the 2.75” flanges. The bolts gave very easily, and we went through dozens of bolts whose heads we broke off. Another unfortunate incident was the demise of a viewport that we did not protect enough from the baking process. Finally, when we thought the vacuum was good, we managed to burn off the rubidium in the getters (which, by the way, came preinstalled in the glass chamber made by Precision Glass Blowing of Colorado, with the getters provided to them by SAES Getters).

Fortunately, Selim had another vacuum system, already assembled with getters and a glass cell that he meant to use for another experiment, that was not being used at the
Figure 5.15. The final vacuum chamber that we ended up with, which is a 1 cm by 1 cm by 4 cm experimental chamber with a turbo pump and a 2 ml/s Gamma Vacuum ion pump. The feedthroughs are attached in series to rubidium getters inside the chamber.

Figure 5.16. The glass cell vacuum system with the coils and the optics around it.

moment (See Fig. 5.15). It was already pumped down, and a light baking and cleaning the ion pump (by running current through it without the magnets) was able to restore
the vacuum pressure to low $10^{-11}$ Torr. We chose to keep the turbo pump in place for the rest of the experiment.

### 5.6. Results of single zone and Ramsey fringe test

In Fig. 5.18 and 5.19, two more pictures of the actual setup show where the optics and other equipment are laid out. The MOT beam is brought over by an optical fiber, and the repump, simply by using mirrors. Once they reach the vacuum chamber area, the optics guiding them into the chamber from 6 different paths are all placed within a 20” × 30” area. Despite the limited space and awkward balancing of power for the six beams (I should have made sure that each waveplate was responsible for a single coordinate), and dealing with non-AR coated glass cell that made viewing the inside very difficult because of reflections from every surface, we were able to see the MOT within two weeks of working with the new glass chamber. The MOT can be seen in Fig. 5.20. At this instance in time, the MOT was fairly large; more than 1 mm by 1 mm. We were able to optimize
Figure 5.18. A view of the optics table from far away to show the setup from far away. The turbo pump, camera, and MOT optics are labeled.

Figure 5.19. The paths of the MOT beams and the repump beams. The Raman beam combines with the MOT beam that goes up, but is not in place yet.
the MOT by wiggling the coils around, balancing the power in the MOT beams, turning
the quarter waveplates, and, most importantly, by watching the pressure. With such a
small vacuum system, it was not difficult to accidentally make the MOT unstable by the
influx of rubidium background vapor. We found that turning the current up to 3 A in the
beginning of the day, and then slowly tuning it down during the day a little, was ideal for
our system.

Once the MOT was obtained, we performed a single zone Raman interaction test. The
experimental pulse sequence is given in Fig. 5.21. The result of the Ramsey fringe test
in Fig. 5.22 shows that the \( \pi \) pulse time is 25 \( \mu s \), which we set the subsequent Raman
pulse width to. The result of the actual Single zone off-resonant Raman interaction test
is given in Fig. 5.23 with ten averages per data point. All eleven peaks that we would
expect from arbitrary magnetic field are clearly visible with good signal to noise ratio. In
5.2. Collective state detection

In the previous section, where preliminary results were collected, the detection scheme employed was not conducive for detecting a single collective state. This requires signal processing beyond an APD and a Labview program.
Figure 5.22. Result from Raman Rabi oscillation test by varying the Raman pulse width (while keeping the total duration from the Raman pulse to the detection time constant). The credit for taking this data goes to Renpeng Fang.

In the detection zone, we probe the population in the most excited state, $|E_N\rangle$, where all the individual atoms are in state $|b\rangle$. This can be done by exciting the atoms with an off-resonant beam of laser with frequency detuned from $|a\rangle$ to the excited states. Due to Raman Rabi condition, atoms in the excited states will then transition to $|b\rangle$ while emitting a photon that matches that off-resonant transition frequency. Only in the case where none of the atoms are in state $|a\rangle$, which is when the collective state of the system is $|E_N\rangle$, will there be no photon emission. Therefore, counting the number of occurrences where no photons are detected and building a histogram allows us to plot the $|E_N\rangle$ state signal.
Figure 5.23. Result from single Raman pulse test. The credit for taking this data goes to Renpeng Fang.

The electric field of the emitted photon can be calculated from $\epsilon_0 E^2 A c \tau / 2 = \hbar \omega_2$ where $A$ is the area, $c$ the speed of light, and $\tau$ the interaction time, to form the field interaction volume. The single photon Rabi frequency is proportional to the electric field through the expression $g = |e \cdot \langle r \rangle| E / \hbar$ where $|e \cdot \langle r \rangle|$ is the dipole moment of the atom. The Raman Rabi frequency for a single atom is $\Omega = g \Omega_0 / (2 \Delta)$ where $\Delta$ is the average detuning of the two laser frequencies from the transition frequencies. In the bad cavity limit, $\Omega \gg k$ where $k$ is the cavity decay rate, the atoms will not reabsorb the photon that it just emitted. Therefore, the single excitation of one leg of the lambda system will drive the transition to the other state in a nonreversible process that can be modeled as a decay with an effective decay rate that is singular to each $|E_j\rangle$ state. Duan, Lukin, Cirac,
and Zoller have derived this quantity for the decay from the first state as:

$$
(5.8)\quad \gamma_0 = \frac{4NL|g\Omega_0|^2}{\Delta^2c} = N\gamma_{sa}
$$

where $\gamma_{sa} = 16L\Omega^2/c$. The other excited states effective decay rates can also be calculated following the same logic:

$$
(5.9)\quad \gamma_j = (j + 1)(N - j)\gamma_{sa}
$$
The slowest decay rate is \( \gamma_0 = \gamma_{N-1} = N\gamma_{sa} \). To make sure that even the longest lived state has a chance to decay completely, the interrogation period has to be at least \( \gamma_0 T = \pi \). Then we collect the florescence from the anti-Stokes photons in an efficient method similar to that of Coherent anti-Stokes Raman scattering spectroscopy (CARS). Once the florescence is collected, we multiply the signal by double the frequency of the 1.5 GHz voltage controlled oscillator (VCO), which drives the two acousto optic modulators that detune the lasers from resonance. After filtering the signal with a low pass filter (LPF), the voltage output is plotted. Determining the threshold at which photon emission occurs, and counting the occurrences of zero photon emission contributes a point on the histogram. In this manner, we will be able to detect the collective state \( |E_N\rangle \) and produce the first collective state atomic clock. More on this is in Chapter 3 in the detection section.

Due to our need to purchase all new signal processing equipment, we have yet to perform the collective state detection test. This is, of course, the next thing on our list of to dos.
CHAPTER 6

Conclusion

The two major portions of my thesis work are that of the Rydberg assisted light shift imbalance induced blockade, and that of the collective state atomic clock. With the former, we showed that the ensemble-based qubits realized in the manner we described can be used to implement a controlled-NOT (CNOT) gate. Many such gates can be linked to one another, via nearest neighbor quantum coupling, to realize an elementary quantum computer. Due to our system’s relative insensitivity to the number of atoms held in a FORT, our approach may prove to be a more viable alternative for scalable quantum computing using neutral atoms. With the latter, we found that an atomic clock, given the same dark time period, could have a significant reduction in the Ramsey fringe linewidth, by a factor of $\sqrt{N}$, by measuring the amplitude of a collective state with a heterodyne detection scheme. We have shown that the reduction occurs due to multipath interference among the collective states, and does not violate the fundamental quantum limit. The performance of the COSAC has been compared to that of the CC by analyzing quantum and classical fluctuations in frequency. When the effects of detector efficiency and collection efficiency are considered, it can be seen that the COSAC may perform 10 times better than a typical CC employing fluorescence detection.

Currently, we are still working on detecting a collective state signal, but we believe that we are very close, given our recent progress that shows good signal to noise ratio in our system. We are transitioning to a frequency synthesizer with smaller frequency step
and better accuracy, and a new timing board and a more sophisticated Labview program and timing operations. Most importantly, we are gathering the electronics necessary to perform the null measurement scheme that is so key to our experiment. While it was not easy to come this far, we believe that we are very close to obtaining our results and publishing soon. What we have accomplished thus far is by no means trivial, and we have accrued knowledge about collective states of atoms theoretically, and with experimental realization of their detection, that we would not have without going on this journey.
References


APPENDIX A

Programs for Rydberg assisted light shift imbalance induced blockade

Although more than ten different codes exist for calculating various conditions for the light shift blockade, these are the basic programs that I started with in order to build the other ones.

A.1. MATLAB code for solving $[H,\rho]$ part of the density matrix, called by the other programs - for a single atom

% This function finds the M matrix for calculating the evolution
% of the density matrix. It takes three parameters: H, the
% Hamiltonian, S, the source matrix, and decay, the level from
% which the decay occurs. For example, if the decay occurs from
% the 3rd level, then decay = 3.

function M = find_M(H, S, decay)
% return the size of the Hamiltonian in one of the dimensions
N = size(H,1);

% initialize the M matrix
M = zeros(N^2, N^2);
for q = 1:N
    for r = 1:N
        % indices for H*rho part
        i_1 = 1 + (q-1)*N;
        i_2 = 1 + (r-1)*N;

        % indices for rho*conj_transp(H) part
        i_3 = q;
        i_4 = r;

        for s = 1:N
            % add the terms for H*rho part
            M(i_1, i_2) = M(i_1, i_2) - 1i*H(q, r);
            i_1 = i_1 + 1;
            i_2 = i_2 + 1;

            % add the terms for rho*conj_transp(H) part
            M(i_3, i_4) = M(i_3, i_4) + 1i*conj(H(q, r));
            i_3 = i_3 + N;
            i_4 = i_4 + N;
        end
    end

% include the source terms here:
M(q + (q-1)*N, decay + (decay-1)*N) = M(q + (q-1)*N, decay +
A.2. MATLAB code for solving $[H,\rho]$ part of the density matrix, called by the other programs - for an ensemble of atoms

```matlab
function M = find_M_dicke(H, S1, S2, S3, decay1, decay2, decay3)

N = size(H,1);

M = zeros(N^2, N^2);

for q = 1:N
    for r = 1:N
        % indices for H*rho part
        i_1 = 1 + (q-1)*N;
        i_2 = 1 + (r-1)*N;

        % indices for rho*conj_transp(H) part
```

$$(\text{decay - 1}) \times N + S(q,q);$$
\[ i_3 = q; \]
\[ i_4 = r; \]

for \( s = 1:N \)

\[
\text{\% add the terms for } H\text{\textdagger} \text{rho part}
\]
\[
M(i_1, i_2) = M(i_1, i_2) - 1i*H(q, r);
\]
\[
i_1 = i_1 + 1;
\]
\[
i_2 = i_2 + 1;
\]

\[
\text{\% add the terms for } \text{rho*conj\textunderscore transp}(H) \text{ part}
\]
\[
M(i_3, i_4) = M(i_3, i_4) + 1i*\text{conj}(H(q, r));
\]
\[
i_3 = i_3 + N;
\]
\[
i_4 = i_4 + N;
\]

end

end

\[
\text{\% include the source terms here:}
\]
\[
M(q + (q-1)*N, \text{decay1} + (\text{decay1}-1)*N) = M(q + (q-1)*N, \text{decay1} + (\text{decay1}-1)*N) + S1(q, q);
\]
\[
M(q + (q-1)*N, \text{decay2} + (\text{decay2}-1)*N) = M(q + (q-1)*N, \text{decay2} + (\text{decay2}-1)*N) + S2(q, q);
\]
\[
M(q + (q-1)*N, \text{decay3} + (\text{decay3}-1)*N) = M(q + (q-1)*N, \text{decay3} + (\text{decay3}-1)*N) + S3(q, q);
\]

end
A.3. Matlab code for the steady state solution of Λ type three level system, with ability to calculate it for N atoms

% Program name: steady_state_3.m
% Author: May Kim
% Date created: March 12, 2013
%
% This program plots the steady state solution of the state
% of interest by solving the evolution of the density matrix
% with the given Hamiltonian while varying delta = dela - delb,
% the difference in the two detunings. The output of the program
% is the plot of the population of the state of interest with
% respect to delta, and also the value of delta for which the
% maximum value of the population occurs. Using this information,
% the user can then change the delta and the step to achieve
% better resolution next time around.
%
% Instructions for use:
% 1. Set the frequency parameters: lines 28 - 31
% 2. Set delta (start with 500 initially): line 36
% 3. Set the step size (start with 1): line 43
% 4. Set the source matrix: lines 47 - 49
% 5. Set the level from which decay occurs: line 52
% 6. Set the level that you want plotted: line 55
% 7. Set the Hamiltonian: lines 72 - 74
clear all;

% USER SETTINGS

% USER: enter the Rabi frequencies and detuning in units of gamma
oma = 20;                 % Rabi frequency 1
omb = 20;                 % Rabi frequency 2
del = 0;                  % average of the two detunings del = dela + delb
)
gamma = 1;                % natural linewidth (set small to minimize
effect)

omc = 0;
omb = 0;
er = 0;
delr = 0;

% USER: enter the difference in the two detunings delta = dela - delb in
% units of gamma
%(start with 500 initially; the program output will tell you how to change
it)
delta = 0;

% USER: initialize the step size and the total number of steps wanted
% step = 1 initially
% step = 0.0000001 is the best accuracy that can be had
% step_tot = 1000 shouldn't be changed; if it needs to be changed, only
% change once in the beginning.
step = 1; % step size to vary delta by
step_tot = 1000; % total number of steps taken

% USER: enter the source matrix (leave out rho33)
S = [gamma/2 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 gamma/2 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ;
     0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 ];

% USER: enter the level from which the decay occurs
decay = 2;
% USER: enter the level for which you want to see the effect of the detuning
level = 3;

% return the size of the source matrix in one of the dimensions
N = size(S,1);

% the plot dimensions
dx = linspace(delta-(step_tot*step/2), delta+((step_tot)*step/2), step_tot);
d_y = zeros(1,step_tot);

% initialize the data arrays (for results)
d = delta - (step_tot*step/2);

for l = 1:step_tot

    % USER: enter the Hamiltonian
    H = [delta sqrt(2)*oma/2 0 0 0 0
         0 0 0 0 0 0
         0 0 0 0 0 0
         0 0 0 0 0 0];
\[
\begin{align*}
\sqrt{2} \cdot \text{oma}/2 - \text{del} + \text{delta}/2 & \quad \text{omb}/2 & \quad 0 & \quad 0 \\
\sqrt{2} \cdot \text{oma}/2 & \quad 0 & \quad 0 & \quad 0 & \quad 0 & \quad 0; \\
0 & \quad 0 & \quad 0 & \quad 0 & \quad 0 & \quad 0; \\
0 & \quad \text{omb}/2 & \quad 0 & \quad \text{oma}/2 & \quad 0 \\
0 & \quad 0 & \quad \text{omc}/2 & \quad 0 & \quad 0 & \quad 0; \\
0 & \quad 0 & \quad 0 & \quad 0 & \quad 0 & \quad 0; \\
0 & \quad \text{oma}/2 & \quad - (\text{del} + \text{delta}/2) \quad \sqrt{2} \cdot \text{omb}/2 \\
\sqrt{2} \cdot \text{oma}/2 & \quad 0 & \quad \text{omc}/2 & \quad 0 & \quad 0 & \quad 0; \\
0 & \quad 0 & \quad 0 & \quad 0 & \quad \sqrt{2} \cdot \text{omb}/2 & \quad - \text{delta} & \quad 0 & \quad 0; \\
0 & \quad \text{omc}/2 & \quad 0 & \quad 0 & \quad 0; \\
0 & \quad 0 & \quad 0 & \quad 0 & \quad 0; \\
0 & \quad \text{omd}/2 & \quad 0 & \quad 0 & \quad 0; \\
0 & \quad 0 & \quad 0 & \quad \text{omc}/2 & \quad 0 \\
0 & \quad 0 & \quad \text{oma}/2 & \quad - (\text{delr} + \text{del} + \text{delta}/2) \quad \text{omb}/2 & \quad 0 & \quad 0; \\
0 & \quad \text{omd}/2 & \quad 0 & \quad 0 & \quad 0; \\
\end{align*}
\]
\[
0 \quad 0 \quad 0 \quad 0 \quad \sqrt{2} \cdot \frac{omc}{2}
\]
\[
0 \quad 0 \quad \frac{omb}{2} \quad -\left(\delta l + \delta t a\right) \quad 0
\]
\[
0 \quad \frac{omd}{2} \quad \sqrt{2} \cdot \frac{omc}{2} \quad 0 \quad 0 \quad 0
\]
\[
0 \quad 0 \quad 0 \quad 0 \quad 0 \quad 0
\]
\[
0 \quad \frac{oma}{2} \quad 0 \quad 0 \quad 0 \quad 0
\]
\[
0 \quad 0 \quad 0 \quad 0 \quad 0 \quad 0
\]
\[
0 \quad 0 \quad \frac{omd}{2} \quad 0 \quad 0 \quad 0
\]
\[
\frac{oma}{2} - \delta l \quad \frac{omb}{2} \quad 0 \quad 0 \quad 0
\]
\[
0 \quad 0 \quad 0 \quad 0 \quad 0 \quad 0
\]
\[
0 \quad \frac{omb}{2} - \delta l a \quad 0 \quad \frac{omc}{2} \quad 0 \quad 0
\]
\[
0 \quad 0 \quad 0 \quad 0 \quad \sqrt{2} \cdot \frac{omc}{2} \quad 0 \quad 0 \quad 0
\]
\[
0 \quad 0 \quad 0 \quad 0 \quad 0 \quad 0 \quad 0 \quad 0
\]
\[
0 \quad 0 \quad \frac{omd}{2} \quad \sqrt{2} \cdot \frac{omd}{2} \quad 0 \quad 0 \quad 0 \quad 0
\]
\[
0 \quad 0 \quad \frac{omc}{2} \quad \sqrt{2} \cdot \frac{omd}{2} \quad -3 \cdot \frac{\delta l a}{2} - \delta l r \quad \sqrt{2} \cdot \frac{omc}{2} \quad 0 \quad 0
\]
\[
\begin{pmatrix}
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \sqrt{2} \ast \text{omd/2} & -(del+er)
\end{pmatrix}
\]

% call the function findM(H, S, decay) to find the M matrix
M = findM(H, S, decay);

% eliminate the last row of elements in the M matrix (redundant)
% add the equation in which all the rhos to sum up to 1
count = 1;
for m = 1:N^2
    if m == count
        M(N^2,m) = 1;
        count = count + N + 1;
    else
        M(N^2,m) = 0;
    end
end

% y = M*x and is all zero in steady state except for the last element
% which is 1 (which the population adds up to)
y = zeros(N^2,1);
y(N^2) = 1;
% solve x = (M^-1)*y
x = linsolve(M,y);

% pick out the element that's wanted and save it
d.y(1,1) = x(level + (level-1)*N);

% increment delta by step size for the next iteration
d = d + step;
end

% find the maximum value of the population, its index, and the detuning
% that gets you there.
[d_max, d_max_i] = max(d.y);
d.best = delta-(steptot*step/2) + (d_max_i-1)*step;

% plot the population in steady state solution versus delta
figure
plot(d.x, real(d.y))
format long g;
fprintf('The maximum exited state population in the steady state solution
     is %d.\n', d_max)
fprintf('The maximum occurs when the two photon detuning Delta = dela -
     delb is %d (accurate to %d).\n', d_best, step)
A.4. Matlab code for time evolution of Λ type three level system, with ability to calculate it for \( N \) atoms

```
% % Program name: evolution_3.m
% % Author: May Kim
% % Date created: March 12, 2013
%
% This program plots the evolution of the population according to the 3 level Hamiltonian specified (for 3 pi rotation).
%
% Instructions for use:
% 1. Set the parameters: lines 20 – 24
% 2. Set the Hamiltonian: lines 27 – 29
% 3. Set the source matrix: lines 32 – 34
% 4. Set the level from which the decay occurs: line 37
% 5. Set the number of points to plot: line 42

clear all;

%%%%%%%%%%%%%%%% USER SETTINGS %%%%%%%%%%%%%%%%%

% USER: enter the Rabi frequencies and detuning in units of gamma
oma = 0.01; % Rabi frequency 1
omb = 100; % Rabi frequency 2
del = 1000; % average of the two detunings del = dela + delb)
delta = 2.497858; % difference in the two detunings
gamma = .1; % natural linewidth (set small to minimize effect)

% USER: enter the Hamiltonian
H = [delta/2 oma/2 0;
    oma/2 -del-1i*gamma/2 omb/2;
    0 omb/2 -delta/2];

% USER: enter the source matrix (leave out rho_33)
S = [gamma/2 0 0;
      0 0 0;
      0 0 gamma/2];

% USER: enter the level from which the decay occurs
decay = 2;

% USER: enter the number of points you want to calculate between the points % on the plot (the larger, the better, but the computation time will % increase significantly)
np = 1*10^9;
\% return the size of the Hamiltonian in one of the dimensions
N = size(H,1);

\% call the function find\_M(H, S, decay) to find the M matrix
M = find\_M(H, S, decay);

\% find the time for which a pi rotation occurs (Raman freq * tau = pi)
tau = 2*del*pi/oma/omb;

\% set the x axis of the plot to allow 3 pi rotations
\tt = linspace(0,5*round(tau),(5*round(tau)+1));

\% set the dt for calculating the evolution
dt = max(tt)/((length(tt)-1)*np);

\% initialize the arrays and matrices for calculating the evolution
tic
A = zeros(N^2,1);
A(1) = 1;
One1 = diag(ones(1,N^2));
Atxt = zeros(N^2,length(tt));

for m = 1:length(tt)
Atxt(:,m) = (M*dt+One1)^(tt(m)/dt+1)*A;

end
toc

% PLOT

figure
for k=1:N
    subplot(N,1,k), plot(tt,real(Atxt((k-1)*N+k,:)))
end

% PLOT FOR MORE THAN ONE ATOM

% ens = 1; % number of ensembles
% n = 3; % number of atoms
%
% c = zeros(ens, length(tt));
%
% mult = factorial(n)/(factorial(n/3)^3);
%
% for m = 1:length(tt)
%     c(1,m) = Atxt(1,m)^n;
%     c(2,m) = n*Atxt(5,m)*Atxt(1,m)^(n-1);
%     c(3,m) = n*Atxt(9,m)*Atxt(1,m)^(n-1);
%     c(4,m) = n*(n-1)*Atxt(5,m)*Atxt(9,m)*Atxt(1,m)^(n-2);
c(5,m) = .5 * n * (n-1) * Atxt(9,m) * 2 * Atxt(1,m) * (n-2);  
c(6,m) = .5 * n * (n-1) * (n-2) * Atxt(9,m) * 2 * Atxt(5,m) * Atxt(1,m) * (n-3);  
c(7,m) = mult * Atxt(1,m) * (n/3) * Atxt(5,m) * (n/3) * Atxt(9,m) * (n/3);  
end  
figure  
for k=1:ens  
    subplot(ens,1,k), plot(tt, real(c(k,:)))  
end
APPENDIX B

Programs for collective state atomic clock

B.1. Python code using Monty Python type simulation that calculates noise that occurs in a collective state atomic clock (COLA) as a function of the number of trials, and the number of atoms

```python
import math as m
import numpy as np
import matplotlib.pyplot as plt
import random2 as r

def monte(a):
    rand = r.randint(0,100)
    if rand < a*100.:
        return 1
    else:
        return 0

# function signal(x, num, run): for a given range (x), number of atoms (num)
, and
```
def signal(x, num, run):
    y = np.zeros(len(x))
    for i in range(len(x)):
        y[i] = run*(np.cos(x[i]))**(2*num)
    return y

# function signal(x, num, run): for a given range (x), number of atoms (num)
# and number of trials (run), generates a random number between num ± sqrt of num
# (r_num) then builds the histogram for a given number of trials (run) to return
# y = (cos x)^(2 r_num)

def ran_sig(x, num, run):
    y = np.zeros(len(x))
    for j in range(run):
        for i in range(len(x)):
            r_num = np.random.randint(round(num - np.sqrt(num)), np.int(num + np.sqrt(num)))
            y[i] = y[i] + monte((np.cos(x[i]))**(2*r_num))
    return y

# function ran_sig_ave(x, num, r_ave): for a given range (x), number of atoms

# (num), and number to average by (r_ave), finds a random number between
# r_num = num+sqrt(num) and num-sqrt(num) and returns the function
# y = (cos x)^\(2 \cdot r\_num\) averaged by r_ave times.

```python
def ran_sig_ave(x, num, r_ave):
    y = np.zeros(len(x))
    for i in range(len(x)):
        r_num = 0
        for j in range(r_ave):
            r_num = r_num + r.randint(round(num - m.sqrt(num),0), int(num + m.sqrt(num)))
        r_num = r_num/r_ave
        y[i] = (m.cos(x[i]))**\(2 \cdot r\_num\)
    return y
```

# parameters:

```python
num = 10**7  # number of atoms
rng = m.pi/m.sqrt(num)/2
dot = rng/1000.
```

```python
x = np.arange(-rng, rng, dot)  # x range
run = 100  # number of experimental runs
```

```python
y1 = ran_sig(x, num, run)
y2 = signal(x,num + m.sqrt(num), run)
```
y3 = signal(x, num - m.sqrt(num), run)

plt.plot(x, y1)
plt.plot(x, y2)
plt.plot(x, y3)
plt.show()

B.2. Python program for generating a random number, used by the code in the previous section (taken off the Internet; source listed in the code)

"""Random variable generators."

integers
    uniform within range

sequences
    pick random element
    pick random sample
generate random permutation

distributions on the real line:
    uniform
    triangular
normal (Gaussian)
lognormal
negative exponential
gamma
beta
pareto
Weibull

distributions on the circle (angles 0 to 2pi)

circular uniform
von Mises

General notes on the underlying Mersenne Twister core generator:

* The period is 2**19937−1.
* It is one of the most extensively tested generators in existence.
* Without a direct way to compute N steps forward, the semantics of jumpahead(n) are weakened to simply jump to another distant state and rely on the large period to avoid overlapping sequences.
* The random() method is implemented in C, executes in a single Python step, and is, therefore, threadsafe.
from __future__ import division, print_function
import sys
from warnings import warn as _warn
from types import MethodType as _MethodType, BuiltinMethodType as _BuiltinMethodType
from math import log as _log, exp as _exp, pi as _pi, e as _e, ceil as _ceil
from math import sqrt as _sqrt, acos as _acos, cos as _cos, sin as _sin
from os import urandom as _urandom
from binascii import hexlify as _hexlify
import hashlib as _hashlib
PY3 = sys.version_info[0] >= 3
try:
    long
except NameError:
    long = int
try:
    xrange
except NameError:
    xrange = range
```
__all__ = ["Random", "seed", "random", "uniform", "randint", "choice", "sample",
"randrange", "shuffle", "normalvariate", "lognormvariate",
"expovariate", "vonnivesvariate", "gammavariate", "triangular",
"gauss", "beta_ardiate", "_eratevariate", "_eibullv_arianate",
"getstate", "setstate", "jumpahead", "WichmannHill", "getrandbits",
"SystemRandom"]

NV_MAGICCONST = 4 * exp(-0.5)/sqrt(2.0)
TWOPI = 2.0*pi
LOG4 = log(4.0)
SG_MAGICCONST = 1.0 + log(4.5)
BPF = 53  # Number of bits in a float
RECIP_BPF = 2**-BPF

# Translated by Guido van Rossum from C source provided by
# Adrian Baddeley. Adapted by Raymond Hettinger for use with
# the Mersenne Twister and os.urandom() core generators.

import _random

class Random(_random.Random):
    
    """Random number generator base class used by bound module functions."
```
Used to instantiate instances of Random to get generators that don’t share state. Especially useful for multi-threaded programs, creating a different instance of Random for each thread, and using the jumpahead() method to ensure that the generated sequences seen by each thread don’t overlap.

Class Random can also be subclassed if you want to use a different basic generator of your own devising: in that case, override the following methods: random(), seed(), getstate(), setstate() and jumpahead(). Optionally, implement a getrandbits() method so that randrange() can cover arbitrarily large ranges.

""

VERSION = 3  # used by getstate/setstate

def __init__(self, x=None):
    """Initialize an instance.

    Optional argument x controls seeding, as for Random.seed()."
    """
self.seed(x)
self.gauss_next = None

def seed(self, a=None):
    
    """Initialize internal state from hashable object.

    None or no argument seeds from current time or from an operating system specific randomness source if available.

    If a is not None or an int or long, hash(a) is used instead.
    """

    if a is None:
        try:
            a = long(hexlify(urandom(16)), 16)
        except NotImplementedError:
            import time
            a = long(time.time() * 256) # use fractional seconds

        super(Random, self).seed(a)
        self.gauss_next = None

    def getstate(self):
        """Return internal state; can be passed to setstate() later."""
return self.VERSION, super(Random, self).getstate(), self.

gauss_next

def setstate(self, state):
    """Restore internal state from object returned by getstate()."""
    version = state[0]
    if version == 3:
        version, internalstate, self.gauss_next = state
        super(Random, self).setstate(internalstate)
    elif version == 2:
        version, internalstate, self.gauss_next = state
        # In version 2, the state was saved as signed ints, which
        # causes
        # inconsistencies between 32/64-bit systems. The state is
        # really unsigned 32-bit ints, so we convert negative ints
        # from
        # version 2 to positive longs for version 3.
        try:
            internalstate = tuple(long(x) % (2**32) for x in
                                    internalstate)
        except ValueError as e:
            raise TypeError(e)
        super(Random, self).setstate(internalstate)
    else:
        raise ValueError("state with version %s passed to ")
"Random.setstate() of version %s" % (version, self.VERSION)

def jumpahead(self, n):
    """Change the internal state to one that is likely far away from the current state. This method will not be in Py3.x, so it is better to simply reseed."
    ""
    # The super.jumpahead() method uses shuffling to change state, # so it needs a large and "interesting" n to work with. Here, # we use hashing to create a large n for the shuffle.
    s = repr(n) + repr(self.getstate())
    n = int(_hashlib.new('sha512', s.encode()).hexdigest(), 16)
    if PY3:
        raise NotImplementedError('jumpahead')
    super(Random, self).jumpahead(n)

    # Methods below this point do not need to be overridden when # subclassing for the purpose of using a different core generator.

## pickle support

    def __getstate__(self): # for pickle
        return self.getstate()
def _setstate__(self, state):  # for pickle
    self.setstate(state)

def _reduce__(self):
    return self._class_, (), self.getstate()

### integer methods

def randrange(self, start, stop=None, step=1, int=int, default=None, maxwidth=1<<BPF):
    """Choose a random item from range(start, stop[, step]).

    This fixes the problem with randint() which includes the endpoint; in Python this is usually not what you want. Do not supply the 'int', 'default', and 'maxwidth' arguments."
    
    # This code is a bit messy to make it fast for the common case while still doing adequate error checking.
    istart = int(start)
    if istart != start:
        raise ValueError("non-integer arg 1 for randrange()")
    if stop is default:
        if istart > 0:
            if istart >= maxwidth:
    return self._randbelow(istart)
    return int(self.random() * istart)
    raise ValueError("empty range for randrange()")

    # stop argument supplied.
    istop = int(stop)
    if istop != stop:
        raise ValueError("non-integer stop for randrange()")
    width = istop - istart
    if step == 1 and width > 0:
        # Note that
        #     int(istart + self.random() * width)
        # instead would be incorrect. For example, consider istart
        # = -2 and istop = 0. Then the guts would be in
        # -2.0 to 0.0 exclusive on both ends (ignoring that random()
        # might return 0.0), and because int() truncates toward 0, the
        # final result would be -1 or 0 (instead of -2 or -1).
        #     istart + int(self.random() * width)
        # would also be incorrect, for a subtler reason: the RHS
        # can return a long, and then randrange() would also return
        # a long, but we're supposed to return an int (for backward
        # compatibility).

        if width >= maxwidth:
            return int(istart + self._randbelow(width))
```python
return int(istart + int(self.random() * width))

if step == 1:
    raise ValueError("empty range for randrange() (%d,%d, %d)" % (istart, istop, width))

# Non-unit step argument supplied.
istep = int(step)
if istep != step:
    raise ValueError("non-integer step for randrange()")

if istep > 0:
    n = (width + istep - 1) // istep
elif istep < 0:
    n = (width + istep + 1) // istep
else:
    raise ValueError("zero step for randrange()")

if n <= 0:
    raise ValueError("empty range for randrange()")

if n >= maxwidth:
    return istart + istep*self._randbelow(n)
return istart + istep*int(self.random() * n)

def randint(self, a, b):
```
"""Return random integer in range \([a, b]\), including both end points.
"""

return self.randrange(a, b+1)

def _randbelow(self, n, _log=_log, int=int, _maxwidth=1<<BPF,
               _Method=_MethodType, _BuiltinMethod=_BuiltinMethodType):
    """Return a random int in the range \([0, n)\)

Handles the case where \(n\) has more bits than returned by a single call to the underlying generator.
"""

try:
    getrandbits = self.getrandbits
except AttributeError:
    pass
else:
    # Only call self.getrandbits if the original random() builtin method
    # has not been overridden or if a new getrandbits() was supplied.
    # This assures that the two methods correspond.
if type(self.random) is _BuiltinMethod or type(getrandbits) is _Method:
    k = int(1.00001 + _log(n−1, 2.0))  # 2**k > n−1 > 2**(k−2)
    r = getrandbits(k)
    while r >= n:
        r = getrandbits(k)
    return r

if n >= _maxwidth:
    _warn("Underlying random() generator does not supply \n" "enough bits to choose from a population range this large")
    return int(self.random() * n)

### sequence methods

def choice(self, seq):
    """Choose a random element from a non-empty sequence."""
    return seq[int(self.random() * len(seq))]  # raises IndexError if seq is empty

def shuffle(self, x, random=None, int=int):
    """x, random=random.random -> shuffle list x in place; return None.

    Optional arg random is a 0-argument function returning a random float in [0.0, 1.0); by default, the standard random.random."""
if random is None:
    random = self.random

for i in reversed(xrange(1, len(x))):  
    # pick an element in x[:i+1] with which to exchange x[i]
    j = int(random() * (i+1))
    x[i], x[j] = x[j], x[i]

def sample(self, population, k):
    """Chooses k unique random elements from a population sequence.

    Returns a new list containing elements from the population while
    leaving the original population unchanged. The resulting list is
    in selection order so that all sub-slices will also be valid random
    samples. This allows raffle winners (the sample) to be partitioned
    into grand prize and second place winners (the subslices).

    Members of the population need not be hashable or unique. If the
    population contains repeats, then each occurrence is a possible
    selection in the sample.

    To choose a sample in a range of integers, use xrange as an
    argument.

    This is especially fast and space efficient for sampling from a
    large population:  sample(xrange(10000000), 60)"""
# Sampling without replacement entails tracking either potential
# selections (the pool) in a list or previous selections in a set.

# When the number of selections is small compared to the
# population, then tracking selections is efficient, requiring
# only a small set and an occasional reselection. For
# a larger number of selections, the pool tracking method is
# preferred since the list takes less space than the
# set and it doesn’t suffer from frequent reselections.

n = len(population)
if not 0 <= k <= n:
    raise ValueError("sample larger than population")
random = self.random
_int = int
result = [None] * k
setsize = 21  # size of a small set minus size of an empty list
if k > 5:
    setsize += 4 ** _ceil(_log(k * 3, 4))  # table size for big sets
if n <= setsize or hasattr(population, "keys"):
    # An n-length list is smaller than a k-length set, or this is a
    # mapping type so the other algorithm wouldn’t work.
pool = list(population)

for i in xrange(k):  # invariant: non-selected at [0, n-i)
    j = _int(random() * (n-i))
    result[i] = pool[j]
    pool[j] = pool[n-i-1]  # move non-selected item into vacancy
else:
    try:
        selected = set()
        selected.add = selected.add
        for i in xrange(k):
            j = _int(random() * n)
            while j in selected:
                j = _int(random() * n)
            selected.add(j)
            result[i] = population[j]
    except (TypeError, KeyError):  # handle (at least) sets
        if isinstance(population, list):
            raise
        return self.sample(tuple(population), k)

return result

## ------------------- real-valued distributions -------------------
def uniform(self, a, b):
    """Get a random number in the range [a, b) or [a, b] depending on
    rounding.""
    return a + (b-a) * self.random()

def triangular(self, low=0.0, high=1.0, mode=None):
    """Triangular distribution.

    Continuous distribution bounded by given lower and upper limits,
    and having a given mode value in-between.

    http://en.wikipedia.org/wiki/Triangular_distribution
    """
    u = self.random()
    c = 0.5 if mode is None else (mode - low) / (high - low)
    if u > c:
        u = 1.0 - u
        c = 1.0 - c
    low, high = high, low
    return low + (high - low) * (u * c) ** 0.5
def normalvariate(self, mu, sigma):
    """Normal distribution.

    mu is the mean, and sigma is the standard deviation.
    ""

    # mu = mean, sigma = standard deviation

    # Uses Kinderman and Monahan method. Reference: Kinderman,
    # A.J. and Monahan, J.F., "Computer generation of random
    # variables using the ratio of uniform deviates", ACM Trans

    random = self.random
    while 1:
        u1 = random()
        u2 = 1.0 - random()
        z = NV,MAGICCONST*(u1-0.5)/u2
        zz = z*z/4.0
        if zz <= -log(u2):
            break
    return mu + z*sigma
def lognormvariate(self, mu, sigma):
    """Log normal distribution.

    If you take the natural logarithm of this distribution, you'll get
    a normal distribution with mean mu and standard deviation sigma.
    mu can have any value, and sigma must be greater than zero.
    """
    return exp(self.normalvariate(mu, sigma))

def expovariate(self, lambd):
    """Exponential distribution.

    lambd is 1.0 divided by the desired mean. It should be
    nonzero. (The parameter would be called "lambda", but that is
    a reserved word in Python.) Returned values range from 0 to
    positive infinity if lambd is positive, and from negative
    infinity to 0 if lambd is negative.
    """
def vonmisesvariate(self, mu, kappa):
    # Circular data distribution.
    
    mu is the mean angle, expressed in radians between 0 and 2*pi, and kappa is the concentration parameter, which must be greater than or equal to zero. If kappa is equal to zero, this distribution reduces to a uniform random angle over the range 0 to 2*pi.

    # mu: mean angle (in radians between 0 and 2*pi)
    # kappa: concentration parameter kappa (>= 0)
    # if kappa = 0 generate uniform random angle

    # Based upon an algorithm published in: Fisher, N.I.,
random = self.random
if kappa <= 1e-6:
    return TWOPI * random()

a = 1.0 + sqrt(1.0 + 4.0 * kappa * kappa)
b = (a - sqrt(2.0 * a))/(2.0 * kappa)
r = (1.0 + b * b)/(2.0 * b)

while 1:
    u1 = random()

    z = cos(pi * u1)
    f = (1.0 + r * z)/(r + z)
c = kappa * (r - f)

    u2 = random()

    if u2 < c * (2.0 - c) or u2 <= c * exp(1.0 - c):
        break
u3 = random()
if u3 > 0.5:
    theta = (mu % TWOPI) + acos(f)
else:
    theta = (mu % TWOPI) - acos(f)

return theta

### -------------------------------- gamma distribution --------------------------------

def gammavariate(self, alpha, beta):
    """Gamma distribution. Not the gamma function!

    Conditions on the parameters are alpha > 0 and beta > 0.

    The probability distribution function is:

    \[
    pdf(x) = \frac{x^{\alpha - 1} \exp(-x / \beta)}{\Gamma(\alpha) \beta^{\alpha}}
    \]

    """

    x ** (alpha - 1) * math.exp(-x / beta)
    pdf(x) = -------------------------------
              math.gamma(alpha) * beta ** alpha
# alpha > 0, beta > 0, mean is alpha*beta, variance is alpha*beta
#
# Warning: a few older sources define the gamma distribution in terms
# of alpha > -1.0
#
if alpha <= 0.0 or beta <= 0.0:
    raise ValueError( 'gammavariate: alpha and beta must be > 0.0')

random = self.random

# Uses R.C.H. Cheng, "The generation of Gamma variables with non-integral shape parameters",

ainv = _sqrt (2.0 * alpha - 1.0)
bbb = alpha - LOG4
ccc = alpha + ainv

while 1:
    u1 = random()
    if not 1e-7 < u1 < .9999999:
        continue
    u2 = 1.0 - random()
v = _log(u1/(1.0-u1))/ainv

x = alpha*_exp(v)

z = u1*u1*u2

r = bbb+ccc*v-x

if r + SG.MAGICCONST - 4.5*z >= 0.0 or r >= _log(z):
    return x * beta

elif alpha == 1.0:
    # expovariate(1)
    u = random()
    while u <= 1e-7:
        u = random()
    return -_log(u) * beta

else:  # alpha is between 0 and 1 (exclusive)

    # Uses ALGORITHM GS of Statistical Computing – Kennedy & Gentle

    while 1:
        u = random()
        b = (_e + alpha)/_e
        p = b*u
        if p <= 1.0:
            x = p ** (1.0/alpha)
        else:
$$x = -\log((b-p)/alpha)$$

$$u1 = \text{random}()$$

if \(p > 1.0:\)
    if $$u1 <= x ** (alpha - 1.0):$$
        break
    elif $$u1 <= \exp(-x):$$
        break
    return $$x * \beta$$

### Gauss (faster alternative)

```python
def gauss(self, mu, sigma):
    """ Gaussian distribution. 

    mu is the mean, and sigma is the standard deviation. This is slightly faster than the normalvariate() function.

    Not thread-safe without a lock around calls.
    """

    # When x and y are two variables from [0, 1], uniformly distributed, then
    #
    #    \cos(2 \pi x) \sqrt{-2 \log(1-y)}
```

# \[ \sin(2\pi x) \cdot \sqrt{-2 \log(1-y)} \] #

# are two independent variables with normal distribution
# (\( \mu = 0, \sigma = 1 \)).
# (Lambert Meertens)
# (corrected version; bug discovered by Mike Miller, fixed by LM)

# Multithreading note: When two threads call this function
# simultaneously, it is possible that they will receive the
# same return value. The window is very small though. To
# avoid this, you have to use a lock around all calls. (I
# didn’t want to slow this down in the serial case by using a
# lock here.)

random = self.random
z = self.gauss_next
self.gauss_next = None

if z is None:
    x2pi = random() * TWOPI
    g2rad = \( \sqrt{-2.0 \cdot \log(1.0 - \text{random}())} \)
    z = \( \cos(x2pi) \cdot g2rad \)
    self.gauss_next = \( \sin(x2pi) \cdot g2rad \)

return \( \mu + z \cdot \sigma \)
## betavariate(beta)

### See


### for Ivan Frohne's insightful analysis of why the original implementation:

```python
def betavariate(self, alpha, beta):
    """ Beta distribution.  

    Conditions on the parameters are alpha > 0 and beta > 0.  
    Returned values range between 0 and 1.  

    """
    # This version due to Janne Sinkkonen, and matches all the std
```
# texts (e.g., Knuth Vol 2 Ed 3 pg 134 "the beta distribution").

```python
y = self.gammavariate(alpha, 1.)
if y == 0:
    return 0.0
else:
    return y / (y + self.gammavariate(beta, 1.))
```

## Pareto

```python
def pareto(self, alpha):
    """Pareto distribution. alpha is the shape parameter."""
    # Jain, pg. 495
    u = 1.0 - self.random()
    return 1.0 / pow(u, 1.0/alpha)
```

## Weibull

```python
def weibull(self, alpha, beta):
    """Weibull distribution.

    alpha is the scale parameter and beta is the shape parameter.
    """
    # Jain, pg. 499; bug fix courtesy Bill Arms
    ```
\[
u = 1.0 - \text{self.random()}
\]
\[
\text{return alpha} \ast \text{pow(-\log(u), 1.0/beta)}
\]

### Wichmann–Hill

```python
class WichmannHill(Random):

    VERSION = 1  # used by getstate/setstate

    def seed(self, a=None):
        """Initialize internal state from hashable object.

        None or no argument seeds from current time or from an operating
        system specific randomness source if available.

        If a is not None or an int or long, hash(a) is used instead.

        If a is an int or long, a is used directly. Distinct values
        between
        0 and 27814431486575L inclusive are guaranteed to yield distinct
        internal states (this guarantee is specific to the default
        Wichmann–Hill generator).
        ""
```
if a is None:
    try:
        a = long(hexlify(urandom(16)), 16)
    except NotImplementedError:
        import time
        a = long(time.time() * 256)  # use fractional seconds

if not isinstance(a, (int, long)):
    a = hash(a)

a, x = divmod(a, 30268)
a, y = divmod(a, 30306)
a, z = divmod(a, 30322)
self._seed = int(x)+1, int(y)+1, int(z)+1

self.gauss.next = None

def random(self):
    """Get the next random number in the range [0.0, 1.0)."""

    # Wichman–Hill random number generator.
    #
    # Algorithm AS 183:
    # An efficient and portable pseudo–random number generator
# This part is thread-unsafe:
# BEGIN CRITICAL SECTION
x, y, z = self._seed
x = (171 * x) % 30269
y = (172 * y) % 30307
z = (170 * z) % 30323
self._seed = x, y, z
# END CRITICAL SECTION

# Note: on a platform using IEEE-754 double arithmetic, this can
# never return 0.0 (asserted by Tim; proof too long for a comment).
return (x/30269.0 + y/30307.0 + z/30323.0) % 1.0

def getstate(self):
    """Return internal state; can be passed to setstate() later."""

#
# see also:
# Correction to Algorithm AS 183
#
# McLeod, A. I. (1985)
# A remark on Algorithm AS 183
return self.VERSION, self._seed, self.gauss.next

def setstate(self, state):
    """Restore internal state from object returned by getstate().""
    version = state[0]
    if version == 1:
        version, self._seed, self.gauss.next = state
    else:
        raise ValueError("state with version %s passed to 
        "Random.setstate() of version %s" %
        (version, self.VERSION))

def jumpahead(self, n):
    """Act as if n calls to random() were made, but quickly.

    n is an int, greater than or equal to 0.

    Example use: If you have 2 threads and know that each will
    consume no more than a million random numbers, create two Random
    objects r1 and r2, then do
    r2.setstate(r1.getstate())
    r2.jumpahead(1000000)
    Then r1 and r2 will use guaranteed-disjoint segments of the full
    period.
    """
if not n >= 0:
    raise ValueError("n must be >= 0")

x, y, z = self._seed
x = int(x * pow(171, n, 30269)) % 30269
y = int(y * pow(172, n, 30307)) % 30307
z = int(z * pow(170, n, 30323)) % 30323
self._seed = x, y, z

def _whseed(self, x=0, y=0, z=0):
    """Set the Wichmann–Hill seed from (x, y, z).
    These must be integers in the range [0, 256).
    ""

    if not type(x) == type(y) == type(z) == int:
        raise TypeError('seeds must be integers')
    if not (0 <= x < 256 and 0 <= y < 256 and 0 <= z < 256):
        raise ValueError('seeds must be in range(0, 256)')
    if 0 == x == y == z:
        # Initialize from current time
        import time
        t = long(time.time() * 256)
        t = int((t&0xffffffff) ^ (t>>24))
        t, x = divmod(t, 256)
t, y = divmod(t, 256)
t, z = divmod(t, 256)

# Zero is a poor seed, so substitute 1
self._seed = (x or 1, y or 1, z or 1)

self.gauss.next = None

def whseed(self, a=None):
    """Seed from hashable object's hash code.

    None or no argument seeds from current time. It is not guaranteed that objects with distinct hash codes lead to distinct internal states.

    This is obsolete, provided for compatibility with the seed routine used prior to Python 2.1. Use the .seed() method instead.
    """

    if a is None:
        self.__whseed()
        return

    a = hash(a)
a, x = divmod(a, 256)
a, y = divmod(a, 256)
a, z = divmod(a, 256)
x = (x + a) % 256 or 1
y = (y + a) % 256 or 1
z = (z + a) % 256 or 1
self.__whseed(x, y, z)

class SystemRandom(Random):
    """Alternate random number generator using sources provided
    by the operating system (such as /dev/urandom on Unix or
    CryptGenRandom on Windows).
    Not available on all systems (see os.urandom() for details).
    """
def random(self):
    """Get the next random number in the range [0.0, 1.0)."""
    return (long(_hexlify(_urandom(7)), 16) >> 3) * RECIP_BPF
def getrandbits(self, k):
    """getrandbits(k) -> x. Generates a long int with k random bits.
    """
    if k <= 0:
        raise ValueError('number of bits must be greater than zero')
    if k != int(k):
```python
raise TypeError('number of bits should be an integer')

bytes = (k + 7) // 8  # bits / 8 and rounded up
x = long(hexlify(urandom(bytes)), 16)
return x >> (bytes * 8 - k)  # trim excess bits

def _stub(self, *args, **kwds):
    "Stub method. Not used for a system random number generator."
    return None
seed = jumpahead = _stub

def _notimplemented(self, *args, **kwds):
    "Method should not be called for a system random number generator."
    raise NotImplementedError('System entropy source does not have state.')
getstate = setstate = _notimplemented

## ------------ test program ------------

def _test_generator(n, func, args):
    import time
    print(n, 'times', func.__name__)
    total = 0.0
    sqsum = 0.0
    smallest = 1e10
    largest = -1e10
```
t0 = time.time()
for i in range(n):
    x = func(*args)
    total += x
    sqsum = sqsum + x*x
    smallest = min(x, smallest)
    largest = max(x, largest)

    t1 = time.time()
    print(round(t1-t0, 3), 'sec,')
    avg = total/n
    stddev = _sqrt(sqsum/n - avg*avg)
    print('avg %g, stddev %g, min %g, max %g' % 
    (avg, stddev, smallest, largest))

    def _test (N=2000):
        _test_generator(N, random, ()
        _test_generator(N, normalvariate, (0.0, 1.0))
        _test_generator(N, lognormvariate, (0.0, 1.0))
        _test_generator(N, vonmisesvariate, (0.0, 1.0))
        _test_generator(N, gammavariate, (0.01, 1.0))
        _test_generator(N, gammavariate, (0.1, 1.0))
        _test_generator(N, gammavariate, (0.1, 2.0))
        _test_generator(N, gammavariate, (0.5, 1.0))
        _test_generator(N, gammavariate, (0.9, 1.0))
_test_generator(N, gammavariate, (1.0, 1.0))
_test_generator(N, gammavariate, (2.0, 1.0))
_test_generator(N, gammavariate, (20.0, 1.0))
_test_generator(N, gammavariate, (200.0, 1.0))
_test_generator(N, gauss, (0.0, 1.0))
_test_generator(N, betavariate, (3.0, 3.0))
_test_generator(N, triangular, (0.0, 1.0, 1.0/3.0))

# Create one instance, seeded from current time, and export its methods
# as module-level functions. The functions share state across all uses
# (both in the user’s code and in the Python libraries), but that’s fine
# for most programs and is easier for the casual user than making them
# instantiate their own Random() instance.

_inst = Random()
seed = _inst.seed
random = _inst.random
uniform = _inst.uniform
triangular = _inst.triangular
randint = _inst.randint
choice = _inst.choice
randrange = _inst.randrange
sample = _inst.sample
shuffle = _inst.shuffle
normalvariate = _inst.normalvariate
lognormvariate = _inst.lognormvariate
expovariate = _inst.expovariate
vonnisesvariate = _inst.vonnisesvariate
gammavariate = _inst.gammavariate
gauss = _inst.gauss
betavariate = _inst.betavariate
paretovariate = _inst.paretovariate
weibullvariate = _inst.weibullvariate
gestate = _inst.getstate
setstate = _inst.setstate
jumpahead = _inst.jumpahead
getrandbits = _inst.getrandbits

if __name__ == '__main__':
    __test__()

**B.3. Python code for the exact solution of the propagation operator to calculate the effects of Doppler broadening on the collective state signal**

from numpy import *
from scipy import *
from decimal import *

import matplotlib.pyplot as plt
plt.clf()
def w(om0, delt, tau):
    om1 = sqrt(om0**2 + delt**2)
    phi = om1*tau/2
    c = cos(phi)
    s = sin(phi)

    if delt == 0 and om0 == 0:
        return [[1, 0],
                [0, 1]]
    else:
        return [[c-j*delt/om1*s, -j*(om0/om1)*s],
                [-j*(om0/om1)*s, c+j*delt/om1*s]]

## CONSTANTS
m = .08491178973214/(6.0221413*10**(23)) ## mass of Rb 85 [kg]

m_87 = .086.90918052713/(6.0221413*10**(23))

c = 299792458 ## speed of light [m/s]
k = 1.3806488*10**(-23) ## Boltzmann constant [m^2 kg s^-2 K ^-1]

hbar = 1.05457173*10**(-34) ## h_bar [m^2 kg / s]

rb_gr = 2*pi*3.035732439060*10**9 ## Rubidium 85 D2 line F=2 to F=3

    ground state energy difference in radians
gamma_rb = 2*\pi*6.066618*10**6  # Spontaneous emission rate of D2 line [MHz]

## INITIALIZE

temp = gamma_rb*hbar/2/k  # temperature of the MOT [K]
c0 = [1,0]  # initial conditions
om0_1 = 5*10**6  # interaction zone 1 Raman Rabi frequency
tau_2 = 3*10**(−5)  # dark zone time

## MAXWELL-BOLTZMANN VELOCITY DISTRIBUTION IN ATOMS

mb_step = 100
v_av = sqrt(8*k*temp/pi/m_87)  # average speed in [m/s]
mb_x = arange(0,3*v_av,3*v_av/mb_step)  # domain
mb_y = sqrt((m_87/(2*pi*k*temp))**2*(-m_87*mb_x**2/(2*k*temp)))  # distribution
mb_sum = sum(mb_y)

n_a = 10**4  # number of atoms

for k in range(4):
    n_a = n_a*10

## PARAMETERS CALCULATED FROM INITIAL VALUES
tau_1 = pi/(2*om0_1)  # interaction zone 1 time for pi/2 pulse

x_max = pi/tau_2*2/sqrt(n_a)*1.4  # detuning maximum

x = arange(-x_max/2,x_max/2,x_max/100)  # detuning range

pop = 0

p_c = ones(len(x))  # collective signal

p_d = ones(len(x))  # corrected signal

## CALCULATION OF THE SIGNAL

for i in range(len(x)):
    w_int_1 = w(om0_1, x[i], tau_1)  # interaction zone 1
    w_int_2 = w(om0_1, x[i], tau_1)  # interaction zone 2
    w_dark = w(0,x[i], tau_2)  # dark zone
    w_tot = dot(w_int_2, dot(w_dark, w_int_1))

    pop_x = dot(w_tot, c0)
    pop = abs(real(pop_x[1]**2))
    p_c[i] = pop**n_a

for j in range(mb_step):
    w_int_1 = w(om0_1, x[i]+mb_x[j]/c*rb_gr, tau_1)  # interaction zone 1
\[
\begin{align*}
\text{w\_int\_2} &= w(\omega_0, x[i]+mb_x[j]/c*rb_gr, \tau_1) \quad \# \text{interaction zone 2} \\
\text{w\_dark} &= w(0, x[i]+mb_x[j]/c*rb_gr, \tau_2) \quad \# \text{dark zone} \\
\text{w\_tot} &= \text{dot}(\text{w\_int\_2}, \text{dot}(\text{w\_dark}, \text{w\_int\_1})) \\
\text{pop\_x} &= \text{dot}(\text{w\_tot}, c0) \\
\text{pop} &= \text{abs}(\text{real}(\text{pop\_x}[1]**2)) \\
\text{p\_d}[i] &= \text{p\_d}[i] \times \text{pop}^{\text{pop}(\text{a}*\text{mb}\_y[j]/\text{mb\_sum}/2)} \\
\text{w\_int\_1} &= w(\omega_0, x[i]-mb_x[j]/c*rb_gr, \tau_1) \quad \# \text{interaction zone 1} \\
\text{w\_int\_2} &= w(\omega_0, x[i]-mb_x[j]/c*rb_gr, \tau_1) \quad \# \text{interaction zone 2} \\
\text{w\_dark} &= w(0, x[i]-mb_x[j]/c*rb_gr, \tau_2) \quad \# \text{dark zone} \\
\text{w\_tot} &= \text{dot}(\text{w\_int\_2}, \text{dot}(\text{w\_dark}, \text{w\_int\_1})) \\
\text{pop\_x} &= \text{dot}(\text{w\_tot}, c0) \\
\text{pop} &= \text{abs}(\text{real}(\text{pop\_x}[1]**2)) \\
\text{p\_d}[i] &= \text{p\_d}[i] \times \text{pop}^{\text{pop}(\text{a}*\text{mb}\_y[j]/\text{mb\_sum}/2)} \\
\end{align*}
\]

```python
## PLOT SIGNAL
ax = plt.subplot(2,2,k+1)
plt.axis(ymax=1)
p1, = plt.plot(x, p_c, '--')
p2, = plt.plot(x, p_d)
```
B.4. Python code for the effect of detector efficiency on the collective state signal

```python
from numpy import *
from scipy import *
import matplotlib.pyplot as plt

plt.clf()

## initialization
n_a = 2*10**6  # number of atoms
t = 3*10**(-5)  # dark zone time period
```
## Detuning range

\[ x_{\text{max}} = \frac{(\pi+2)}{(t\cdot \sqrt{n_a})} \]

\[ x = \text{arange}(-x_{\text{max}}\cdot 1.5, x_{\text{max}}\cdot 1.5, 2\cdot x_{\text{max}}/1000) \]

\[ p_2 = (\cos(x\cdot t/2))^2 \] # population of state 2 after Ramsey fringe experiment

ax1 = plt.subplot(2, 2, 1)

## Experimental parameters

\[ \eta = 0.7 \]
\[ \gamma = 10^{-4} \]
\[ \tau = 10^{-10} \]

## Ideal signal, lower bound, and upper bound

\[ s_0 = p_2^2 \]

\[ s_{\text{lb}} = 1-\eta\cdot (1-p_2^2\cdot n_a) \cdot (1-e^{(-n_a^2\cdot 2\cdot \gamma \cdot \tau)} / 4) \]

\[ s_{\text{ub}} = 1-\eta\cdot (1-p_2^2\cdot n_a) \cdot (1-e^{(-n_a \cdot \gamma \cdot \tau)}) \]

\[ \text{textstr} = r'\$\eta=%.2f$' \quad r'\$\gamma_{\text{sa}}=10^{%.0f}$' \quad \%((\text{math. log10}(\gamma \cdot \tau)))' \]

ax1.text(0.05, .95, textstr, fontsize=16, transform=ax1.transAxes, verticalalignment='top', horizontalalignment='left')

plt.title('(a)')
plt.tick_params(axis='both', which='major', labelsize=14)
plt.locator_params(nbins=6)

p1, = plt.plot(x, s_0, color='black')
p2, = plt.plot(x, s_lb, linestyle='--', color='black')
p3, = plt.plot(x, s_ub, linestyle=':', color='black')

# plt.legend([p1, p2, p3], ["ideal signal", "lower bound", "upper bound"], prop
# ={'size':14})
plt.axis(xmin=-150, xmax=150)
plt.ylabel('Collective state signal', fontsize=14)
plt.xlabel('$\delta$ (Hz)', fontsize=14)

ax2 = plt.subplot(2,2,2)

## experimental parameters
eta = .99
gam = 10**4
taus = 10**-10

## ideal signal, lower bound, and upper bound
s_0 = p_2**n_a
s_lb = 1-eta*(1-p_2**n_a)*(1-e**(-n_a*2*gam*taus/4))
s_ub = 1-eta*(1-p_2**n_a)*(1-e**(-n_a*gam*taus))

textstr = r'$\eta=%.2f$\n$r\gamma_{sa}=10^{%.0f}$' %(eta, (math.
log10(gam*taus))))
## Ideal Signal, Lower Bound, and Upper Bound

### Experimental Parameters

**eta** = 0.7  
**gam** = 10**5  
**taus** = 10**-5
\[ s_0 = p_2^{*n_a} \]
\[ s_{lb} = 1 - \eta \ast (1 - p_2^{*n_a}) \ast (1 - e^{*(-n_a*2*\gamma*taus/4)}) \]
\[ s_{ub} = 1 - \eta \ast (1 - p_2^{*n_a}) \ast (1 - e^{*(-n_a*\gamma*taus)}) \]

```python
textstr = r'$\eta=%.2f$\n$r\gamma_{sa}\tau=%.0f$' % (eta, gam*taus)
ax3.text(0.05, .95, textstr, fontsize=16, transform=ax3.transAxes,
    verticalalignment='top', horizontalalignment='left')
```

```python
plt.title('(c)')
plt.tick_params(axis='both', which='major', labelsize=14)
plt.locator_params(nbins=6)
```

```python
p1, = plt.plot(x, s_0, color='black')
p2, = plt.plot(x, s_lb, linestyle='--', color='black')
p3, = plt.plot(x, s_ub, linestyle=':', color='black')

# plt.legend([p1, p2, p3], ['ideal signal', 'lower bound', 'upper bound'], prop = {'size': 14})
plt.axis(xmin=-150, xmax=150)
plt.ylabel('Collective state signal', fontsize=14)
plt.xlabel('$\delta$ (Hz)', fontsize=14)
```

```
ax4 = plt.subplot(2, 2, 4)
## experimental parameters
eta = .99
gam = 10**5
```
taus = 10**-5

## ideal signal, lower bound, and upper bound

\[ s_0 = p_2^{n_a} \]
\[ s_{lb} = 1-\eta(1-p_2^{n_a})*(1-e^{(n_a*2*gam*taus/4)}) \]
\[ s_{ub} = 1-\eta(1-p_2^{n_a})*(1-e^{(n_a*gam*taus)}) \]

```
textstr = r'$\eta=%.2f$ \n$\gamma_{sa}$tau=%.0f$' %(eta , gam*taus)
ax4.text(0.05,.95,textstr , fontsize=16,transform=ax4.transAxes ,
    verticalalignment='top',horizontalalignment='left ')
```

```
plt.title('(d)')
plt.tick_params(axis='both', which='major', labelsize=14)
plt.locator_params(nbins = 6)
```

```
p1, = plt.plot(x, s_0, color='black')
p2, = plt.plot(x, s_lb, linestyle='--', color='black')
p3, = plt.plot(x, s_ub, linestyle=':', color='black')
# plt.legend([p1,p2,p3],['ideal signal', 'lower bound', 'upper bound'], prop
      ={'size':14})
plt.axis(xmin=-150,xmax=150)
plt.ylabel('Collective state signal', fontsize=14)
plt.xlabel('$\delta$ (Hz)', fontsize=14)
```
B.5. Wolfram Mathematica code for the collective state decay

Clear[n, k0, y1, y2, y3, y4]

DSolve[{y4'[t] == -k0 * n * y4[t], y4[0] == 1}, {y4[t]}, t]

{ {y4[t] \[\rightarrow\] \[Eexp\]-k0nt} }  

Simplify[DSolve[{y3'[t] == k0 * n * \[Eexp\]-k0nt - k0 * 2(n - 1) * y3[t], y3[0] == 0}, {y3[t]}, t]]

{ {y3[t] \[\rightarrow\] \[\frac{\Eexp2k0(-1+n)t(1+\Eexp-k0(2+n)t)n}{-2+n}\] } }  

Simplify[

DSolve[{y2'[t] == -k0 * n * y2[t] + k0 * 2(n - 1) * \(\frac{\Eexp-2k0(-1+n)t(1+\Eexp-k0(2+n)t)n}{-2+n}\) ,
y2[0] == 0}, {y2[t]}, t]]

{ {y2[t] \[\rightarrow\] \[\frac{2\Eexp-2k0(-1+n)t(1+\Eexp-k0(2+n)t)n(1+\Eexp-k0(2+n)t)n}{-2+n}\]} }  

Simplify[

DSolve[

{y1'[t] == k0 * n * \(\frac{2\Eexp-2k0(-1+n)t(1+\Eexp-k0(2+n)t)n(1+\Eexp-k0(2+n)t)n}{-2+n}\) ,
y1[0] == 0}, {y1[t]}, t]]

{ {y1[t] \[\rightarrow\] \[\frac{\Eexp-2k0nt(2k0nt+2k0nt\Eexp2k0nt+2k0nt\Eexp-2k0nt-2k0nt\Eexp-2k0nt(1+\Eexp-k0(2+n)t)n)}{(-2+n)^2}\] } }
\[
\text{Simplify} \left[ \frac{e^{-2k0nt} (e^{2k0nt} (-2+n)^2 - e^{2k0nt} n^2 - 2 e^{k0nt} (-1+n)(-2+k0(-2+n)nt))}{(-2+n)^2} + \right. \\
\left. \frac{2 e^{-2k0(-1+n) t} (-1+n) n (1 + e^{k0(-2+n) t} (-1+k0(-2+n) t))}{(-2+n)^2} + \right. \\
\left. \frac{e^{-2k0(-1+n) t} (-1+n) n (1 + e^{k0(-2+n) t} (-1+k0(-2+n) t))}{-2+n} + e^{-k0nt} \right] \\
\]

1

Clear\[n, k0, y1, y2, y3, y4\]
\[n = 3;\]
\[k0 = .1;\]
\[y1 = \frac{e^{-2k0nt} (e^{2k0nt} (-2+n)^2 - e^{2k0nt} n^2 - 2 e^{k0nt} (-1+n)(-2+k0(-2+n)nt))}{(-2+n)^2};\]
\[y2 = \frac{2 e^{-2k0(-1+n) t} (-1+n) n (1 + e^{k0(-2+n) t} (-1+k0(-2+n) t))}{(-2+n)^2};\]
\[y3 = \frac{e^{-2k0(-1+n) t} (-1+n) n (1 + e^{k0(-2+n) t} (-1+k0(-2+n) t))}{-2+n};\]
\[y4 = e^{-k0nt};\]

Plot\[\{y1, y2, y3, y4\}, \{t, 0, 20\}, \text{PlotRange} \to \{0, 1\},\]
PlotLegends \to \{\rho\text{11, }\rho\text{22, }\rho\text{33, }\rho\text{44},\]
PlotStyle \to \{\text{Dashed, Dashing[Tiny], Dashing[Large], Thick}\}\]

Clear\[n, k0, y1, y2, y3, y4\]
\[\text{DSolve}\[\{y3'[t] == -k0 * 2(n - 1) * y3[t], y3[0] == 1\}, \{y3[t]\}, t\\]
\[\{\{y3[t] \to e^{-2(-k0+k0n)t}\}\}\]

\[\text{Simplify} \left[ \text{DSolve} \left[ \{y2'[t] == k0 * 2(n - 1) * e^{-2(-k0+k0n)t} - k0 * n * y2[t], y2[0] == 0\}, \right. \right. \]
\[\left. \left. \{y2[t]\}, t\right]\right]\]
\[\{\{y2[t] \to \frac{2 e^{-2k0(-1+n) t} (-1+n) n (1 + e^{k0(-2+n) t} (-1+n))}{-2+n}\}\} \]
Simplify[
 DSolve[
 {y1'[t] == k0 * n * \(\frac{2e^{-2k0(-1+n)t}}{-2+n} - 1 + e^{k0(-2+n)t}) - 1 + n\), y1[0] == 0},
 {y1[t]}, t]
]

\[
\begin{align*}
  n &= 3; \\
  k0 &= .1; \\
  y1 &= \frac{e^{-2k0(-1+n)t}(e^{2k0(-1+n)t}(-2+n) - 2e^{k0(-2+n)t}(-1+n) + n)}{-2+n}; \\
  y2 &= \frac{-2e^{-2k0(-1+n)t}(1+e^{k0(-2+n)t})(-1+n)}{-2+n}; \\
  y3 &= e^{-2(-k0+k0n)t}; \\
  \text{Plot}[[y1, y2, y3], \{t, 0, 20\}, \text{PlotRange} \to \{0, 1\}, \\
  \text{PlotLegends} \to \{\rho11, \rho22, \rho33\}, \text{PlotStyle} \to \{\text{Dashed}, \text{Dashing}[\text{Tiny}], \text{Thick}\}]
\end{align*}
\]

Clear[n, k0, y1, y2, y3, y4]

DSolve[{y2'[t] == -k0 * n * y2[t], y2[0] == 1}, {y2[t]}, t]

\[
\begin{align*}
  n &= 3; \\
  k0 &= .1; \\
  \text{Plot}[[1 - e^{-3t}, e^{-3t}], \{t, 0, 20\}, \text{PlotRange} \to \{0, 1\}, \text{PlotLegends} \to \{\rho11, \rho22\}, \text{PlotStyle} \to \{\text{Dashed}, \text{Thick}\}]
\end{align*}
\]
B.6. Python code of rubidium 85 D2 line data that other programs can call

```python
from numpy import *
from scipy import *

# Fundamental physical constants

# speed of light [m/s]
c = 2.99792458*10**8

# permeability of vacuum [N/A^2]
mu_0 = 4*pi*10**(-7)

# permittivity of vacuum [F/m]
epsilon_0 = (mu_0*c**2)**(-1)

# Planck's constant [J*s]
h = 6.6260689633*10**(-34)
hbar = h/(2*pi)

# Rubidium physical properties

# atomic number
z = 37

# atomic mass [kg]
```

mass = 1.40999319970*10**(25)

################################ D2 line data ################################

# wavelength in vacuum [m]
d2_lambda = 780.24136827127*10**(9)

# lifetime [s]
d2_tau = 26.234877*10**(-9)

# decay rate [Hz] radians/s
d2_gamma_r = 2*pi*6.066618*10**6

# decay rate [Hz] cycles/s
d2_gamma = 6.066618*10**6

# hyperfine splitting [Hz] cycles/s
d2_omega_23 = 3.035732439060*10**9
d2_omega_1prime2prime = 29.37290*10**6
d2_omega_2prime3prime = 63.40161*10**6
d2_omega_3prime4prime = 120.64068*10**6

d2_omega_1prime3prime = d2_omega_1prime2prime + d2_omega_2prime3prime
d2_omega_1prime4prime = d2_omega_1prime3prime + d2_omega_3prime4prime
\[ d_{2\omega 2'4'} = d_{2\omega 2'3'} + d_{2\omega 3'4'} \]

# g-factors for \( F = 2, 3, 1 \) prime, \( 2 \) prime, \( 3 \) prime, and \( 4 \) prime

\[
\begin{align*}
    d_{2\cdot g2} &= -1/3 \\
    d_{2\cdot g3} &= 1/3 \\
    d_{2\cdot g1\text{,prime}} &= -1 \\
    d_{2\cdot g2\text{,prime}} &= 1/9 \\
    d_{2\cdot g3\text{,prime}} &= 7/18 \\
    d_{2\cdot g4\text{,prime}} &= 1/2
\end{align*}
\]

# # # # # # # # # # # # # # # D2 dipole matrix elements and saturation intensitiies  # # # # # # # # # # # # # # #

# cycling transition dipole moment and saturation intensity \([\text{nW/cm}^2]\)

\[
\begin{align*}
    d_{2\cdot dm\text{,cycle}} &= 1/\sqrt{2} \\
    d_{2\cdot isat\text{,cycle}} &= 1.6693235
\end{align*}
\]

## other dipole matrix elements and saturation intensities \([\text{nW/cm}^2]\) for sigma plus transitions

# \( F=2 \) m=0 to \( F'=1,2,3 \) m=1 sigma +

\[
\begin{align*}
    d_{2\cdot dm\text{,2011}} &= \sqrt{1/20} \\
    d_{2\cdot dm\text{,2021}} &= \sqrt{7/36} \\
    d_{2\cdot dm\text{,2031}} &= \sqrt{4/45}
\end{align*}
\]
\[
\begin{align*}
\text{d2\_isat\_2011} &= \text{d2\_isat\_cycle} \times (\text{d2\_dm\_cycle}/\text{d2\_dm\_2011})^2 \\
\text{d2\_isat\_2021} &= \text{d2\_isat\_cycle} \times (\text{d2\_dm\_cycle}/\text{d2\_dm\_2021})^2 \\
\text{d2\_isat\_2031} &= \text{d2\_isat\_cycle} \times (\text{d2\_dm\_cycle}/\text{d2\_dm\_2031})^2 \\
\end{align*}
\]

# F=3 m=0 to F’=(2,3,4) m=1 sigma +

\[
\begin{align*}
\text{d2\_dm\_3021} &= \text{sqrt}(1/63) \\
\text{d2\_dm\_3031} &= \text{sqrt}(5/36) \\
\text{d2\_dm\_3041} &= \text{sqrt}(5/28) \\
\end{align*}
\]

\[
\begin{align*}
\text{d2\_isat\_3021} &= \text{d2\_isat\_cycle} \times (\text{d2\_dm\_cycle}/\text{d2\_dm\_3021})^2 \\
\text{d2\_isat\_3031} &= \text{d2\_isat\_cycle} \times (\text{d2\_dm\_cycle}/\text{d2\_dm\_3031})^2 \\
\text{d2\_isat\_3041} &= \text{d2\_isat\_cycle} \times (\text{d2\_dm\_cycle}/\text{d2\_dm\_3041})^2 \\
\end{align*}
\]

B.7. Python code for rubidium 87 D1 line data that other programs can use

```python
from numpy import *
from scipy import *

# Fundamental physical constants

# speed of light [m/s]
c = 2.99792458*10**8

# permeability of vacuum [N/A^2]
mu_0 = 4*pi*10**(-7)
```
# permittivity of vacuum [F/m]
epsilon_0 = (mu_0*c**2)**(-1)

# Planck's constant [J*s]
h = 6.6260689633*10**(-34)
hbar = h/(2*pi)

############################ Rubidium physical properties ############################

# atomic number
z = 37

# atomic mass [kg]
mass = 1.44316064872*10**(-25)

############################ D2 line data ################################

# wavelength in vacuum [m]
d1_lambda = 794.97885115623*10**(-9)

# lifetime [s]
d1_tau = 27.67927*10**(-9)

# decay rate [Hz] cycles/s
d1_gamma = 5.750056*10**6
# decay rate [Hz] radians/s
d1_gamma_r = 2*pi*d1_gamma

# hyperfine splitting [Hz] cycles/s
d1_omega_12 = 6.8346826109042909*10**9

d1_omega_1prime2prime = 814.515*10**6

# g-factors for F = 2, 3, 1 prime, 2 prime, 3 prime, and 4 prime
d1_g1 = -1/2

d1_g2 = 1/2

d1_g1_prime = -1/6

d1_g2_prime = 1/6

# # # # # # # # # # # # # # # D2 dipole matrix elements and saturation intensities
# # # # # # # # # # # # # # #

# cycling transition dipole moment and saturation intensity [mW/cm^2]
dm_cycle = 1/sqrt(2)
isat_cycle = 1.6693235

# # other dipole matrix elements and saturation intensities [mW/cm^2] for
# sigma plus transitions

# F=1 m=0 to F'=(1,2) m=1 sigma +
\[ d_{1\_dm\_1011} = -\sqrt{\frac{1}{12}} \]
\[ d_{1\_dm\_1021} = -\sqrt{\frac{1}{4}} \]

\[ d_{1\_isat\_1011} = \text{isat\_cycle} \times \left( \frac{\text{dm\_cycle}}{d_{1\_dm\_1011}} \right)^2 \]
\[ d_{1\_isat\_1021} = \text{isat\_cycle} \times \left( \frac{\text{dm\_cycle}}{d_{1\_dm\_1021}} \right)^2 \]

# F=2 m=0 to F’=(1,2) m=1 sigma +
\[ d_{1\_dm\_2011} = \sqrt{\frac{1}{12}} \]
\[ d_{1\_dm\_2021} = \sqrt{\frac{1}{4}} \]

\[ d_{1\_isat\_2011} = \text{isat\_cycle} \times \left( \frac{\text{dm\_cycle}}{d_{1\_dm\_2011}} \right)^2 \]
\[ d_{1\_isat\_2021} = \text{isat\_cycle} \times \left( \frac{\text{dm\_cycle}}{d_{1\_dm\_2021}} \right)^2 \]

# F=1 m=-1 to F’=(1,2) m=0 sigma +
\[ d_{1\_dm\_1m110} = -\sqrt{\frac{1}{12}} \]
\[ d_{1\_dm\_1m120} = -\sqrt{\frac{1}{12}} \]

\[ d_{1\_isat\_1m110} = \text{isat\_cycle} \times \left( \frac{\text{dm\_cycle}}{d_{1\_dm\_1m110}} \right)^2 \]
\[ d_{1\_isat\_1m120} = \text{isat\_cycle} \times \left( \frac{\text{dm\_cycle}}{d_{1\_dm\_1m120}} \right)^2 \]

# F=2 m=-1 to F’=(1,2) m=0 sigma +
\[ d_{1\_dm\_2m110} = \sqrt{\frac{1}{4}} \]
\[ d_{1\_dm\_2m120} = \sqrt{\frac{1}{4}} \]

\[ d_{1\_isat\_2m110} = \text{isat\_cycle} \times \left( \frac{\text{dm\_cycle}}{d_{1\_dm\_2m110}} \right)^2 \]
\[ d_{1\text{isat}} \cdot m_{120} = \text{isat\_cycle} \cdot (d_{m\text{cycle}}/d_{1\_dm\_2m120})^{2} \]

\[
\text{# magnetic principal number maximum:}
\]
\[
m_{F1\_max} = 1 \quad \text{## } F = 1
\]
\[
m_{F2\_max} = 2 \quad \text{## } F = 2
\]
\[
m_{F1\_prime\_max} = 1 \quad \text{## } F' = 1
\]
\[
m_{F2\_prime\_max} = 2 \quad \text{## } F' = 2
\]

\[
\text{# number of Zeeman sublevels}
\]
\[
\text{levels}_{F1} = m_{F1\_max} \cdot 2 + 1 \quad \text{## } F = 1
\]
\[
\text{levels}_{F2} = m_{F2\_max} \cdot 2 + 1 \quad \text{## } F = 2
\]
\[
\text{levels}_{F1\_prime} = m_{F1\_prime\_max} \cdot 2 + 1 \quad \text{## } F' = 1
\]
\[
\text{levels}_{F2\_prime} = m_{F2\_prime\_max} \cdot 2 + 1 \quad \text{## } F' = 2
\]

**B.8. Python code to calculate the effect of magnetic field in Zeeman sublevels of rubidium 87 D1 line - including \( F = 1, F = 2, \) and \( F' = 1 \)**

```python
from numpy import *
from scipy import *
from decimal import *
from rb87_data import *
from find_m import *
from ham_b import *
```
from copy_array import *

import matplotlib.pyplot as plt

getcontext().prec = 50
plt.clf()

# MAGNETIC FIELD HAMILTONIAN

mu_B = 5.788381806638*10**(-9)  # Bohr magneton in eV/G
hbar = 6.5821192815*10**(-16)  # h_bar in eV * s

b = [B_x, B_y, B_z]  # Creates array of the magnetic fields
### Selim's alpha notation

\[
\alpha_{F1} = d_1 g_1 \mu_B \left( \frac{1}{\hbar \times 2 \times \pi} \right) \quad \text{##} \quad \mu_B / \hbar \, \text{in} / \text{s} / \text{G}
\]

\[
\alpha_{F2} = d_1 g_2 \mu_B \left( \frac{1}{\hbar \times 2 \times \pi} \right) \quad \text{##} \quad \mu_B / \hbar \, \text{in} / \text{s} / \text{G}
\]

\[
\alpha_{F1\prime} = d_1 g_{1\prime} \mu_B \left( \frac{1}{\hbar \times 2 \times \pi} \right) \quad \text{##} \quad \mu_B / \hbar \, \text{in} / \text{s} / \text{G}
\]

\[
\alpha_{F2\prime} = d_1 g_{2\prime} \mu_B \left( \frac{1}{\hbar \times 2 \times \pi} \right) \quad \text{##} \quad \mu_B / \hbar \, \text{in} / \text{s} / \text{G}
\]

#### USER INPUT

#### Arrays to pass onto the magnetic field Hamiltonian function

\[
\alpha = [\alpha_{F1}, \alpha_{F2}, \alpha_{F1\prime}]
\]

\[
\mathsf{m}_{\text{max}} = [\mathsf{m}_{F1\text{max}}, \mathsf{m}_{F2\text{max}}, \mathsf{m}_{F1\prime\text{max}}]
\]

\[
\mathsf{levels} = [\mathsf{levels}_{F1}, \mathsf{levels}_{F2}, \mathsf{levels}_{F1\prime}]
\]

\[
\mathsf{ham} = \text{zeros(shape=(sum(levels),sum(levels))), dtype=complex}.
\]

#### Call the magnetic field Hamiltonian function

\[
\mathsf{h}_b = \text{ham_b(\mathsf{ham}, \alpha, \mathsf{levels}, \mathsf{m_{\text{max}}, b})}
\]

\[
\text{print(\mathsf{h}_b)}
\]

#### INITIALIZATION OF NECESSARY VARIABLES

## PUT RABI FREQUENCY

```python
levels_raman = levels_F1 + levels_F2  # Number of levels participating in Raman transition

om = zeros(levels_raman)  # Initialize Raman Rabi frequency vector
for i in range(levels_raman):
    om[i] = 6*pi*10**6  # in cycles/s
```

## PUT DETUNING FROM THE EXCITED STATE

```python
detun = 0
```

## PUT PLOT PARAMETERS

```python
pts = 1000  # number of points to plot
```

## Pi pulse time

```python
#om_raman = om[0]*om[1]/(2*detun)
om_raman = om[0]
t_pi = pi/om_raman

maximum = om_raman*90  # detuning range to plot
delta = arange(-maximum/2,maximum/2,maximum/pts)
```
## Source matrices

\[
s_1 = \text{zeros}(\text{shape}=(\text{sum(levels)}, \text{sum(levels)}), \text{dtype}={\text{complex}})
\]

\[
s_2 = \text{zeros}(\text{shape}=(\text{sum(levels)}, \text{sum(levels)}), \text{dtype}={\text{complex}})
\]

\[
s_3 = \text{zeros}(\text{shape}=(\text{sum(levels)}, \text{sum(levels)}), \text{dtype}={\text{complex}})
\]

## Number of points to compute between each data point

## The M matrix will be raised to this power, essentially, to compute the
time evolution

\[
n_p = 10^{15}
\]

\[
dt = 1/n_p
\]

## Initial state vector, where all the atoms are in F1

\[
a = \text{zeros}(\text{sum(levels)}\times2)
\]

for \(i\) in range(levels_F1):

\[
a[i\ast(\text{sum(levels)}+1)] = 1/\text{levels}_F1
\]

## Identity matrix

\[
\text{mat}_1 = \text{eye}(\text{sum(levels)}\times2)
\]

## Final state vector after a pi pulse

\[
\text{atxt} = \text{zeros}(\text{shape}=(\text{sum(levels)}\times2, \text{pts}), \text{dtype}={\text{complex}})
\]
for m in range(pts):
    ham = zeros(shape=(sum(levels),sum(levels)), dtype=complex)
    copy_array_2d(h_b, ham)

    ### Hamiltonian due to interaction with electric field
    count = 0
    for i in range(len(ham)):
        ### Interaction between F1 and F1' (sigma+), and energy of F1 states
        dif_F11_prime = (levels_F1 - levels_F1_prime)/2
        dif_F12 = (levels_F2 - levels_F1)/2
        dif_F21_prime = (levels_F2 - levels_F1_prime)/2
        if i > m_F1_prime_max - 1 + dif_F11_prime and i < levels_F1 -
            dif_F11_prime:
            ham[i,i] = ham[i,i] + delta[m]/2
            ham[i,i+levels_F1+levels_F2-1] = ham[i,i+levels_F1+levels_F2-1]
                    + om[count]/2
            ham[i+levels_F1+levels_F2-1,i] = ham[i+levels_F1+levels_F2-1,i]
                    + om[count]/2
            count = count + 1

        ### Interaction between F2 and F1' (sigma+), and energy of F2 states
if $i > \text{levels}_F1 + m_{F1\text{prime}\text{max}} - 1 + \text{dif}_{F21\text{prime}}$ and $i < \text{levels}_F1 + \text{levels}_F2 - \text{dif}_{F21\text{prime}}$

$\text{ham}[i,i] = \text{ham}[i,i] - \Delta [m]/2$

$\text{ham}[i,i+\text{levels}_F1] = \text{ham}[i,i+\text{levels}_F1] + \text{o}[\text{count}]/2$

$\text{ham}[i+\text{levels}_F1,i] = \text{ham}[i+\text{levels}_F1,i] + \text{o}[\text{count}]/2$

$\text{count} = \text{count} + 1$

elif $i > \text{levels}_F1 + \text{levels}_F2 - 1$ and $i < \text{levels}_F1 + \text{levels}_F2 + \text{levels}_F1\text{prime} - \text{m}_{F1\text{prime}\text{max}}$

$\text{ham}[i,i] = \text{ham}[i,i] - \text{detun}$

# print(i,i,ham[i,i])

## Calls the external program to find the M matrix

mat = find_M(\text{ham, s1,s2,s3,0,0,0})

## Calculation of the final state vector

\text{atxt}[::,m] = \text{dot(linalg.matrix_power(mat*dt+mat_1,int(t_pi/dt+1)),a)}
plt.subplot(4,1,1)
# for k in range(levels_F1):
#     plt.plot(delta, real(atxt[k*(len(ham)+1),:]))
plt.axis(xmin=-maximum/2,xmax=maximum/2)
p0, = plt.plot(delta, real(atxt[0*(len(ham)+1),:])), linewidth=2)
p1, = plt.plot(delta, real(atxt[1*(len(ham)+1),:])), linewidth=2)
p2, = plt.plot(delta, real(atxt[2*(len(ham)+1),:])), linewidth=2)
plt.legend([p0, p1, p2], ["|m=2>","|m=1>","|m=0>","|m=-1>"]) plt.ylabel("|F = 1> Population")

plt.subplot(4,1,2)
# for k in range(levels_F2):
#     plt.plot(delta, real(atxt[(k+levels_F1)*(len(ham)+1),:]))
plt.axis(xmin=-maximum/2,xmax=maximum/2)
p3, = plt.plot(delta, real(atxt[(0+levels_F1)*(len(ham)+1),:])), linewidth=2)
p4, = plt.plot(delta, real(atxt[(1+levels_F1)*(len(ham)+1),:])), linewidth=2)
p5, = plt.plot(delta, real(atxt[(2+levels_F1)*(len(ham)+1),:])), linewidth=2)
p6, = plt.plot(delta, real(atxt[(3+levels_F1)*(len(ham)+1),:])), linewidth=2)
p7, = plt.plot(delta, real(atxt[(4+levels_F1)*(len(ham)+1),:])), linewidth=2)
plt.legend([p3, p4, p5, p6, p7], ["|m=2>"","|m=1>"","|m=0>"","|m=-1>"","|m=-2>"]) plt.ylabel("|F = 2> Population")

plt.subplot(4,1,3)
plt.axis(xmin=-maximum/2,xmax=maximum/2)
p8, = plt.plot(delta, real(atxt[(0+levels_F1+levels_F2)*(len(ham)+1),:]))
B.9. Python code to find $[H, \rho]$ used by the above program

```python
from numpy import *
from scipy import *
from decimal import *

getcontext().prec = 50

## This function finds the M matrix for calculating the evolution of the
## density matrix. It takes seven parameters: h, the Hamiltonian; (s1, s2, s3)
## , the
```
## source matrices; and (decay1, decay2, decay3), the levels from which the
decays occur.

## For example, if the first decay occurs from the 2nd level, then decay =
2.

def find_M(h, s1, s2, s3, decay1, decay2, decay3):
    ## return the size of the Hamiltonian in one of the dimensions
    lx = len(h)

    ## initialize the M matrix
    mat = zeros((lx**2, lx**2), dtype=complex)

    for q in range(lx):
        for r in range(lx):
            # indices for H*rho part
            i_1 = q*lx
            i_2 = r*lx

            # indices for rho*conj_transp(H) part
            i_3 = q
            i_4 = r

            for s in range(lx):
                # add the terms for H*rho part
                mat[i_1, i_2] = mat[i_1, i_2] - 1j*h[q, r]
\[ i_1 = i_1 + 1 \]
\[ i_2 = i_2 + 1 \]

```python
# add the terms for rho*conj_transp(H) part
mat[i_3, i_4] = mat[i_3, i_4] + 1j*conjugate(h[q, r])
i_3 = i_3 + lx;
i_4 = i_4 + lx;

# include the source terms here:
mat[q + q*lx, decay1 + (decay1-1)*lx-1] = mat[q + q*lx, decay1 + (decay1-1)*lx-1] + s1[q,q]
mat[q + q*lx, decay2 + (decay2-1)*lx-1] = mat[q + q*lx, decay2 + (decay2-1)*lx-1] + s2[q,q]
mat[q + q*lx, decay3 + (decay3-1)*lx-1] = mat[q + q*lx, decay3 + (decay3-1)*lx-1] + s3[q,q]
return mat
```

B.10. Python code called on by the program above to find the portion of the Hamiltonian that is dependent on the external magnetic field

```python
# USER INPUT
from numpy import *
from scipy import *
from decimal import *
from rb_87_data import *
```

```python
B.10. Python code called on by the program above to find the portion of the Hamiltonian that is dependent on the external magnetic field
```
\[
\mu_B = 5.788381806638 \times 10^{-9} \quad \text{Bohr magneton in eV/G}
\]
\[
hbar = 6.5821192815 \times 10^{-16} \quad \text{hbar in eV} \times s
\]

```python
def ham_b(ham, alpha, levels, m_max, b):
    count = 0
    for m in range(len(levels)):
        for i in range(levels[m]):
            ham[i + count, i + count] = alpha[m] * b[2] * (m_max[m] - i)
        for k in range(levels[m]):
            if k == i + 1:
                ham[i + count, k + count] = alpha[m] * \sqrt{(m_max[m] + 1 - (m_max[m] - k) * (m_max[m] - k + 1)) / 2 * (b[0] - 1 \times j * b[1])}
            if k == i - 1:
                ham[i + count, k + count] = alpha[m] * \sqrt{(m_max[m] + 1 - (m_max[m] - i) * (m_max[m] - i + 1)) / 2 * (b[0] + 1 \times j * b[1])}
        count = count + levels[m]
    return ham
```

### B.11. Python code for finding the Raman Rabi frequency, light shift, \( \pi \) pulse time, and linewidth, given the power and radius of the two Raman beams

```python
from numpy import *
```
from sympy import *
from scipy import *
from rb_85_data import *

import matplotlib.pyplot as plt
plt.clf()

# power in each Raman beam [mW]
power_plus = 50 # F = 2
power_minus = 50 # F = 3

# radius of each Raman beam [cm]
r_plus = .75 # F = 2
r_minus = .75 # F = 3

# intensity of each Raman beam [mW/cm**2]
i_plus = power_plus/(pi*r_plus**2) # F = 2
i_minus = power_minus/(pi*r_minus**2)# F = 3

# Rabi frequency of each sigma plus transition [Hz] [cycles/s]
rabi_21 = sqrt(i_plus/(2*d2_isat_2011))*d2_gamma # F = 2, m = 0 to F' = 1, m = 1
rabi_22 = sqrt(i_plus/(2*d2_isat_2021))*d2_gamma # F = 2, m = 0 to F' = 2, m = 1
\( \text{rabi}_{23} = \sqrt{\frac{i_{\text{plus}}}{(2 \times d_{\text{isat}} \times 2031)}} \times d_{2 \text{gamma}} \)  
\# \text{F = 2, m = 0 to F' = 3, m = 1} 

\( \text{rabi}_{32} = \sqrt{\frac{i_{\text{minus}}}{(2 \times d_{\text{isat}} \times 3021)}} \times d_{2 \text{gamma}} \)  
\# \text{F = 3, m = 0 to F' = 2, m = 1} 

\( \text{rabi}_{33} = \sqrt{\frac{i_{\text{minus}}}{(2 \times d_{\text{isat}} \times 3031)}} \times d_{2 \text{gamma}} \)  
\# \text{F = 3, m = 0 to F' = 3, m = 1} 

\( \text{rabi}_{34} = \sqrt{\frac{i_{\text{minus}}}{(2 \times d_{\text{isat}} \times 3041)}} \times d_{2 \text{gamma}} \)  
\# \text{F = 3, m = 0 to F' = 4, m = 1} 

# Detuning to F' = 1, 2, 3, 4 for the two Raman beams [Hz] 
\( \delta_{1} = d_{2 \text{omega}_{23}}/2 - d_{2 \text{omega}_{1prime3prime}} \)  
\# \text{F' = 1} 

\( \delta_{2} = d_{2 \text{omega}_{23}}/2 - d_{2 \text{omega}_{2prime3prime}} \)  
\# \text{F' = 2} 

\( \delta_{3} = d_{2 \text{omega}_{23}}/2 \)  
\# \text{F' = 3} 

\( \delta_{4} = d_{2 \text{omega}_{23}}/2 + d_{2 \text{omega}_{3prime4prime}} \)  
\# \text{F' = 4} 

# Detuning for F = 3 to F' transition, when it is up shifted as well. 
# By our convention, this detuning takes on a minus sign, although it's up shifted. 
\( \delta_{5} = -d_{2 \text{omega}_{23}}/2 - d_{2 \text{omega}_{2prime3prime}} \)  
\# \text{F' = 2} 

\( \delta_{6} = -d_{2 \text{omega}_{23}}/2 \)  
\# \text{F' = 3} 

\( \delta_{7} = -d_{2 \text{omega}_{23}}/2 + d_{2 \text{omega}_{3prime4prime}} \)  
\# \text{F' = 4} 

# Detuning for omega_1 beam from F = 2 
\( \delta_{211} = -(d_{2 \text{omega}_{23}}/2 - d_{2 \text{omega}_{1prime3prime}}) \)  
\# \text{F' = 1} 

\( \delta_{212} = -(d_{2 \text{omega}_{23}}/2 - d_{2 \text{omega}_{2prime3prime}}) \)  
\# \text{F' = 2}
\[ \delta_{213} = -d_2 \omega_{23}/2 \quad \text{# } F' = 3 \]

\# Detuning for omega_2 beam from F = 2
\[ \delta_{221} = \delta_{211} - d_2 \omega_{23} \quad \text{# } F' = 1 \]
\[ \delta_{222} = \delta_{212} - d_2 \omega_{23} \quad \text{# } F' = 2 \]
\[ \delta_{223} = \delta_{213} - d_2 \omega_{23} \quad \text{# } F' = 3 \]

\# Detuning for omega_2 beam from F = 3
\[ \delta_{322} = \delta_{212} \quad \text{# } F' = 2 \]
\[ \delta_{323} = \delta_{213} \quad \text{# } F' = 3 \]
\[ \delta_{324} = -(d_2 \omega_{23}/2 + d_2 \omega_{33}/4) \quad \text{# } F' = 4 \]

\# Detuning for omega_1 beam from F = 3
\[ \delta_{312} = \delta_{322} + d_2 \omega_{23} \quad \text{# } F' = 2 \]
\[ \delta_{313} = \delta_{323} + d_2 \omega_{23} \quad \text{# } F' = 3 \]
\[ \delta_{314} = \delta_{324} + d_2 \omega_{23} \quad \text{# } F' = 4 \]

\# Raman Rabi frequency calculation [Hz] [cycles/s]
\[ \text{raman} = \frac{\text{rabi}_2 \text{rabi}_3}{2 \delta_2} + \frac{\text{rabi}_2 \text{rabi}_3}{2 \delta_3} \]
\[ \text{print}'\text{Raman Rabi frequency: ', raman, ' '[cycles/s]' }\]

\# Light shift calculation [Hz]
\[ \text{beta}_211 = \frac{\text{rabi}_2 \text{rabi}_2}{4 \delta_211} \quad \text{# } F = 2 \text{ to } F' = 1 \text{ omega}_1 \]
\[ \text{beta}_212 = \frac{\text{rabi}_2 \text{rabi}_2}{4 \delta_212} \quad \text{# } F = 2 \text{ to } F' = 2 \text{ omega}_1 \]
\[ \beta_{213} = \frac{\text{rabi}_{23}**2}{4*\delta_{213}} \] # F = 2 to F' = 3 omega_1

\[ \beta_{221} = \frac{\text{rabi}_{21}**2}{4*\delta_{221}} \] # F = 2 to F' = 1 omega_2

\[ \beta_{222} = \frac{\text{rabi}_{22}**2}{4*\delta_{222}} \] # F = 2 to F' = 2 omega_2

\[ \beta_{223} = \frac{\text{rabi}_{23}**2}{4*\delta_{223}} \] # F = 2 to F' = 3 omega_2

\[ \beta_{312} = \frac{\text{rabi}_{32}**2}{4*\delta_{312}} \] # F = 3 to F' = 2 omega_1

\[ \beta_{313} = \frac{\text{rabi}_{33}**2}{4*\delta_{313}} \] # F = 3 to F' = 3 omega_1

\[ \beta_{314} = \frac{\text{rabi}_{34}**2}{4*\delta_{314}} \] # F = 3 to F' = 4 omega_1

\[ \beta_{322} = \frac{\text{rabi}_{32}**2}{4*\delta_{322}} \] # F = 3 to F' = 2 omega_2

\[ \beta_{323} = \frac{\text{rabi}_{33}**2}{4*\delta_{323}} \] # F = 3 to F' = 3 omega_2

\[ \beta_{324} = \frac{\text{rabi}_{34}**2}{4*\delta_{324}} \] # F = 3 to F' = 4 omega_2

\[ \beta_2 = \beta_{211}+\beta_{212}+\beta_{213}+\beta_{221}+\beta_{222}+\beta_{223} \]

\[ \beta_3 = \beta_{312}+\beta_{313}+\beta_{314}+\beta_{322}+\beta_{323}+\beta_{324} \]

\[ \beta = \beta_2 - \beta_3 \]

print('Light shift:', beta, '[cycles/s]')

## Plot of the population of state F = 3 after optical pumping into F = 2, and after Raman pulse
# Number of points to plot
step = 10**3

d_max = 10*raman
d_step = d_max*2/step

# Detuning (big delta) [Hz] [cycles/s]
detune = arange(-d_max+betta, d_max+betta, d_step)

# Generalized Raman Rabi frequency
om_g = sqrt(raman**2+(detune-betta)**2)

# Pi pulse time
t_pi = 1/(2*raman)
print('Pi pulse time:', t_pi, '[s]')

# Find the population
pop = zeros(step)
for i in range(step):
    pop[i] = (raman/om_g[i]*sin(2*pi*om_g[i]*t_pi/2))**2

# Find the linewidth
half = max(pop)/2
line = 0
```python
max_i = 0
for i in range(1, step):
    if pop[i] == max(pop):
        max_i = i

line_i = 0
for i in range(max_i, step):
    if abs((pop[i] - half)/half) < 10**(-2):
        line_i = i

line = 2*(line_i-max_i)*d_step
print('Linewidth:', line, '[cycles/s]')

# Plot the population
plt.plot(detune, pop)
plt.axis(xmin=min(detune), xmax=max(detune))
plt.xlabel('Detuning [Hz]')
plt.ylabel('Signal')
plt.show()
```