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Preparation of high-principal-quantum-number Rydberg states

2023 UROP summer project

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Rydberg States

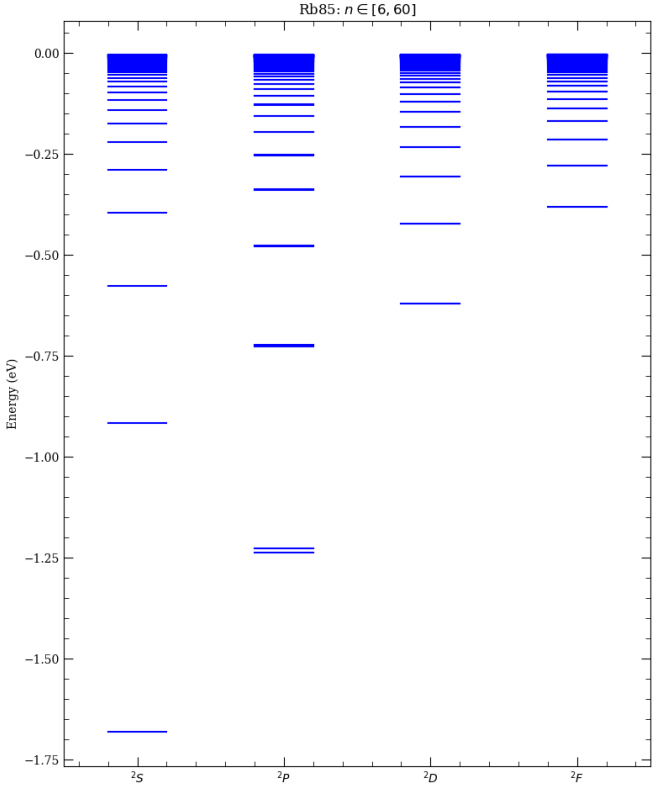

Figure 1: Rb energy levels for different l manifolds

Rydberg States are states with high principal quantum number n, for example, n=50. Classically, this corresponds to a very large electron orbit, making the properties similar to hydrogen [1].

$$E_{n,l,j} = \frac{-Ry}{(n - \delta_{lj})^2} \qquad (1)$$

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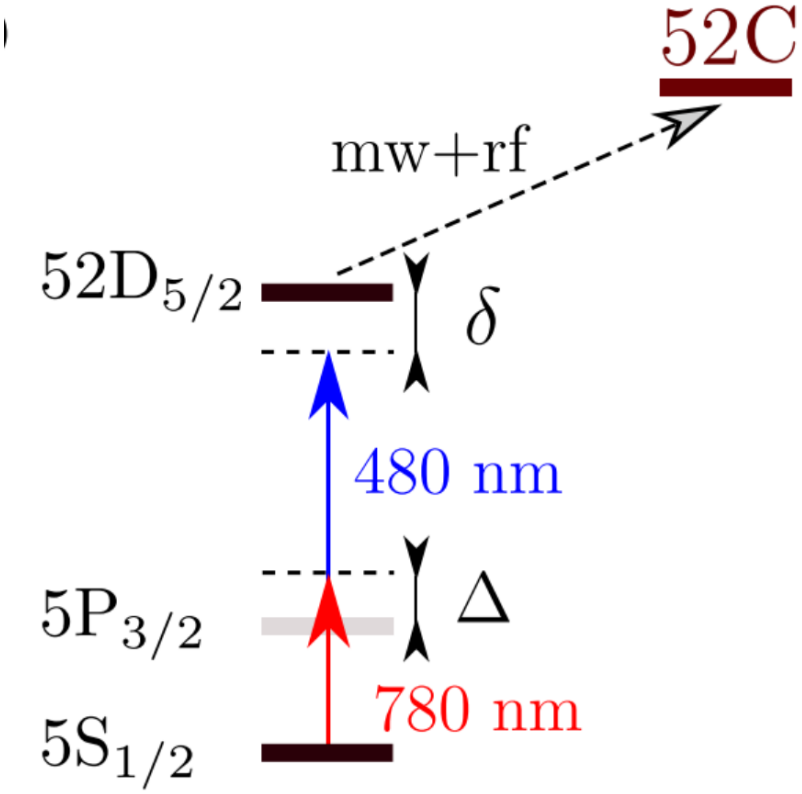
Laser Excitation



We usually excite Rydberg atoms by laser. Heavy, alkaline atoms are preferred.

- Single photon process, ultraviolet regime
- Multi photon process, infared or visible light regime

Figure 2: Preparation of Rydberg States

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Lifetime

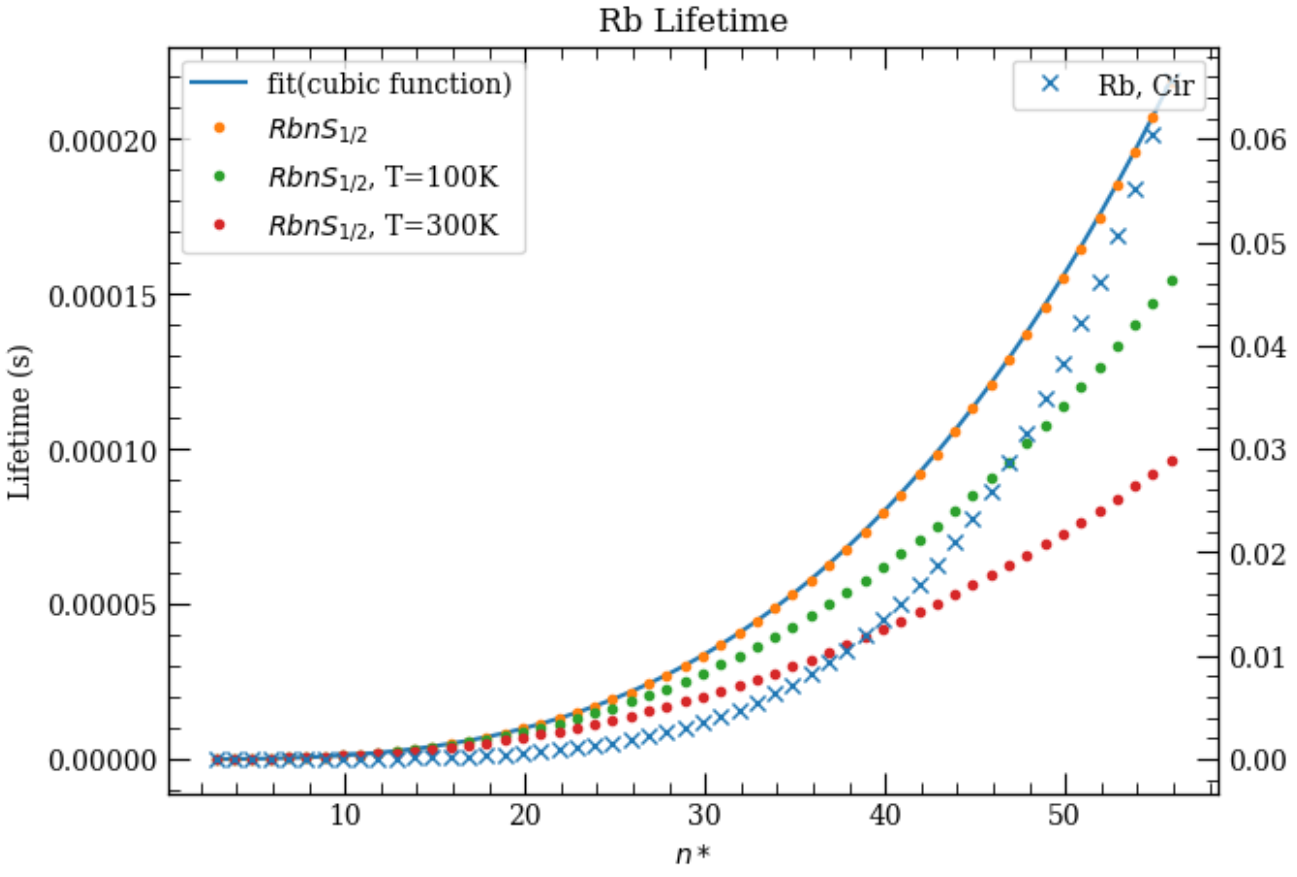

Figure 3: Rb lifetimes. Note the circular state has much longer lifetime than other states.

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Interactions

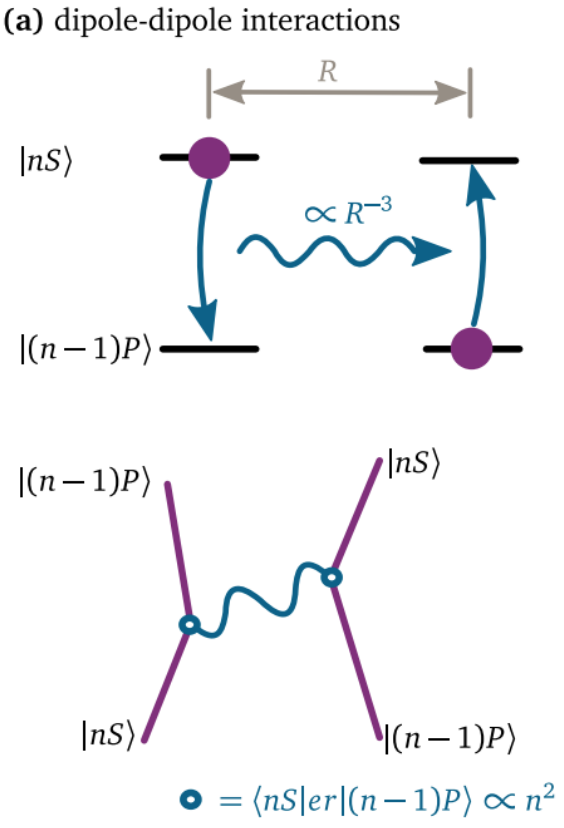

Figure 4: $[2]V(R) = C_3/R^3 \propto n^4/R^3$

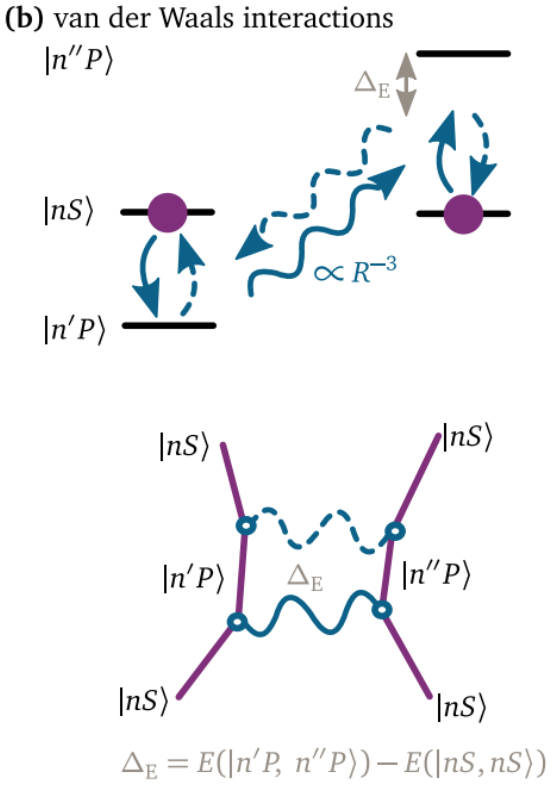

Figure 5: $[2]V(R) = C_6/R^6 \propto n^1 1/R^6$

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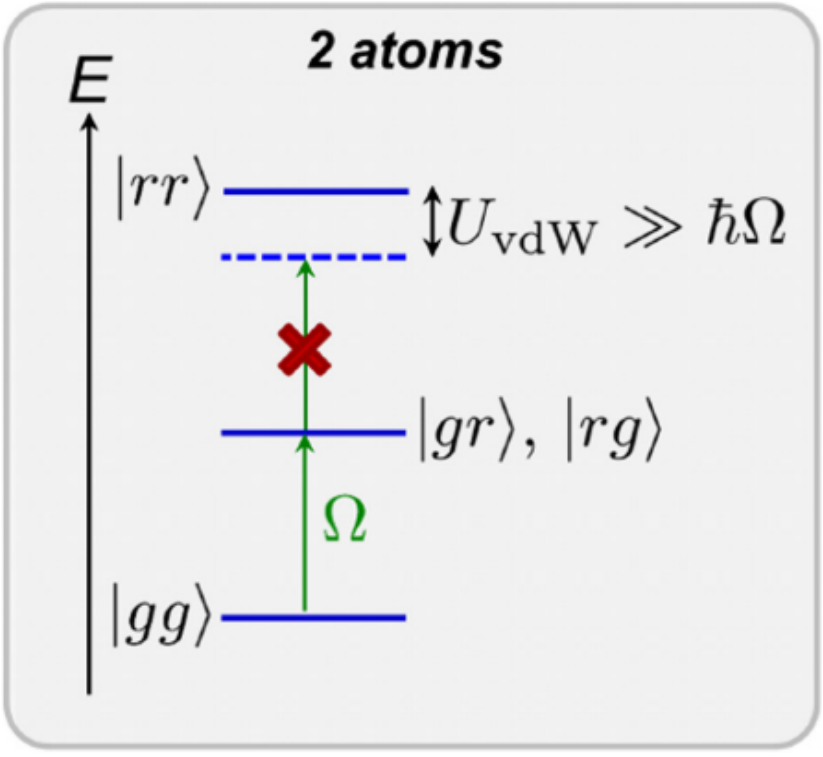
Rydberg Blockade



For one atom, $|g\rangle$ is coupled to $|r\rangle$.
For two nearby atoms, $|rr\rangle$ is shifted by Van de Waals interaction and is thus forbidden.
New rabi oscillation would be between
($|gg\rangle$ and $(|gr\rangle + |rg\rangle)/\sqrt{2}$

Figure 6: Van de Waals interaction prevents the double excitation [1].

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Observation of Rydberg Blockade

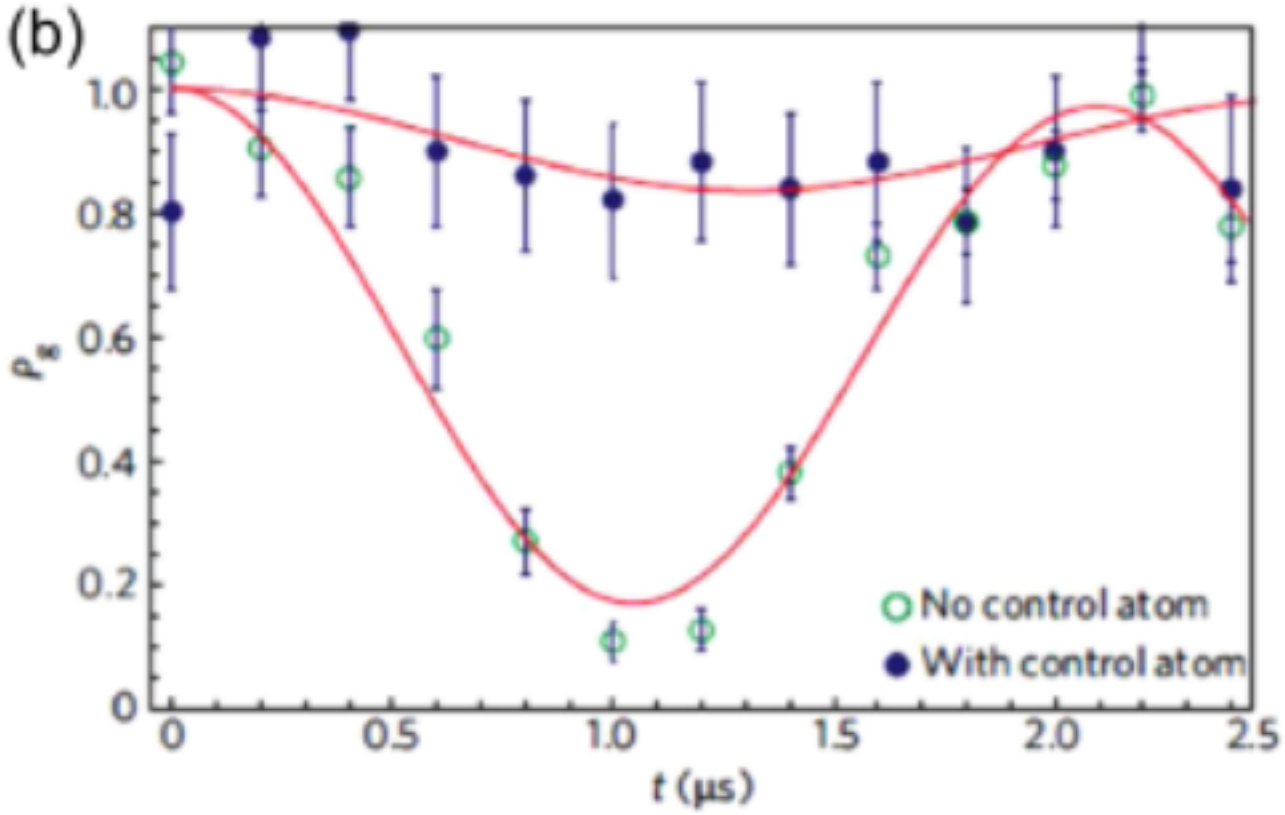

Figure 7: Rabi Oscillations with or without control atoms [3].

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Single Photon Excitation

We apply the optical Bloch equations:

$$\dot{\rho}_{ee} = i\Omega(\tilde{\rho}_{eg} - \tilde{\rho}_{ge})/2 - \gamma_2\rho_{ee}$$
$$\dot{\rho}_{gg} = -i\Omega(\tilde{\rho}_{eg} - \tilde{\rho}_{ge})/2 + \gamma_2\rho_{ee}$$
$$\dot{\tilde{\rho}}_{eg} = -(\gamma_1 - i\Delta)\tilde{\rho}_{eg} + i\Omega(\rho_{ee} - \rho_{gg})/2$$
$$\dot{\tilde{\rho}}_{ge} = -(\gamma_1 + i\Delta)\tilde{\rho}_{ge} + i\Omega(\rho_{gg} - \rho_{ee})/2$$

where γ_2 is the longitudinal decay rate and γ_1 is the transverse (dephasing) rate. We choose to excite the Rb state from 5S1/2 to 50P3/2. All the parameters can be evaluated from dipole matrix elements between the two states. Transition frequency is approximately 10^{15} Hz while the rabi frequency is 10^6 Hz.

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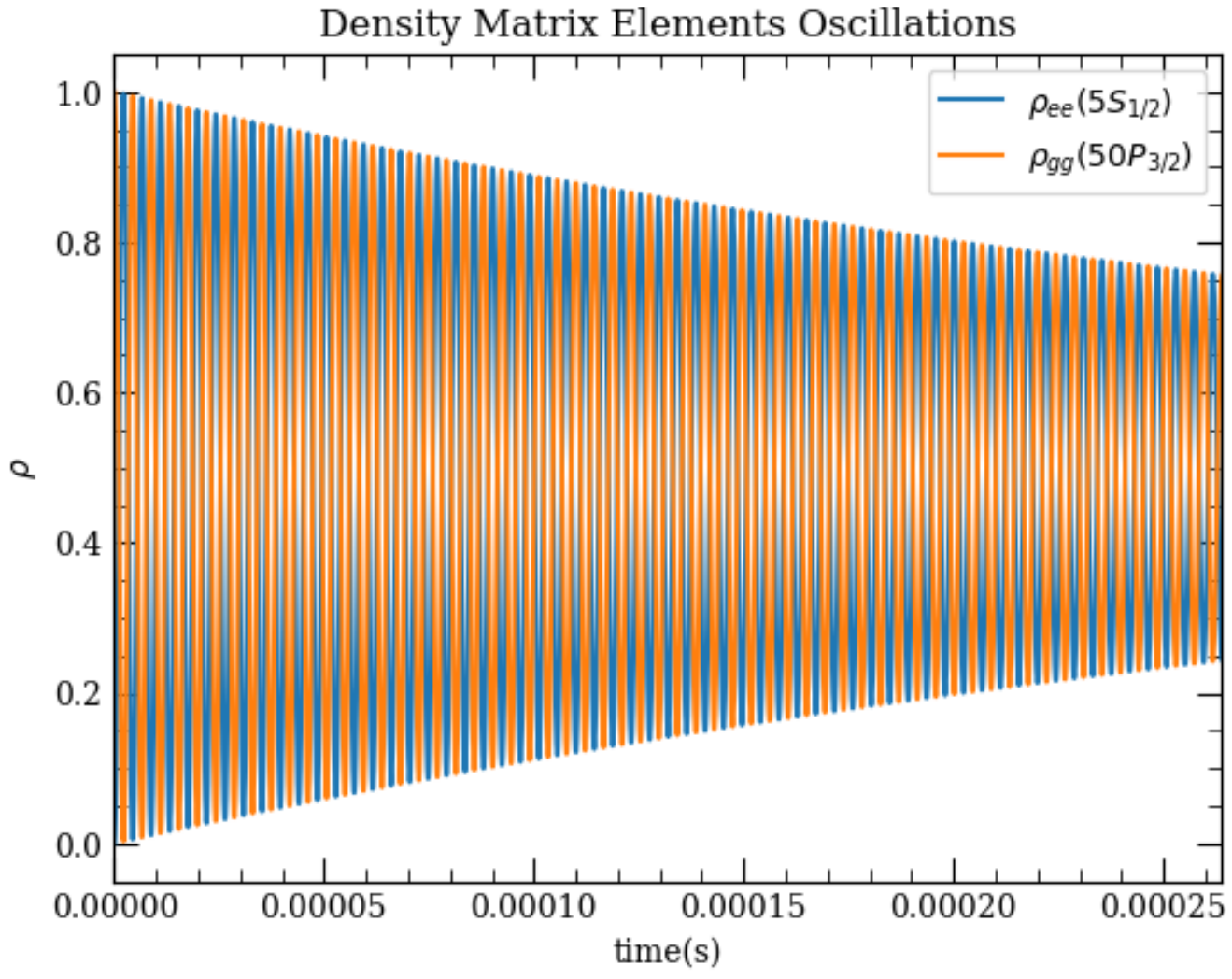
Simulation Results



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Multi-Photon Excitation

We can write down the multi-level OBEs [4]:

$$\frac{\mathrm{d}\langle\sigma_{m,n}^{F,F}\rangle}{\mathrm{d}t} = \sum_q [-(\Omega_n^q)^*\langle\sigma_{m,n+q}^{F,F'}\rangle - \Omega_m^q\langle\sigma_{m+q,n}^{F',F}\rangle + \Gamma_{m,n}^q\langle\sigma_{m+q,n+q}^{F',F'}\rangle + \mathcal{B}_{m,n}^{F,F}],$$

$$\frac{\mathrm{d}\langle\sigma_{m,n}^{F',F'}\rangle}{\mathrm{d}t} = \sum_q [(\Omega_{m-q}^q)^*\langle\sigma_{m-q,n}^{F,F'}\rangle + \Omega_{n-q}^q\langle\sigma_{m,n-q}^{F',F}\rangle - \Gamma\langle\sigma_{m,n}^{F',F'}\rangle + \mathcal{B}_{m,n}^{F',F'}],$$

$$\frac{\mathrm{d}\langle\sigma_{m,n}^{F,F'}\rangle}{\mathrm{d}t} = \sum_q [\Omega_{n-q}^q\langle\sigma_{m,n-q}^{F,F}\rangle - \Omega_m^q\langle\sigma_{m+q,n}^{F',F'}\rangle + (\mathrm{i}\Delta - \Gamma/2)\langle\sigma_{m,n}^{F,F'}\rangle + \mathcal{B}_{m,n}^{F,F'}].$$

where the first two terms describe the interaction with electric fields and the third term describes the sponetaneous emission and the last term is the interaction with magnetic fields. (in our case 0)

Three-level OBEs

$$\dot{\rho}_{ee} = -\gamma_2 \rho_{ee} + \frac{i}{2}\Omega_2(\rho_{ei} - \rho_{ie})$$

$$\dot{\rho}_{ii} = -\gamma_1 \rho_{ii} + b_2 \gamma_2 \rho_{ee} - \frac{i}{2}\Omega_2(\rho_{ei} - \rho_{ie}) + \frac{i}{2}\Omega_1(\rho_{ig} - \rho_{gi})$$

$$\dot{\rho}_{gg} = b_1 \gamma_1 \rho_{ii} - \frac{i}{2}\Omega_1(\rho_{ig} - \rho_{gi})$$

$$\dot{\rho}_{ei} = (-\frac{\gamma_1 + \gamma_2}{2} + i\delta_2)\rho_{ei} + \frac{i}{2}\Omega_2(\rho_{ee} - \rho_{ii}) + \frac{i}{2}\Omega_1 \rho_{eg}$$

$$\dot{\rho}_{eg} = (-\frac{\gamma_2}{2} + i(\delta_1 + \delta_2))\rho_{eg} + \frac{i}{2}\Omega_1 \rho_{ei} - \frac{i}{2}\Omega_2 \rho_{ig}$$

$$\dot{\rho}_{ig} = (-\frac{\gamma_1}{2} + i\delta_1)\rho_{ig} + \frac{i}{2}\Omega_1(\rho_{ii} - \rho_{gg}) - \frac{i}{2}\Omega_2 \rho_{eg}$$

We have three equations for the three-level populations and six equations for the coherence terms, so in total nine differential equations.

Fourier Transform

These are nine linear differential equations. We can apply the Fourier transform on both sides and note that:

$$\tilde{f}'(t) = i\omega\tilde{f}(\omega) \tag{2}$$

The nine differential equations can be transformed to nine algebraic equations:

$$i\omega\tilde{\rho}_{ee} = -\gamma_2\tilde{\rho}_{ee} + \frac{i}{2}\Omega_2(\tilde{\rho}_{ei} - \tilde{\rho}_{ie})$$

$$\vdots$$

The problem of solving differential equations is converted to finding the eigenvalues and eigenvectors of a non-hermitian matrix.

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Non-Hermitian Matrix

$$\begin{bmatrix} -\gamma_2 & 0 & 0 & \frac{i\Omega_2}{2} & \cdots \\ b_2\gamma_2 & -\gamma_1 & 0 & -\frac{i\Omega_2}{2} & \cdots \\ 0 & b_1\gamma_1 & 0 & 0 & \cdots \\ \frac{i\Omega_2}{2} & -\frac{i\Omega_2}{2} & 0 & -\frac{\gamma_2+\gamma_1}{2} + i\delta_2 & \cdots \\ \vdots & \vdots & \vdots & \vdots & \end{bmatrix}$$

We numerically diagonalise the matrix and do the inverse fourier transform and we found that the final solution of ρs are superposition of nine normal modes, with the eigenvalues being complex.

$$\rho = c_1 \mathrm{v}_1 \exp(i w_1 t) + c_2 \mathrm{v}_2 \exp(i w_2 t) \cdots + c_9 \mathrm{v}_9 \exp(i w_9 t) \qquad (3)$$

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Two-Photon Excitation

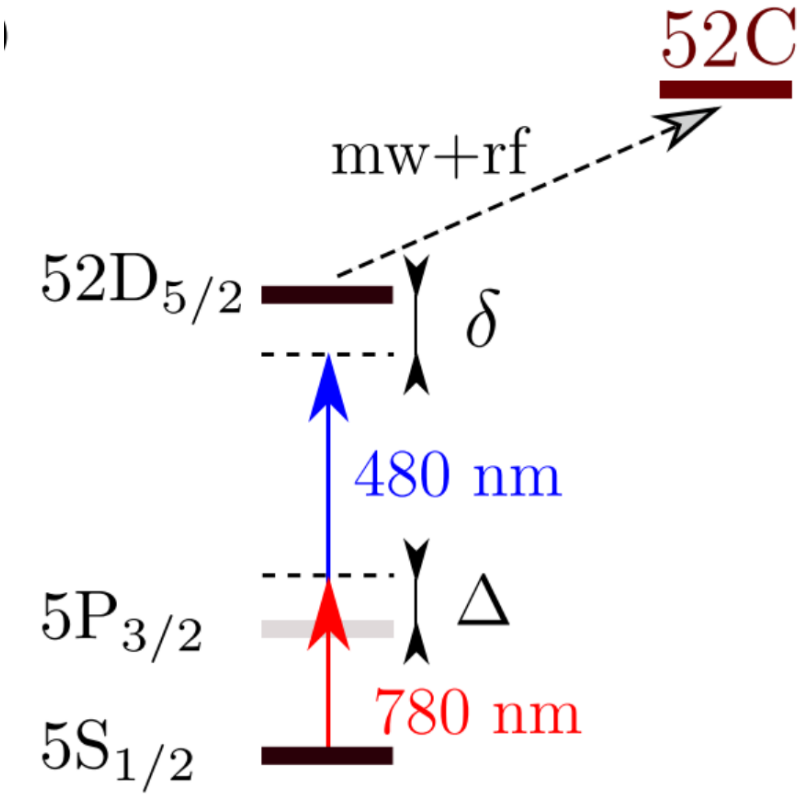

Figure 9: [1]

There are two detunings here: Δ is the detuning between the first laser and the intermediate level, δ is the total detuning. From the adiabatic elimination, we can convert the three-level system to two-level system, with the

$$\Omega_{eff} = \frac{\Omega_1 \Omega_2}{2\Delta}$$

$$\delta_{eff} = \delta - \left(\frac{|\Omega_1|^2}{4\Delta} - \frac{|\Omega_2|^2}{4\Delta}\right)$$

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Simulations

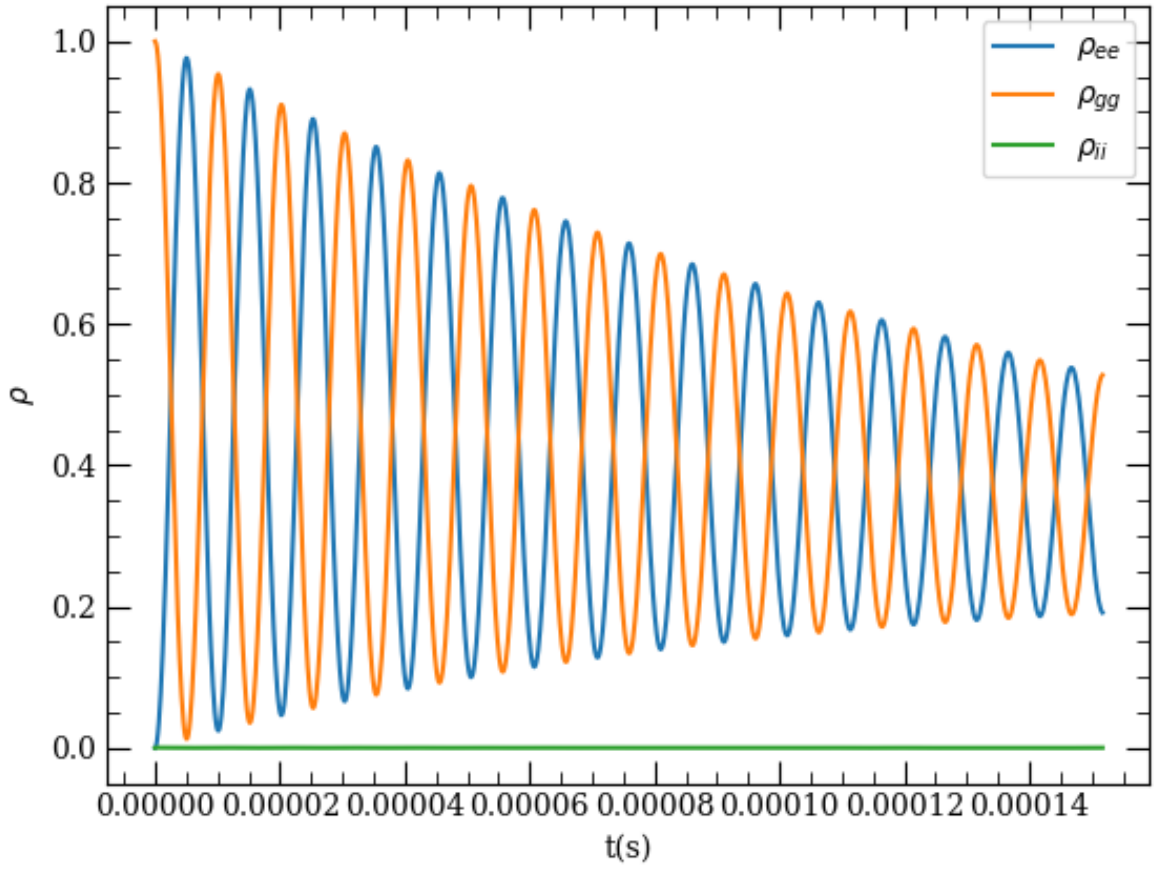

Figure 10: We choose three states as 5S1/2, 5P3/2 and 60S1/2(Rydberg state). We choose large Δ so that the intermediate state is not populated.

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Fidelity

The fidelity is given by

$$f = (1 - P_p)(1 - P_r) \tag{4}$$

where P_p and P_r are probability for decay events from the intermediate and Rydberg states. We assume the resonance of the rydberg state and the ground state with the AC stark shifts. We can derive that the detuning Δ to achieve the maximal fidelity should be:

$$\Delta^2 \approx \frac{\gamma_p}{\gamma}(\Omega_1^2 + \Omega_2^2)/8 \tag{5}$$

where γ_p is the decay rate of the intermediate state and γ is the decay rate of the rydberg state.

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Numerical results

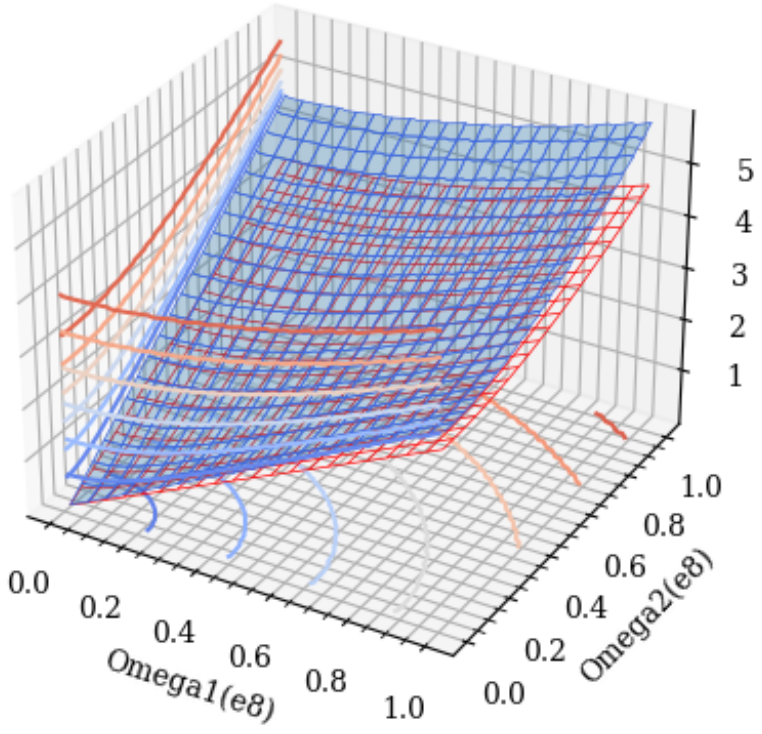

Figure 11: Blue line: numerical value of the optimal Δ. Red line: Analytical results.

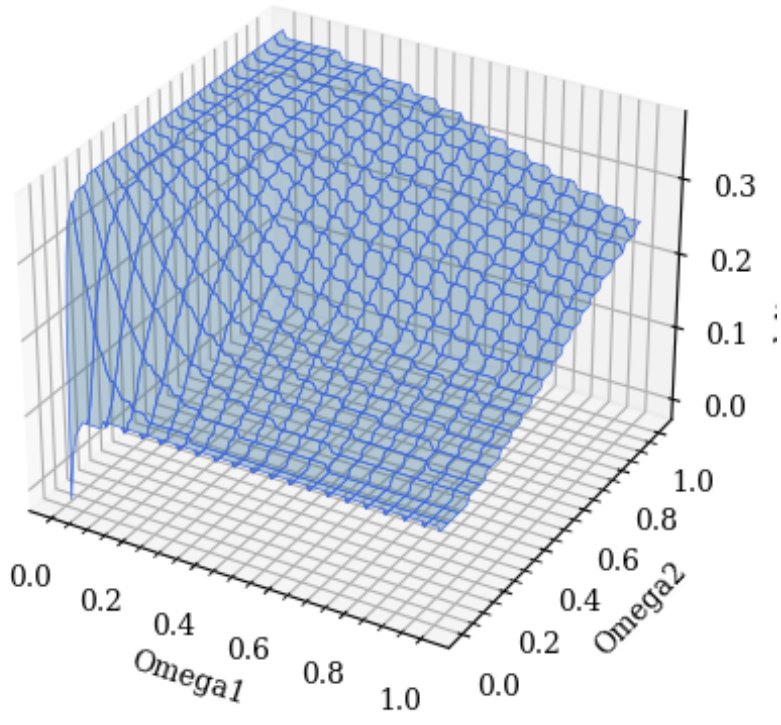

Figure 12: Fractional difference between our analytical results and numerical results

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Circular Rydberg States

Circular Rydberg states are states of maximal angular-momentum in the sense that $l = |m_l| = n - 1$. The wave function has the form [5]:

$$\psi_{nc}(r,\theta,\phi) = \frac{1}{\sqrt{\pi a_0^3}} \frac{1}{nn!} \left(-\frac{r}{na_0} \sin\theta e^{i\phi}\right)^{n-1} e^{-r/na_0} \tag{6}$$

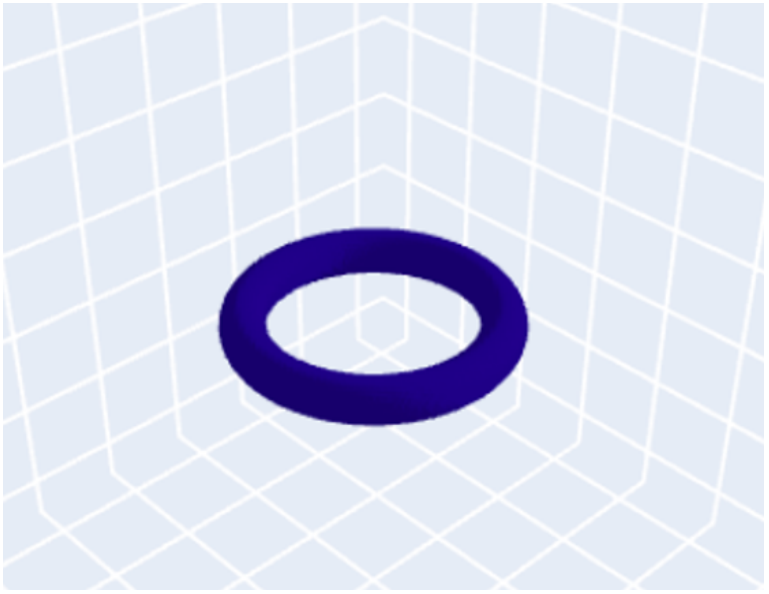

Figure 13: Rb n=50 circular state in an electric field

Rydberg atoms in DC electric field

Due to the degeneracy in the coulomb potential, we can only prepare the circular Rydberg state in an external electric field when this degeneracy is lifted [6]. Hence we have to introduce a Stark shift term into our Hamiltonian.

$$\hat{V}_s = q_e F_z \hat{z} \tag{7}$$

We see that m is still a good quantum number, but l and n are not. States with different ns and ls are mixed. We need to find new good quantum numbers to label our new state in the field.

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Parabolic quantum numbers

Instead of the conventional spherical coordinates (r, θ, ϕ), we can use parabolic coordinates (ξ, η, ϕ) to solve the Schrodinger equation [7], where $x = \sqrt{(\xi\eta}\cos\phi$, $y = \sqrt{\xi\eta}\sin\phi$, $z = \frac{1}{2}(\xi - \eta)$.

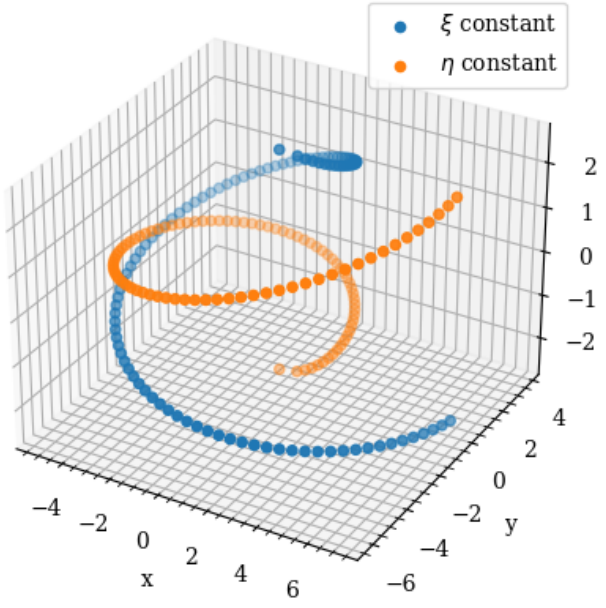

Figure 14: Constant η and constant ξ paraboloids.

In this coordinate system, we obtain three quantum numbers n_1, n_2 and m. It can be proved these three are good quantum numbers with a z-directed electric field. It is noted that $n = n_1 + n_2 + |m_l| + 1$. The wavefunction can be separated into three parts:

$$\psi = f_{n_1}(\xi) f_{n_2}(\eta) e^{im\phi} \qquad (8)$$

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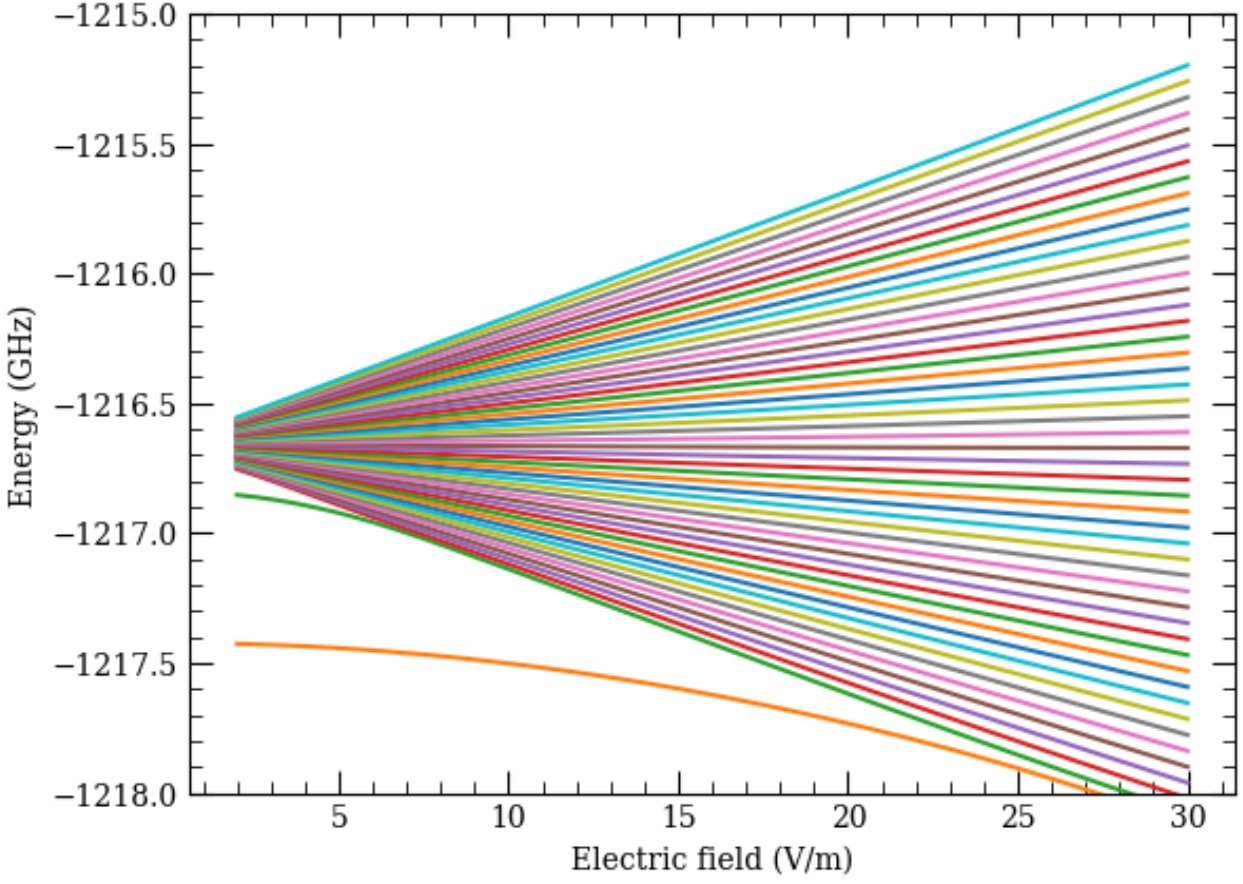
Stark Map of Rb 87

We calculated the stark map for Rb87, we choose the state with m=2 and n=52. The lowest two lines are originally 52G and 52F respectively.



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"Good" quantum numbers

From calculations, we notice that if the electric field is not too large (we will usually use E field less than $500 V/m$), states with different ns are not mixed too much (less than 1%). Also the splitting of different spins will also be small. Hence we choose the principal quantum number n, magnetic quantum number m, one of the parabolic quantum number n_1 as our "good" quantum number to label the states in an external electric field.

In terms of the new quantum numbers, we can write down the first-order stark shift as

$$E^{(1)}_{n,k,m_l} = \frac{3}{2} q_e a_0 n (n_2 - n_1) \times E \tag{9}$$

We see the equal spacing between the two neighbour states with the same m or same n_1. This equal spacing is very important in the preparation of circular states!!!

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Diamond Manifolds

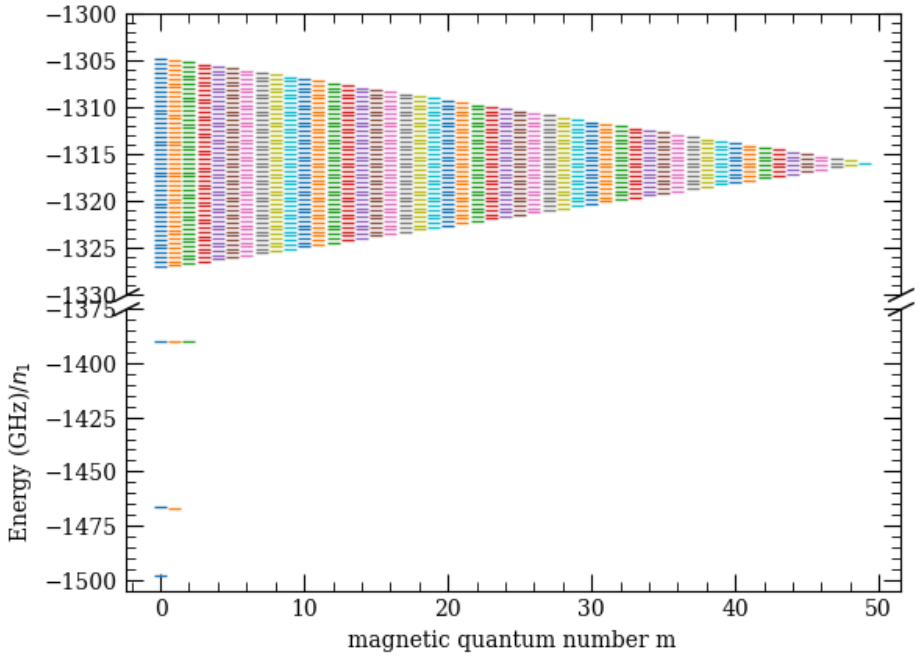

Figure 16: All the Rb states with n=50 in an electric field of 240 V/m. For small m=0,1,2, energies are not equally spaced due to the quantum defects.

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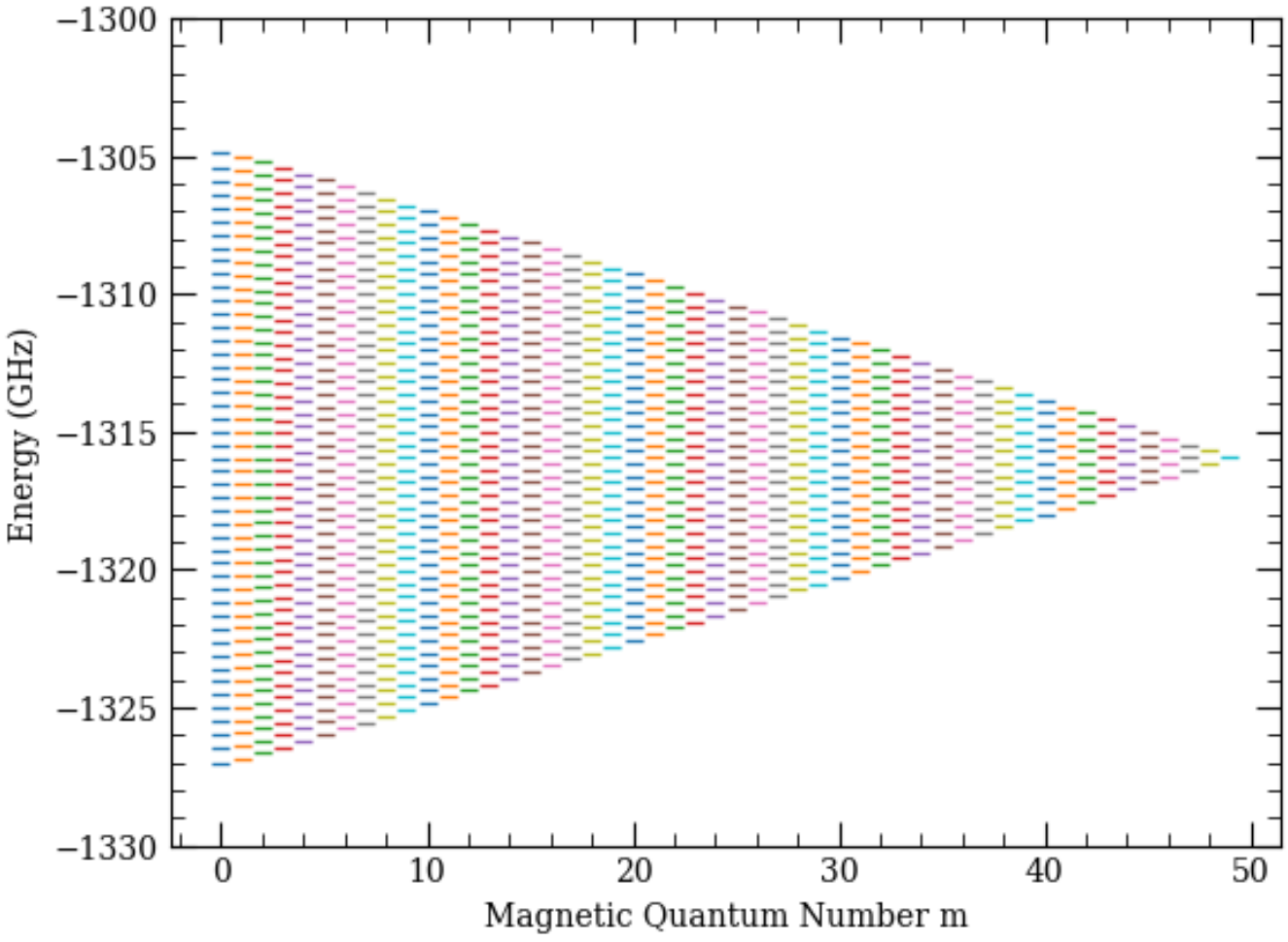
Diamond manifolds



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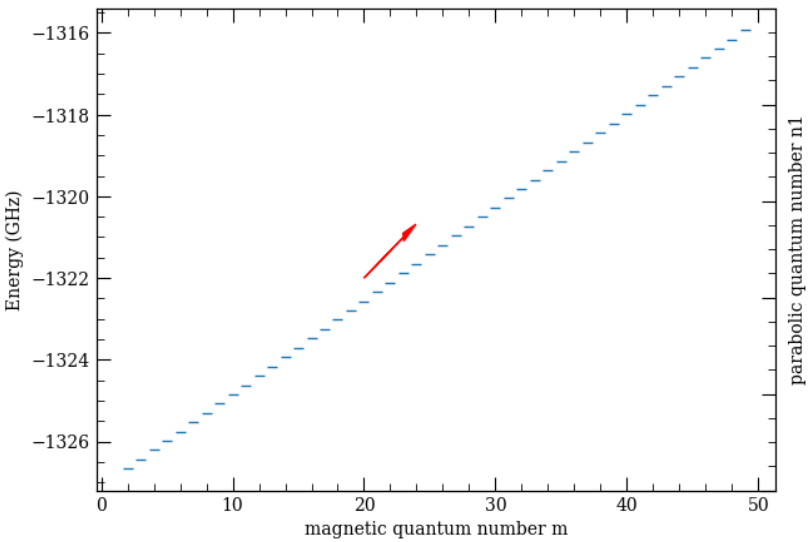
ARP path

We have to find a path through which we can transform our population from low m states to high m states. And an ideal choice is the states with $n_1 = 0$. And note that we start from m=3 [8].



We choose to drive the population through the bottom of the diamond manifold. We apply a laser with a frequency equal to the transition frequency at a certain electric field. (We choose 250V/m in this case)

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Transition Frequency and Coupling

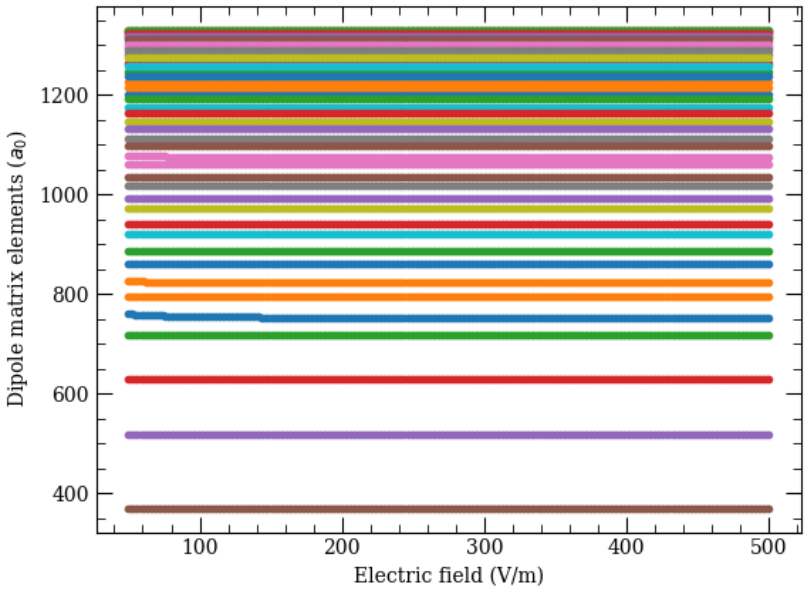

Figure 17: Coupling between neighboring states. Note the magnitude of the coupling are not in order with the state number.

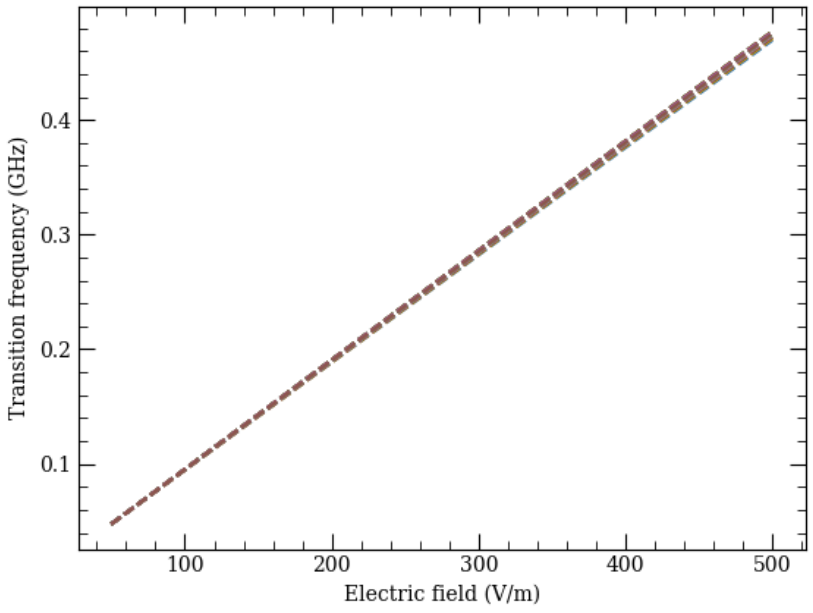

Figure 18: Transition frequencies are approximately the same due to the equal spacing.

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Dressed-State Picture

Now the whole Hamiltonian is periodic in time. We can use Shirley Floquet Hamiltonian [9] to construct a time-independent infinite dimension hamiltonian. We can obtain an effective hamiltonian:

$$\begin{bmatrix} 0 & \frac{\Omega_{01}}{2} & 0 & 0 & \cdots \\ \frac{\Omega_{01}}{2} & \omega_1 - \omega & \frac{\Omega_{12}}{2} & 0 & \cdots \\ 0 & \frac{\Omega_{12}}{2} & \omega_2 - 2\omega & \frac{\Omega_{23}}{2} & \cdots \\ 0 & 0 & \frac{\Omega_{23}}{2} & \omega_3 - 3\omega & \cdots \\ \vdots & \vdots & \vdots & \vdots & \end{bmatrix}$$

Diagonalise this matrix we can obtain the dressed states and ac stark shift.

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Simulation results

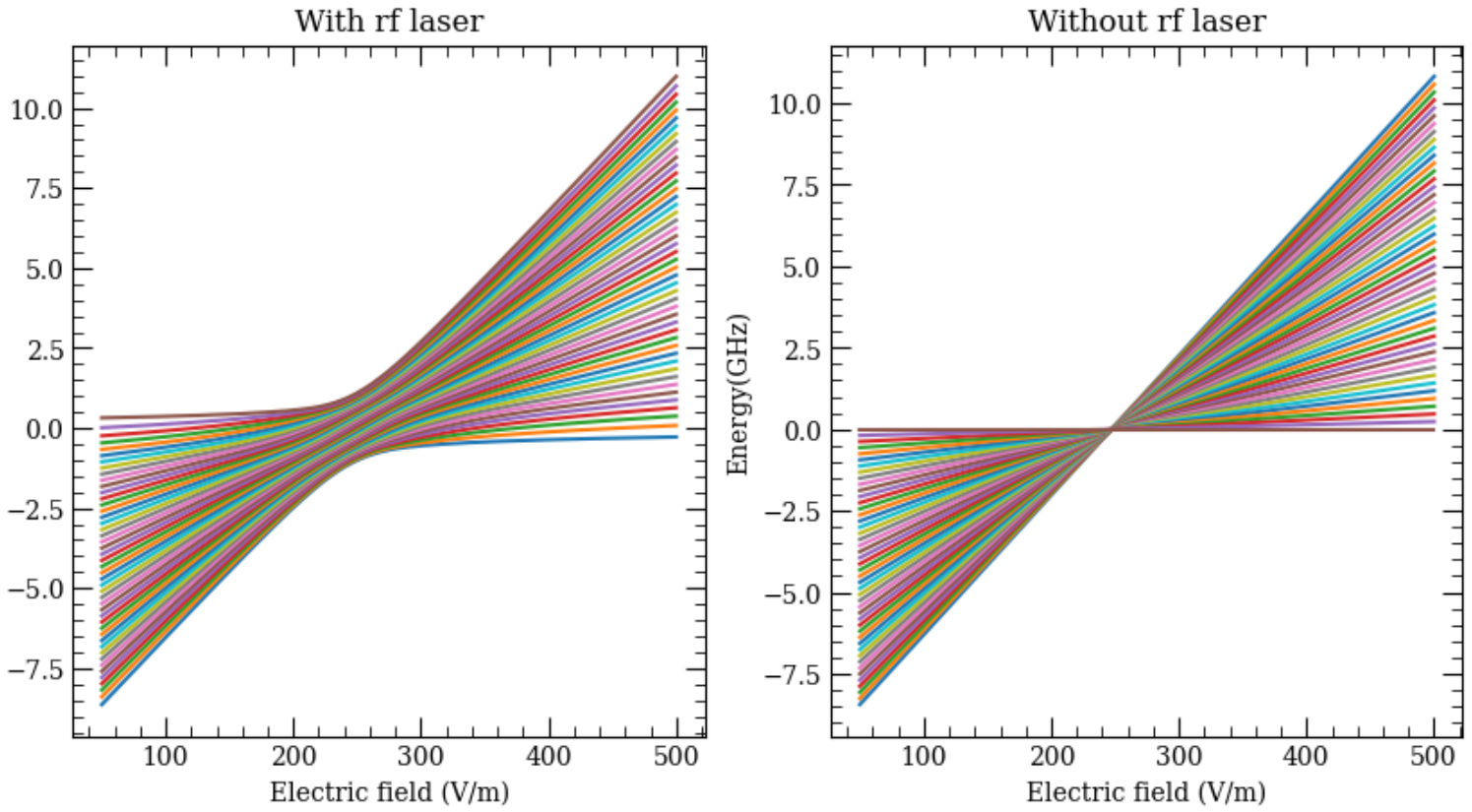

Figure 19: AC stark shifts

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Wavefunction

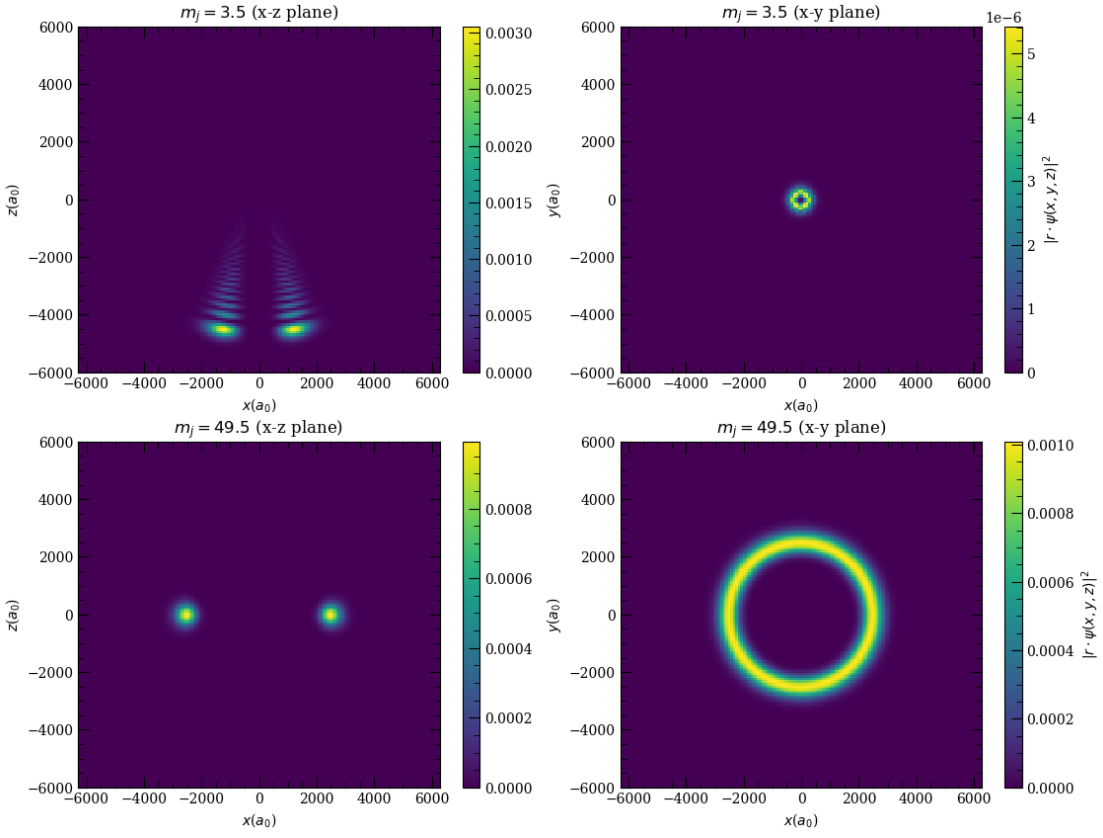

Figure 20: Wavefunctions of low m state and circular state

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Non-Hermitian Physics

So far for the preparation of the circular rydberg state, we only considered the hermitian hamiltonian. Can the non-hermitian processes (decay or dephasing) affect the whole Landau-Zener transition ? One paper shows that in some special cases, e.g. for the PT symmetric system, non-hermitian terms will have non-perturbative effects on the evolution of eigenstates [10].

$$P_{00} = P_{11} = \frac{1}{2 - e^{-\frac{\pi\gamma^2}{\alpha}}} \tag{10}$$

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Unitary transform

What is the unitary transform that maps the spherical states to parabolic states? What symmetry is preserved in the parabolic coordinates in the presence of an electric field?

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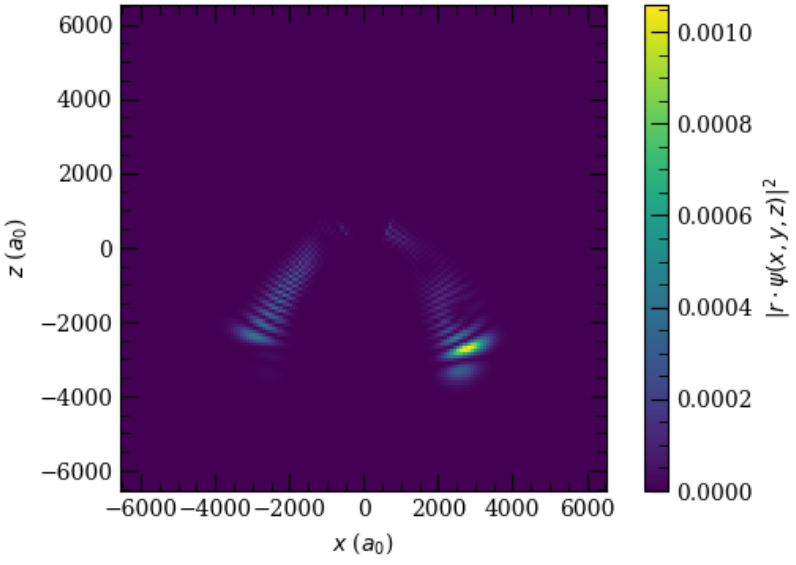
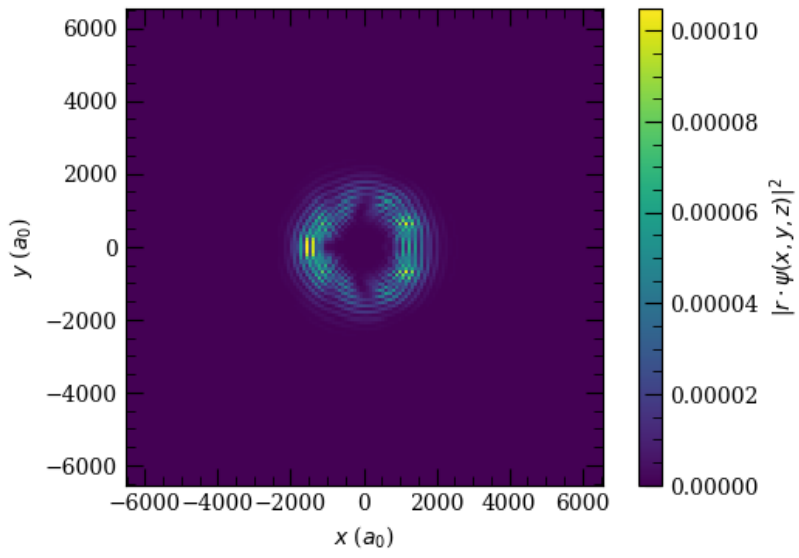
Wavefunction of the dressed state?

We then calculated the wave function of the dressed state; we find the symmetry is broken in x-y plane? A permanent dipole is induced?





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Computational Tools

Many of our codes are from ARC, which is a useful package to calculate rydberg atom properties such as lifetime, energy levels and transition rates.

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Acknowledgement

I would like to thank Prof. Mike Tarbutt, Prof. Ben Sauer and Dr. Kai Voges for providing help and guidance throughout the project.


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