# MODELING POPULATION DYNAMICS OF INCOHERENT AND COHERENT EXCITATION

Hannah Killian & Brandon Rozek
Advisor: Dr. Hai Nguyen
University of Mary Washington Department of Physics

## Abstract

In the three level atomic system, incoherent excitation leads to approximately one third of the population transferred from the ground state to the uppermost excited state. The objective of this study was to maximize the population transfer to the highest energy level while minimizing the amount of population in the intermediate state. Using the properties of coherent light in a two-photon system, theoretical calculations were performed with the time dependent Schrödinger equation. By exploiting the orthonormality of the wave function and applying the near resonance, dipole, and rotating wave approximations, a model was developed to show population dynamics for a 3-level $Rb^{87}$ system. Population transfer is driven by delayed laser pulses, and the model demonstrated that to maximize transfer the pulses should be in a counterintuitive order where the second laser field is applied prior to the first. Nearly complete population transfer was achieved from the ground state to the excited state using this method of coherent excitation.

## Introduction

All atoms have a quantum structure with discrete energy states. In order to change energy levels, the atom must absorb or emit a photon. The interaction of the atom's quantum system with light results in excitation of the atomic population. The goal of this research is to determine the conditions that would maximize population transfer in an atomic system while minimizing population in the transition state.

There are two types of light which can interact with an atom's quantum system. The first type is incoherent light, which is electromagnetic radiant energy of varying frequencies and phases, commonly observed in the form of a light bulb. The second type of light considered for excitation is coherent light, which is electromagnetic energy with a constant phase difference and the same frequency.

Excitation can be implemented in the configuration demonstrated in Figure 1, commonly referred to as the "ladder" system. Levels |1> & |2> and |2> & |3> are connected by dipole matrix elements, while levels |1> & |3> are not. The $\Omega_1$ and $\Omega_2$ represent the light sources exciting the population in |1> and |2> respectively.[1]

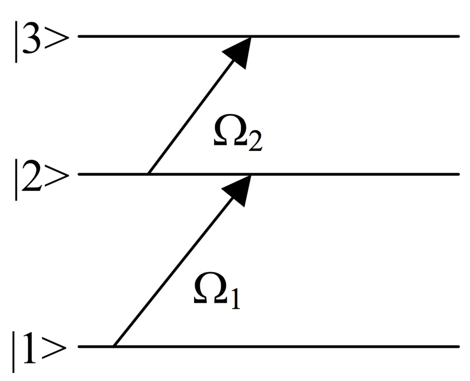


**Figure 1**: The three level atomic "ladder" system.[1]

A pump laser, represented in Figure 1 as $\Omega_1$ , is applied to the ground state and excites the population to the next level. In the $Rb^{87}$ system, the wavelength of the pump laser is 780.3 nm. The second pulse, $\Omega_2$, is produced by the Stokes laser which excites the population to the third state at a wavelength of 1528.99 nm.[2] The order of these two pulses is considered to be "intuitive" if $\Omega_1$ precedes $\Omega_2$. Counterintuitive pulse order is when $\Omega_2$ is applied before $\Omega_1$. The technique of stimulated Raman adiabatic passage (STIRAP) utilizes the counterintuitive pulse order when transferring population between three or more coupled energy levels.[2] In this research, the objective was to create a model to test the effectiveness of STIRAP.

## Incoherent Light

In the 3-level ladder system, $I_1$ and $I_2$ represent the intensities of the incoherent light, and $A_{32}$ and $A_{21}$ represent the spontaneous emission loss from energy levels |3> to |2> and |2> to |1> as shown in Figure 2. The population can be represented mathematically as a series of differential equations:

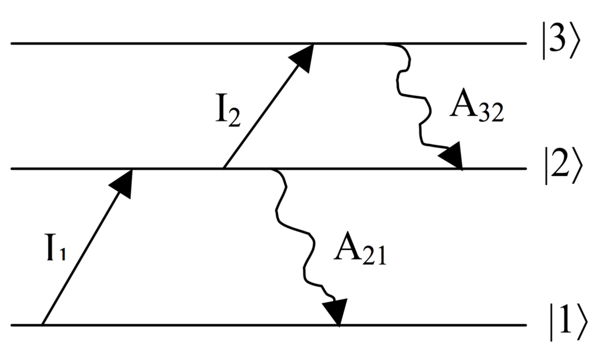


**Figure 2**: The three level incoherent system excited by incoherent light in the form of intensities $I_1$ and $I_2$ which also contains spontaneous emission $A_{32}$ and $A_{21}$.[1]

$$\begin{cases} \dot{n}_1 = n_2A_{21} + n_2BI_1 - n_1BI_1 \\ \dot{n}_2 = -n_2A_{21} - n_2BI_1 + n_1BI_1 - n_2BI_2 + n_3BI_2 + n_3A_{32} \\ \dot{n}_3 = n_2BI_2 - n_3BI_2 - n_3A_{32} \end{cases}$$

Upon solving for $n_3$, the population in the excited state, and applying the approximation in which the intensities are much greater than spontaneous emission:

$$\lim_{BI>>A} n_3 = \lim_{BI>>A} \frac{1}{\frac{A}{BI} + 2\frac{A^2}{B^2I^2} + 2\frac{A}{BI} + 1} = \frac{1}{3}$$

We obtained that the maximum population transferred in the incoherent system is one third of the initial amount in the ground state.

## Coherent Light

For the three level ladder system we make use of the coherent property of light, setting spontaneous emission A = 0. It is assumed that population is conserved between levels. The time-dependent Schrödinger equation (TDSE) is represented in the first two equations, where the latter is Hermitian. The third equation represents the wave function, where $\Psi$ is a solution to the TDSE and $\psi$ satisfies the time-independent Schrödinger equation (TISE).

$$i\hbar\frac{\partial\Psi}{\partial t} = H'\Psi \qquad H' = H_0 + \hat{V}(t) \qquad \Psi(t) = \sum_n c_n(t)\psi_n e^{-i\omega_n(t)}$$

In order to determine the amount of population transferred to the uppermost excited state, we must solve for the $c_n$ term which is the constant coefficient for the transfer. By squaring this term, the probability of exciting from the ground state to the uppermost excited state can be found. Solving for the $c_n$ term involves applying several approximations. First the wave function can be simplified by exploiting its orthonormality, and then the near resonance approximation can be applied where the number of levels are truncated to three, as we are considering a three level system. Next is the dipole approximation, where the electric field is considered to be spatially uniform with respect to the atom. The Rabi frequency governs the coupling between light and the atomic transition. It can be defined as the frequency of oscillation for an atomic system. The model used $Rb^{87}$ whose Rabi frequencies are defined as the following, where $\tau$ is the delay between pulses and $\omega$ represents the Gaussian pulse width.

$$\Omega(t)_{s,p} = \Omega_{0s,p}e^{-\ln(4)\left(\frac{(t-t_0)\pm\frac{1}{2}\tau}{w}\right)^2}$$

Finally, the rotating wave approximation is made by ignoring terms that oscillate at a frequency much higher than the Rabi frequency. The result is a matrix definition for the time derivative of the constant coefficient. The model uses this matrix along with the density matrix to create a series of coupled differential equations. Upon solving these numerically, the population of each state can be determined.

$$\dot{c} = -\frac{i}{2}\begin{bmatrix} 0 & \Omega_1 & 0 \\ \Omega_1 & 2\Delta_2 & \Omega_2 \\ 0 & \Omega_2 & 2\Delta_3 \end{bmatrix} c$$

## Simulation Results

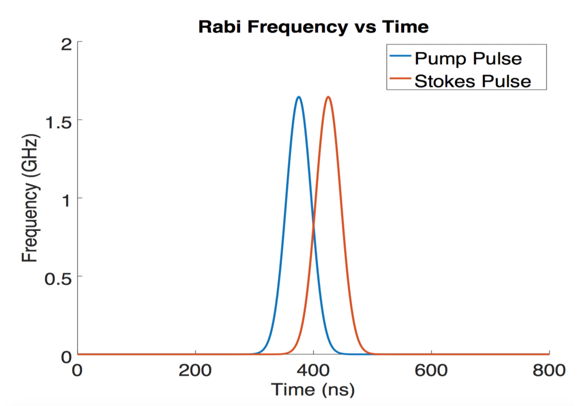


**Figure 3**: Rabi Frequency with respect to time in intuitive pulse order.

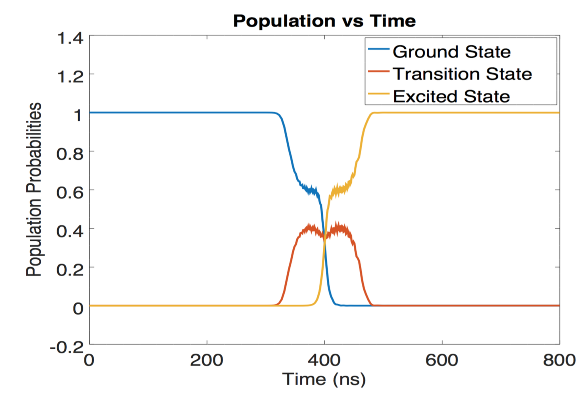


**Figure 4**: Population densities with respect to time in intuitive pulse order, neglecting spontaneous emission.

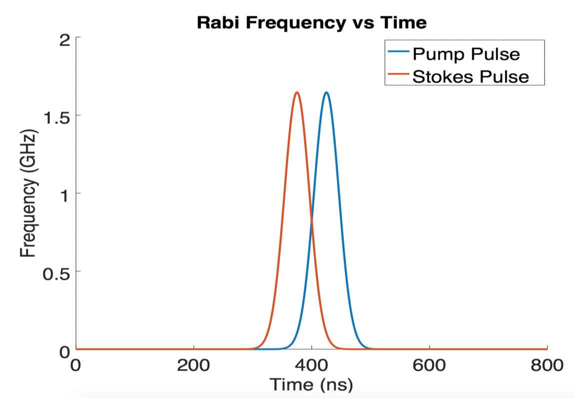


**Figure 5**: Rabi Frequency with respect to time in counterintuitive pulse order.

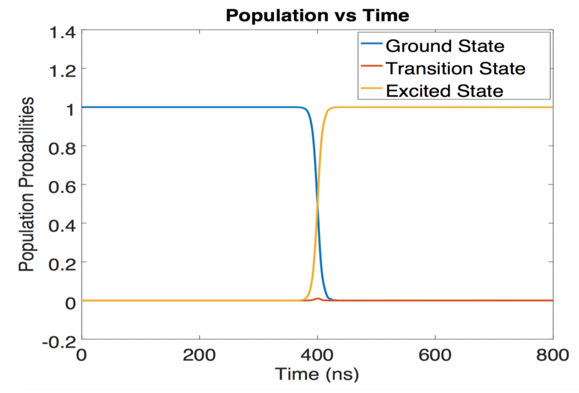


**Figure 6**: Population densities with repsect to time in counterintuitive pulse order, neglecting spontaneous emission.

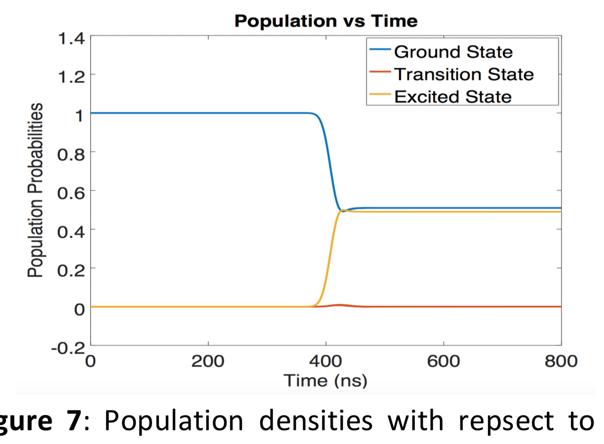


**Figure 7**: Population densities with repsect to time when photon detuning is increased by a factor of 20.

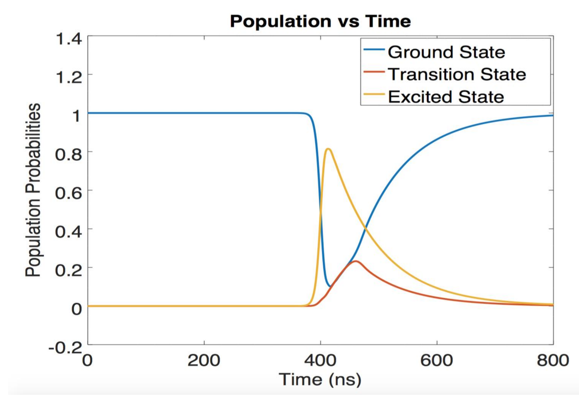


**Figure 8**: Population densities with respect to time in counterintuitive pulse order with spontaneous emission.

## Simulation Analysis

In the model for coherent excitation three main parameters were considered: pulse delay, photon detuning, and spontaneous emission. Pulse delay is the time in nanoseconds between the pump and stokes laser. Photon detuning is a technique that involves detuning the laser below the resonance frequency. Spontaneous emission occurs as an atom in an excited state releases a photon and transitions down to a lower, more stable energy level.

We first consider the pulse delay. In intuitive order, the pump laser is applied prior to the Stokes laser as shown in Figure 3. The population graph in Figure 4 demonstrates that the population is transferred from the ground to the excited state completely. It takes approximately 200ns to achieve complete transfer, and during that time about half of the population occupies an intermediate state. In counterintuitive order, as displayed in Figures 5 and 6, the Stokes laser is applied before the pump laser. In this case, the population is completely transferred in less than 50ns with minimal occupancy of the intermediate state.

In the figures discussed above, the photon detuning was set at 53MHz below resonance, which is the experimentally determined optimal frequency for $Rb^{87}$.[2,3] In Figure 7, this parameter was set at 20x the standard experimental frequency, which caused only half of the population to transfer to the excited state.

Thus far the model has not addressed the reality of spontaneous emission. Figure 8 illustrates its effect on a coherent system with counterintuitive pulse order. The population is nearly immediately transferred into the excited state, but then exponentially decays back to the ground state.

## Applications

The model described a three level $Rb^{87}$ system; however, it can be generalized to apply to any atomic system by changing the values for the Rabi frequency and photon detuning, and the decay rates for spontaneous emission. Additionally, the model could be expanded to include more than three levels.

The theoretical calculations for coherent excitation and the results of the model can be confirmed experimentally. The experimental setup involves trapping the $Rb^{87}$ atoms in a magneto-optical trap (MOT). The atoms are then excited by a series of coupled lasers. Using the STIRAP method of coherent excitation the Stokes laser is first applied, followed by the pump laser in counterintuitive order. The population can be measured in several ways, one of which being through recoil ion momentum spectroscopy (RIMS).[4]

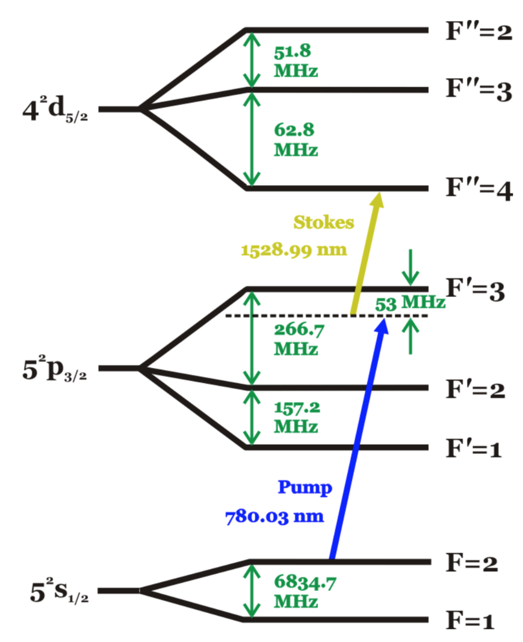


**Figure 9**: The 3-level coherent excitation system for $Rb^{87}$ with experimental values.[2]

## Conclusion

From the theoretical calculations concerning incoherent and coherent excitation, we concluded that the latter leads to a near complete population transfer from the ground state to the uppermost excited state. We then modeled the coherent system, manipulating several of the parameters including pulse delay, photon detuning, and spontaneous emission. It can be concluded that the STIRAP system with counterintuitive pulse order transfers nearly all of the population while minimizing the intermediate state.

## References

[1] DePaola, B. *Notes for Spring 2003 AMO Course: Lectures on Coherent Excitation* [PDF document].

[2] H.A. Camp, Ph.D. thesis, Kansas State University, 2005.

[3] Daniel A. Steck. *Rubidium 87 D Line Data*, 2001.

[4] H. Nguyen , Ph.D. thesis, Kansas State University, 2003.

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