Stimulated Raman Adiabatic Passage (STIRAP)

Quantum Information of Atoms and Photons

Marco Tavis Foster

15/07/2025

Introduction

Population transfer between individual, discrete quantum states is one of the most fundamental processes in quantum mechanics, with many different methods being introduced in order to achieve coherent and controlled transitions. One issue that needs to be contended with is that of spontaneous emission of a photon from excited states, resulting in the excited state transitioning into another in an uncontrolled manner - potentially one not relevant to our process of interest. Spontaneous emission of a photon can occur in all states but we are concerned with that from excited states in particular as they have a much shorter lifespan than ground states. It is desirable to develop a method to transfer populations between two states without the loss of population to other states or sub-levels through emission.

Stimulated Raman Adiabatic Transfer (STIRAP) resolves this problem. It operates with two laser beams that govern the transitions between three states, such as |0> → |e> and |e> → |1>, where |0> and |1> are sub-levels of the ground state and |e> is an excited state (3-level Λ-system). STIRAP completes the transition from |0> to |1> without ever populating the ‘leaky’ excited state, thus achieving (close to) total population transfer. This is achieved through ‘adiabacity’, the idea that a slow-acting perturbation (such as laser light on the atoms) will not affect energy level populations - states initially occupying an energy eigenstate will remain within that eigenstate as the system evolves. This means that as long as we keep the evolution of the perturbations slow we can keep our population within a specific eigenstate - this factor is crucial, as we shall see later. Coherent energy transfer is important for fields such as quantum optics or computing that rely on systems to remain predictable.

Theory

Landau-Zener problem

We begin by looking at the Landau-Zener problem; modelled as a two-level system with time dependent energy bias ω/2 and constant coupling δ. The Hamiltonian of the system is described as:

$$H = \delta\sigma_z + \frac{\omega}{2}(\sigma_+ + \sigma_-) = \begin{bmatrix} \delta & \frac{\omega}{2} \\ \frac{\omega}{2} & -\delta \end{bmatrix} \tag{1}$$

For this Hamiltonian there is a minimal energy gap of ω at t = 0 when δ = 0. The region around this point is known as the avoided level-crossing.

In the adiabatic case (where perturbations are slow acting) the Hamiltonian is perfectly diagonalised:

$$\hat{H}(t) = \begin{bmatrix} E_+(t) & 0 \\ 0 & E_-(t) \end{bmatrix} \tag{2}$$

The energy levels, $E_\pm(t)$, and eigenstates, $|E_\pm(t)>$, can be obtained from the Schrödinger equation:

$$E_\pm(t) = \pm\sqrt{\left(\frac{\omega}{2}\right)^2 + \delta^2} \tag{3}$$

This eigenvalue evolves with time, meaning that if we evolve the system adiabatically we can remain in the same eigenstate but still end up populating different energy levels. When dealing with the Landau-Zener problem the question we want to solve is therefore the probability, P_e, of beginning in the ground state and ending in the excited state in the adiabatic regime.

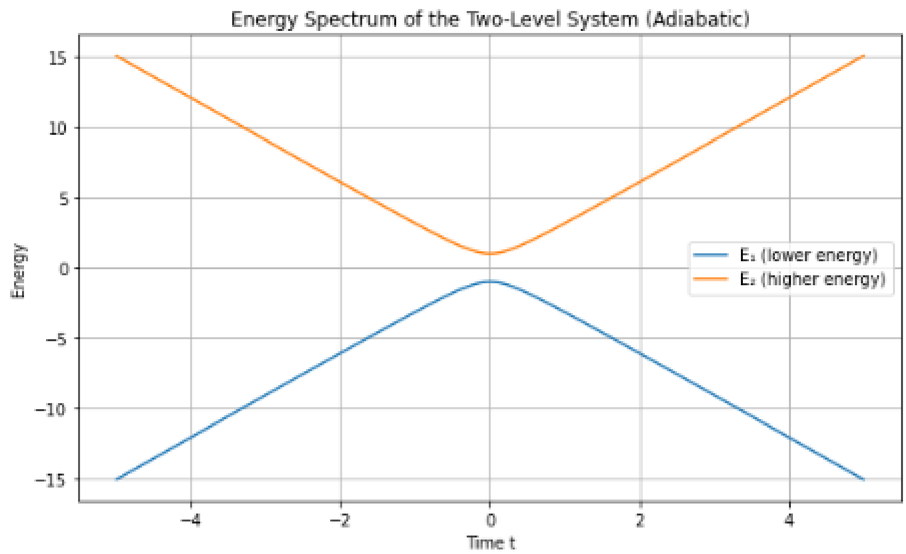


Fig. 1: Adiabatic passage of the two-level system. The avoided level-crossing can be seen at t = 0, with separation ω.

<u>*Application to STIRAP*</u>

The Landau-Zener problem shows us how we can use the adiabatic theorem to stimulate transitions between energy levels while remaining in the same eigenstate. We will see how we can extend this to STIRAP.

First we will discuss how the STIRAP protocol works. We have stated that we are working with three energy levels, |0>, |e> and |1>, and we use two lasers to stimulate the transitions between them, the Pump laser for the |0> → |e> transition and the Stokes laser for the |e> → |1> transition, and having the population entirely inhabit the |0> energy level (with the aim to populate |1> as coherently as possible).

The protocol begins, rather counter-intuitively, by first turning on the *Stokes* laser. We then slowly turn on the Pump laser, slowly turn off the Stokes laser, and then finally slowly turning off the Pump laser again. This does not seem to make much intuitive sense but we will soon see the result of such a protocol and the impact it has on the evolution of the system.

We begin by analysing the Hamiltonian that is formed as a result of this protocol:

$$\mathrm{H}(t) = \frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_P(t) & 0 \\ \Omega_P(t) & 2\Delta_P & \Omega_S(t) \\ 0 & \Omega_S(t) & 2(\Delta_P - \Delta_S) \end{bmatrix} \tag{4}$$

Where $\Omega_P(t)$ and $\Omega_S(t)$ are the pulsed Rabi frequencies of the Pump and Stokes lasers and Δ_P and Δ_S are the detunings of each laser from their respective transitions. The off-diagonals show the coupling strength between states, while the diagonal elements show the detuning from the intermediate state or the two-photon resonance. For STIRAP it is essential that $\Delta_P = \Delta_S$ so that there is no two-photon detuning.

The eigenstates for the Hamiltonian in (4), written in terms of the energy eigenstates |0>, |e> and |1>, are:

$$\begin{aligned} |a_1\rangle &= \sin\Theta\sin\Phi\,|0\rangle + \cos\Phi\,|e\rangle + \cos\Theta\sin\Phi\,|1\rangle \\ |a_0\rangle &= \cos\Theta\,|0\rangle - \sin\Theta\,|1\rangle \\ |a_2\rangle &= \sin\Theta\cos\Phi\,|0\rangle + \sin\Phi\,|e\rangle + \cos\Theta\cos\Phi\,|1\rangle \end{aligned} \tag{5}$$

Quite strikingly we see that $|a_0>$ has no contributions from the lossy excited state |e>. In fact, if we were to apply the adiabatic theorem that we used during the Landau-Zener problem we predict that we could remain in the $|a_0>$ state and successfully transfer the population from |0> to |1> - without ever occupying the state |e>. The state $|a_0>$ is therefore known as the ‘dark state’.

Let’s analyse the significance of Θ (ϕ can be ignored since it does not contribute to the dark state). Θ is the mixing angle and is the ratio between the Pump and Stokes Rabi frequencies:

$$\tan\,\Theta = \frac{\Omega_P(t)}{\Omega_S(t)} \tag{6}$$

Θ can therefore be controlled through varying this ratio. This can be achieved by first turning on the Stokes laser, slowly turning on the Pump laser and turning off the Stokes laser, before finally turning off the Pump laser - as laid out in the protocol. The effect, over time, is that:

- When just the Stokes laser is on $\Omega_P(0) = 0$, meaning that the mixing angle will also be zero and the dark state in Eqn. 5. will only have the |0> term (all atoms occupy the |0> state.
- As $\Omega_S(t)$ is slowly decreased and $\Omega_P(t)$ is slowly increased the effect is that, over time, atoms will transition directly from |0> to |1> as the mixing angle is changed smoothly from 0° to 90°.
- Finally, when just the Pump laser is on, the dark state will no longer have the |0> and all atoms will be occupying the |1> state. The Pump laser is slowly turned off.

Throughout this whole process the state vector will never acquire a contribution from the state |e> (except through imperfections in the adiabatic process).

Simulation

Plotting the Landau-Zener problem

The first diagram for the Landau-Zener problem is depicted in Fig. 1, which shows the case that the system follows the adiabatic passage. In Fig. 2 we see the diabatic passage, which is the case where $\omega = 0$ and the level-crossing is not avoided. In this regime the system does not follow the adiabatic eigenstates $|E_{\pm}(t)\rangle$ and instead follow the basis |0> and |1> states.

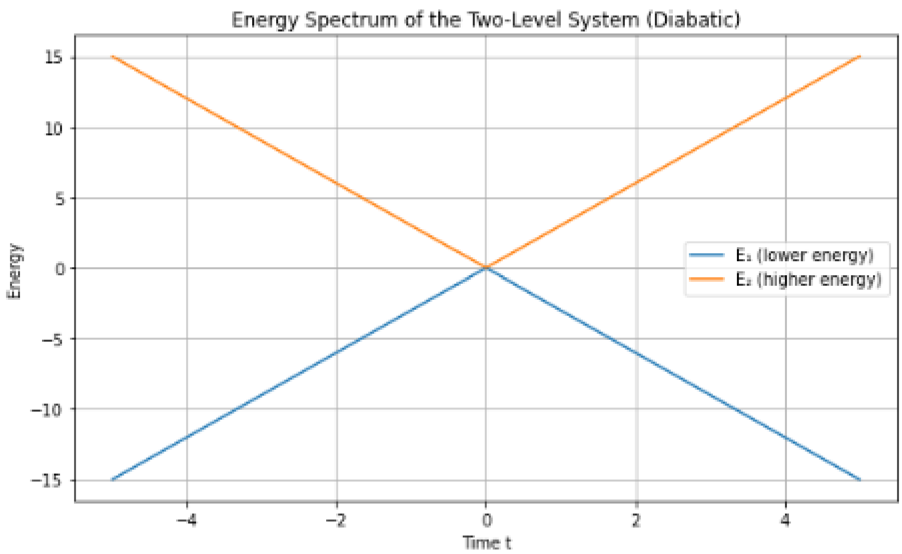


Fig. 2: Diabatic passage of a two-level system, where $\omega = 0$.

Compare numerical calculation vs. analytical

In the Landau-Zener problem we are interested in determining P_e, the probability that the system ends in the excited state given that it began in the ground state. Analytically, for large values of τ, this can be approximated as:

$$P_e \approx 1 - \exp(-\pi\Omega^2/(2\alpha)) \tag{7}$$

We're interested in comparing this with a numerical solution for P_e for different values of the parameters Ω, α and τ while keeping the other two constant.

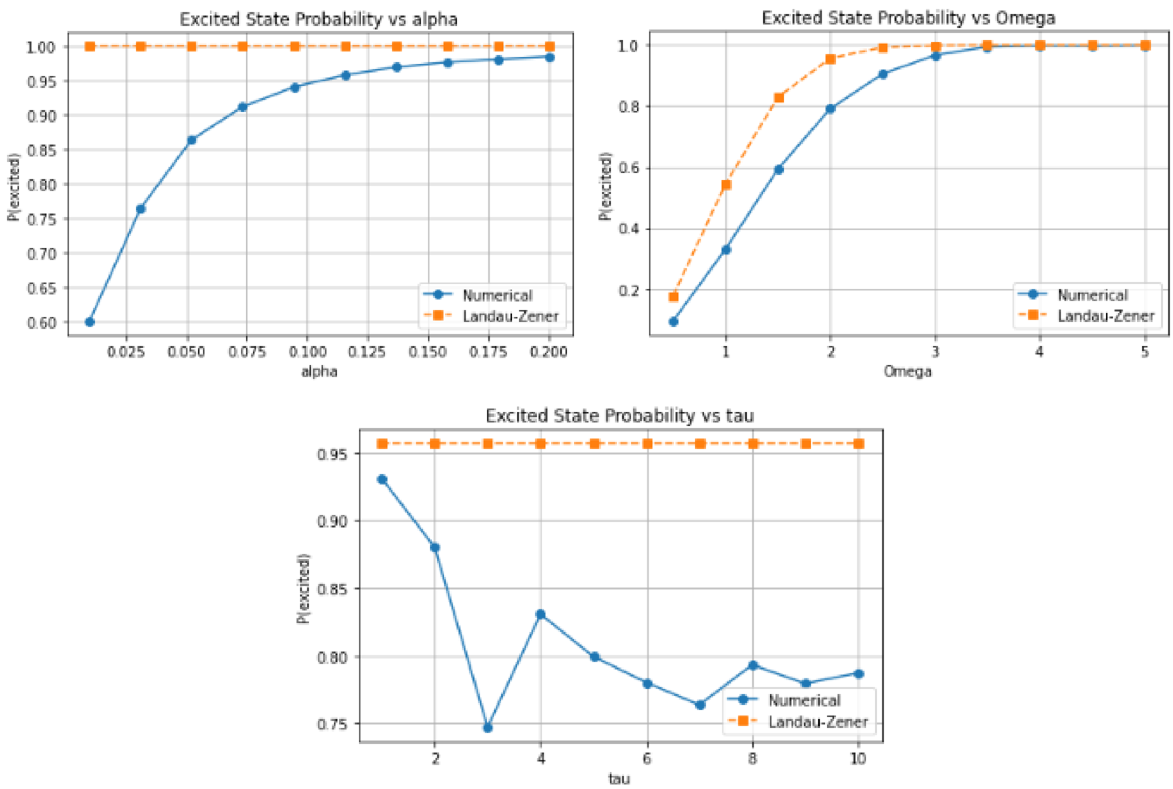


Fig. 3: Evolution of the analytical (orange) and numerical (blue) calculations when a) Ω is varied, b) α is varied, c) τ is varied.

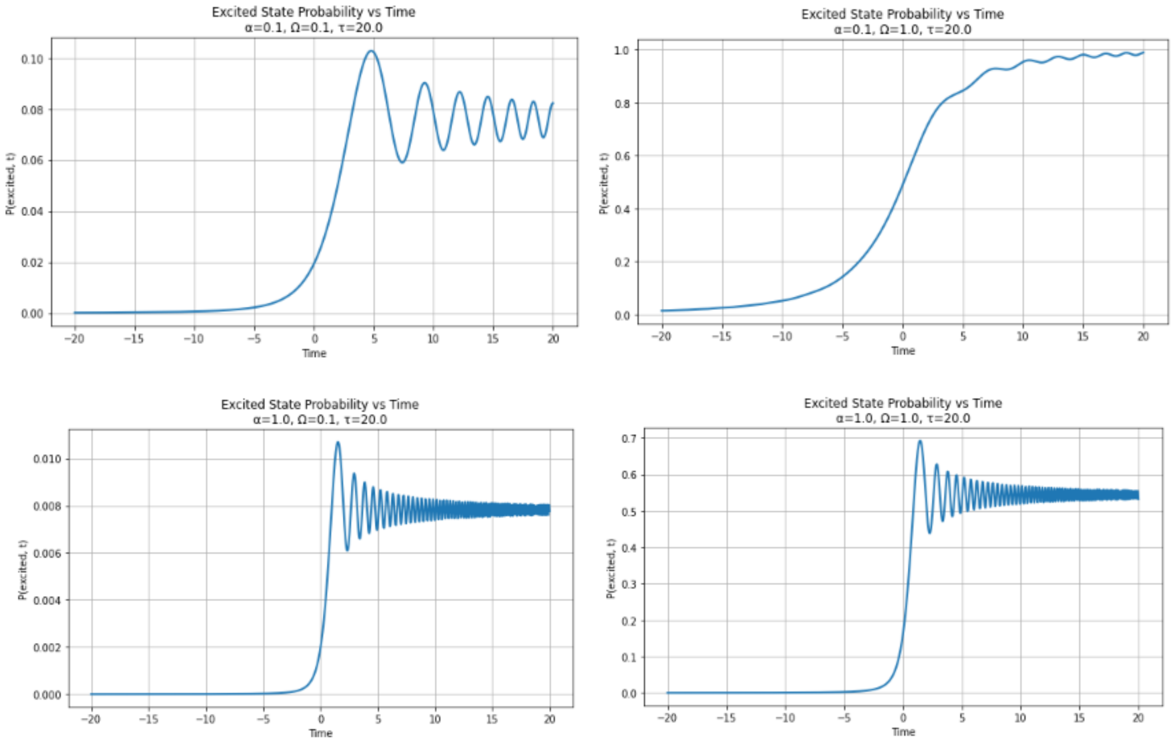


Fig.4: Time evolution of the analytical calculations for varying combinations of Ω and α (τ = 20 for every plot).

As expected we see that varying Ω and α have similar effects on the probability since they both impact the exponential seen in *(7)*. The analytical form of the probability remains a good approximation as long as Ω and α are not similar values - in Fig. 3b I purposefully have them set this way at the beginning so it can be shown the effect this has on both the analytical and numerical values.

Varying τ has a different effect. Our analytical expression for the probability seems unaffected by this variation - and thats because it is. The expression is not dependent on τ at all, instead it was assumed in the first place that τ is large and so we can't rely on the values we get for low τ. We see why in the numerical solution, which undergoes unpredictable fluctuations as τ is being varied, especially at the beginning.

Dark state evolution in STIRAP

We're interested in determining how the dark state evolves through time, taking into account contributions from the basis states |0>, |e>, |1>. We define the Hamiltonian as a function and find its eigenvalues/vectors using np.linalg.eigh. The dark state eigenvector can be found by observing the following:

$$\omega^{+} = \Delta_P + \sqrt{\Delta_P^2 + \Omega_P^2 + \Omega_S^2}, \qquad \omega^{0} = 0,$$
$$\omega^{-} = \Delta_P - \sqrt{\Delta_P^2 + \Omega_P^2 + \Omega_S^2}. \tag{7}$$

Which are the eigenvalues of the equations in *(5)*. We can see that the eigenvalue corresponding to the dark state is always zero, so if we find the minimum absolute eigenvalue (using np.argmin(np.abs(eigvals)) - once degeneracy between eigenvalues is lifted) we can use that to find the dark state and, from there, find the basis states it is composed of through time. We see this evolution in Fig. 5.

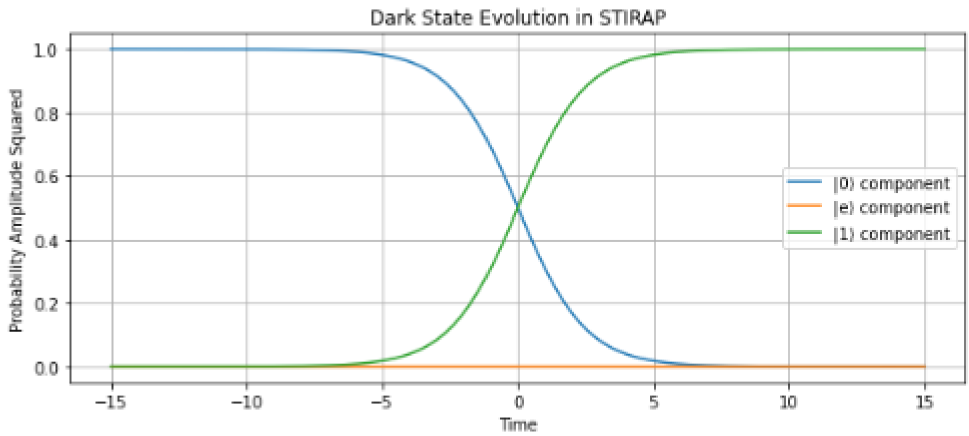


Fig. 5: Time evolution of the dark state of the STIRAP Hamiltonian, expressed in terms of percentage of contribution from each basis state.

Furthermore, *(7)* raises questions about the energy gaps between the eigenvalues of the STIRAP hamiltonian. The initial degeneracy will be lifted when the process starts so it is not harmful to the transition process. Regardless, we are interested in making sure that the gap remains large throughout the process.

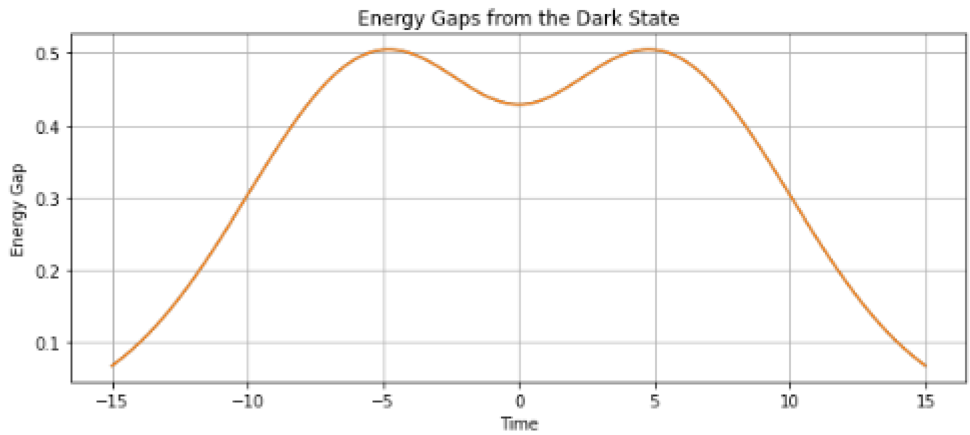


Fig. 6: Energy gap between the dark state eigenvalue and the two other eigenvalues of the STIRAP Hamiltonian.

<u>Population transfer</u>

Finally, we wish to see how the population of states changes as the STIRAP protocol evolves. We use our STIRAP Hamiltonian in the Schrödinger equation using scipy.integrate.solve_ivp. If ‘sol’ is the output of using this function then the population percentages can be found by getting psi_t = sol.y.T and extracting psi_t[:, a] (where a = 1, 2, 3 and represents the three different populations). The process is run from t = -5 to t =5.

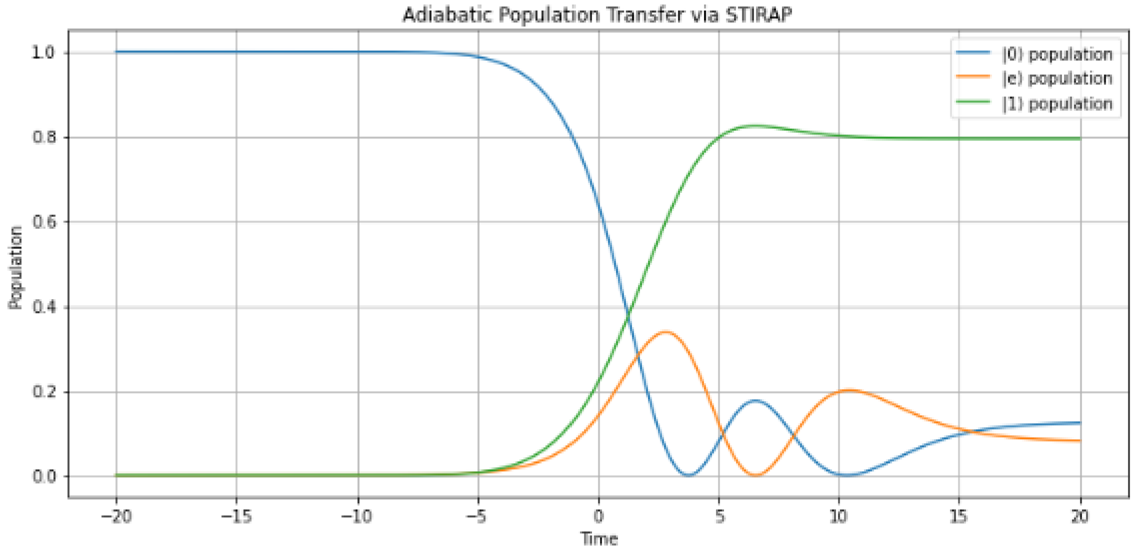
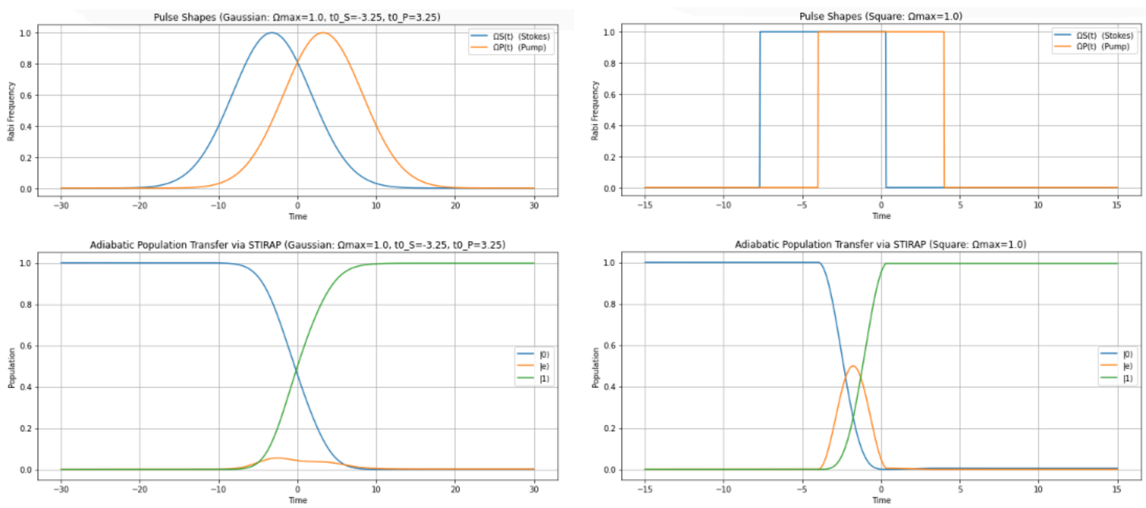


Fig. 7: Time evolution of population present at each basis state, with the STIRAP process occurring from t = -20 to t = 20.

We see an imperfect adiabatic transfer while the process is running, with 80% of the population ultimately ending up in the desired |1> state. The rest transitioned into the unwanted |e> state - which in a real experiment would quickly emit a photon and decay into either one of our relevant states or other states irrelevant to this process. This level of imperfection is due to a non-ideal delay occurring between the two lasers. We can see that this is the case in Fig.6, where the energy gap between eigenstates decreases in the middle of the process since the delay of the Stokes laser is too large. We correct it in Fig.8, as well as exploring the effects of other pulse shapes on the system evolution.



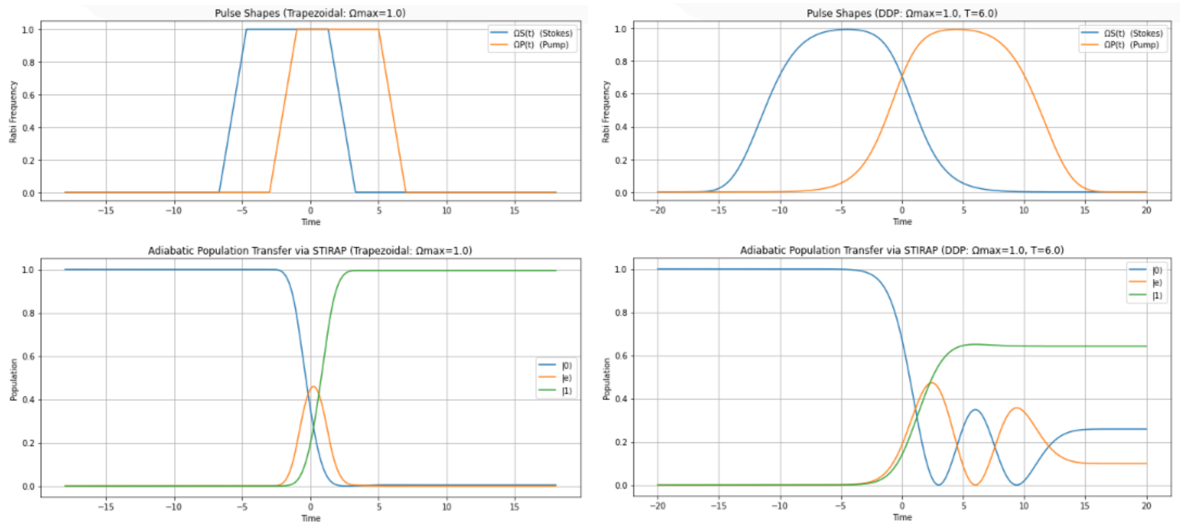


Fig.8: Time evolution of state populations for different pulse shapes. Top-left; gaussian, top-right; square, bottom-left; trapezoidal, bottom-right; DPP-optimised pulse shape

We see for the first three pulse shapes (gaussian, square and trapezoidal) that the gaussian results in far less error. The local adiabatic condition - that quantifies the smoothness required for the pulses to maintain adiabaticity - does not hold for the trapezoidal and especially the square case where the laser is abruptly turned on and off again. This results in non-adiabatic coupling and leakage into |e>.

The fourth shape is based on the Dykhne-Davis-Pechukas (DDP) method, which aims to resolve the issue that the gaussian pulse shape usually requires a large pulse area in order to evolve slowly enough to maintain adiabaticity. We can minimise the probability of leaking from the dark state by maximising the eigenenergy gap which is seen in Fig.6. The eigenvalues have energies given by (3), where $\frac{\omega}{2} = \Omega_{S,P}$. If we shape our lasers so that $\Omega_S^2(t) + \Omega_P^2(t) = const.$, then the adiabatic eigenvalue splitting will remain constant throughout the process and the gap to the bright states never closes. I had trouble recreating this myself so Fig.9. shows the results of Vasilev et al. (2009).

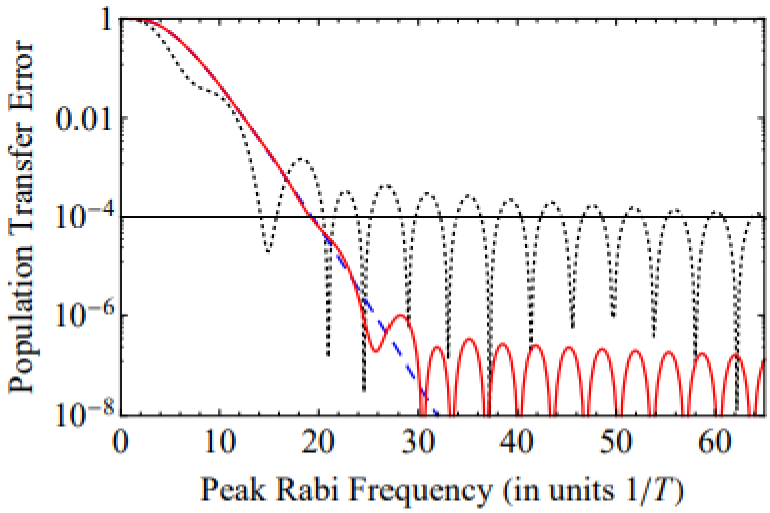


Fig.9: Comparison of the error in population transfer of the normal gaussian pulse shape (dotted curve) and the DDP-optimised pulse shape (solid curve).

We note that, as long as the system remains adiabatic, it remains robust against system imperfections such as laser intensity, pulse timing and pulse shape. This is due to the fact that the mixing angle *(6)* depends solely on the ratio of the Rabi frequency and not on any other factor, so as long as this ratio is controlled then the adiabaticity holds.

Finally we predict that to some degree the Landau-Zener theory can be applied. Although we are not operating in a two-level system and it does not utilise a dark state, if we wanted to simulate a perfectly adiabatic transfer with no contribution from |e> we can reduce this as a two-level system between only |0> and |1>.