Prof. Dr. Jean-Philippe Brantut
Dr. Rohit Prasad Bhatt
Francesca Orsi

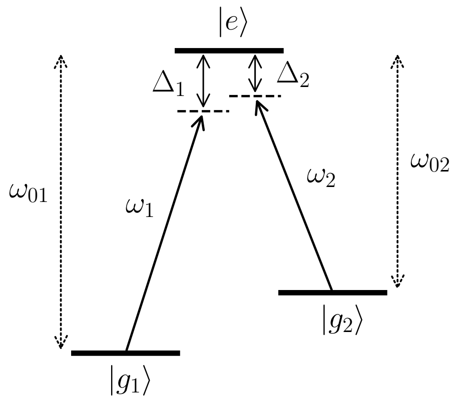
Quantum Optics 2 – Spring semester 2023 – 16/03/2023
Problem Set 4 : Dark States

For questions contact : francesca.orsi@epfl.ch

In this problem set we consider a three-level, Λ-type system illuminated by two optical fields. In the first part we will find a ***dark state*** in such a system, which does not couple to the other states by the applied fields. The second part deals with the stimulated Raman adiabatic passage (STIRAP), which enables us to transfer population from one quantum state to another coherently. This is possible by keeping the atoms in a dark state during the process.

I. COHERENT DARK STATES

We consider a Λ-type three-level system with the two ground states $|g_1\rangle$ and $|g_2\rangle$ and the excited state $|e\rangle$ as depicted in the figure. The system is driven by two optical fields $\mathcal{E}_1 \cos\omega_1 t$ and $\mathcal{E}_2 \cos\omega_2 t$. The atomic Hamiltonian is written as



$$\hat{H}_0 = -\hbar\omega_{01} |g_1\rangle \langle g_1| - \hbar\omega_{02} |g_2\rangle \langle g_2| , \tag{1}$$

where we have taken the excited state to have zero energy. The atom–field interaction Hamiltonian in the dipole approximation is

$$\hat{H}_{\text{int}} = d\mathcal{E}_1 \cos\omega_1 t \cdot (\hat{\sigma}_1 + \hat{\sigma}_1^\dagger) + d\mathcal{E}_2 \cos\omega_2 t \cdot (\hat{\sigma}_2 + \hat{\sigma}_2^\dagger), \tag{2}$$

with $\hat{\sigma}_{1,2} = |g_{1,2}\rangle \langle e|$. Here we assume that the detunings $\Delta_{1,2} = \omega_{1,2} - \omega_{01,02}$ are nearly equal such that the field $\omega_{1,2}$ couples only $|g_{1,2}\rangle$ to $|e\rangle$.

1. Show that in the rotating-wave approximation the atom–field interaction Hamiltonian can be written as

$$\hat{H}_{\text{int}} = \frac{\hbar\Omega_1}{2}\left(\hat{\sigma}_1 e^{i\omega_1 t} + \hat{\sigma}_1^\dagger e^{-i\omega_1 t}\right) + \frac{\hbar\Omega_2}{2}\left(\hat{\sigma}_2 e^{i\omega_2 t} + \hat{\sigma}_2^\dagger e^{-i\omega_2 t}\right), \tag{3}$$

with the Rabi frequencies $\Omega_{1,2}$.

2. Show that in the rotating frame the total Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}$ can be written as

$$\hat{H} = \hbar\Delta_1 |g_1\rangle \langle g_1| + \hbar\Delta_2 |g_2\rangle \langle g_2| + \frac{\hbar\Omega_1}{2}\left(\hat{\sigma}_1 + \hat{\sigma}_1^\dagger\right) + \frac{\hbar\Omega_2}{2}\left(\hat{\sigma}_2 + \hat{\sigma}_2^\dagger\right). \tag{4}$$

We want to find a state which does not couple to the other states even in the presence of the optical fields. To do so we first make a change of basis for the ground states, one of the new states decouples from the excited state. We define the new ground states $|g_\pm\rangle$ as

$$|g_+\rangle = \sin\theta\, |g_1\rangle + \cos\theta\, |g_2\rangle , \tag{5}$$
$$|g_-\rangle = \cos\theta\, |g_1\rangle - \sin\theta\, |g_2\rangle . \tag{6}$$

3. Obtain the Hamiltonian for the new basis.

4. What should $\tan\theta$ be for one of the ground states to decouple from the excited state?

Now let's take spontaneous emission into account. Supposing that the excited state $|e\rangle$ decays to $|g_{1,2}\rangle$ with the decay rate of $\Gamma_{1,2}$, so that the total decay rate of the excited state is $\Gamma = \Gamma_1 + \Gamma_2$, the Lindblad equation of the system reads

$$\frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar}[\hat{H}, \hat{\rho}] + \Gamma_1 \hat{\mathcal{D}}[\hat{\sigma}_1]\hat{\rho} + \Gamma_2 \hat{\mathcal{D}}[\hat{\sigma}_2]\hat{\rho}, \tag{7}$$

with $\hat{\rho}$ the density matrix. The superoperator $\hat{\mathcal{D}}[\hat{\sigma}]\hat{\rho}$ has the form

$$\hat{\mathcal{D}}[\hat{\sigma}]\hat{\rho} = \hat{\sigma}\hat{\rho}\hat{\sigma}^\dagger - \frac{1}{2}(\hat{\sigma}^\dagger\hat{\sigma}\hat{\rho} + \hat{\rho}\hat{\sigma}^\dagger\hat{\sigma}). \tag{8}$$

5. Obtain the Lindblad equation in the new basis. To simplify the result use $\Gamma_1 = \Gamma_2 = \Gamma/2$.

6. Which state becomes dark state in which condition?

II. STIRAP: STIMULATED RAMAN ADIABATIC PASSAGE

The possibility to transfer population from one quantum state to another coherently is of crucial importance for precision measurements and quantum information processing. For many applications it is important that the transfer efficiency does not depend sensitively on the experimental parameters such as the laser frequency, intensity and for pulses the precise timing.

This is possible with stimulated Raman adiabatic passage (STIRAP), which we will explore in this exercise. Consider an atom in state $|g_1\rangle$ which should be transferred coherently to state $|g_2\rangle$. To this aim, two laser pulses (of Gaussian envelope $\Omega_{1,2}(t) = \Omega_0 \exp\left[-\left(\frac{t-t_{1,2}}{T}\right)^2\right]$) with frequencies $\omega_1 = \omega_{01} - \Delta$ and $\omega_2 = \omega_{02} - \Delta$ are applied with a certain delay. The basic idea is that a dark state, which is initially identical with $|g_1\rangle$ is adiabatically transferred to $|g_2\rangle$, i.e., the angle $\theta(t)$ changes continuously from 0 to $\pi/2$.

1. Which laser, ω_1 or ω_2, has to be turned on first, and which next in order to achieve this transfer (without populating state $|e\rangle$ at any time)? Provide a schematic plot with intensity of the two Gaussian pulses $\Omega_1(t)$, $\Omega_2(t)$ as a function of time.

2. Derive the adiabaticity condition $\Omega_0 T \gg 1$ assuming $|t_{1,2}| = T$ in the case $\Delta = 0$. In an experiment in which the states should be transferred adiabatically in an extremely short time, what kind of laser do you need?

3. Using QuTiP plot the populations in the three states as a function of time (download the corresponding jupyter notebook from c4science). Tweak the pulse parameters and assess the adiabaticity condition. You should always find zero population in the state $|e\rangle$.

I. COHERENT DARK STATES

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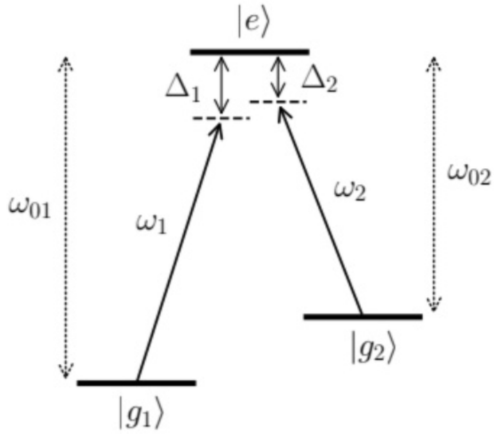

$$\hat{H}_0 = -\hbar\omega_{01} |g_1\rangle \langle g_1| - \hbar\omega_{02} |g_2\rangle \langle g_2| , \tag{1}$$

where we have taken the excited state to have zero energy. The atom–field interaction Hamiltonian in the dipole approximation is

$$\hat{H}_{\text{int}} = \overbrace{d\mathcal{E}_1}^{\hbar\Omega_1} \cos\omega_1 t \cdot (\hat{\sigma}_1 + \hat{\sigma}_1^\dagger) + d\mathcal{E}_2 \cos\omega_2 t \cdot (\hat{\sigma}_2 + \hat{\sigma}_2^\dagger), \tag{2}$$

with $\hat{\sigma}_{1,2} = |g_{1,2}\rangle \langle e|$. Here we assume that the detunings $\Delta_{1,2} = \omega_{1,2} - \omega_{01,02}$ are nearly equal such that the field $\omega_{1,2}$ couples only $|g_{1,2}\rangle$ to $|e\rangle$.

1. Show that in the rotating-wave approximation the atom–field interaction Hamiltonian can be written as

$$\hat{H}_{\text{int}} = \frac{\hbar\Omega_1}{2}(\hat{\sigma}_1 e^{i\omega_1 t} + \hat{\sigma}_1^\dagger e^{-i\omega_1 t}) + \frac{\hbar\Omega_2}{2}(\hat{\sigma}_2 e^{i\omega_2 t} + \hat{\sigma}_2^\dagger e^{-i\omega_2 t}), \tag{3}$$

with the Rabi frequencies $\Omega_{1,2}$.

$e^{i\omega_{\text{driving field}} H_0 t}$

Using $\hat{U} = \exp(-i\omega_1 t |g_1\rangle\langle g_1| - i\omega_2 t |g_2\rangle\langle g_2|) \to \hat{U}_{\text{rot}}$?

$$\boxed{\hat{U}^+ \underbrace{\hat{U} H_{int} \hat{U}^+}_{A} \hat{U} = H_{int}}$$

$e^{-i\omega_1 t |g\rangle\langle g|} = e^{-i\omega_1 t} |g\rangle\langle g|$

$\sigma = |g\rangle\langle e|$
$\sigma^+ = |e\rangle\langle g|$

$$A: \hat{U}\hat{H}_{int}\hat{U}^+ = \left(|e\rangle\langle e| + e^{-i\omega_1 t}|g_1\rangle\langle g_1| + e^{-i\omega_2 t}|g_2\rangle\langle g_2|\right)$$

$$\left(\frac{\hbar\Omega_1}{2}(e^{i\omega_1 t} + e^{-i\omega_1 t})(\hat{\sigma}_1 + \hat{\sigma}_1^+) + \frac{\hbar\Omega_2}{2}(e^{i\omega_2 t} + e^{-i\omega_2 t})(\sigma_2 + \sigma_2^+)\right)$$

$$\left(|e\rangle\langle e| + e^{i\omega_1 t}|g_1\rangle\langle g_1| + e^{i\omega_2 t}|g_2\rangle\langle g_2|\right)$$

RWA

$$= \frac{\hbar\Omega_1}{2}\left((1 + \cancel{e^{-2i\omega_1 t}})\hat{\sigma}_1 + (1 + \cancel{e^{2i\omega_1 t}})\sigma_1^+\right)$$

$$+ \frac{\hbar\Omega_2}{2}\left((1 + \cancel{e^{-2i\omega_2 t}})\hat{\sigma}_2 + (1 + \cancel{e^{2i\omega_2 t}})\sigma_2^+\right)$$

Transform back to the LAB FRAME yields Hamiltonian (3).

2. Show that in the rotating frame the total Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_{\text{int}}$ can be written as

$$\hat{H} = \hbar\Delta_1 |g_1\rangle\langle g_1| + \hbar\Delta_2 |g_2\rangle\langle g_2| + \frac{\hbar\Omega_1}{2}(\hat{\sigma}_1 + \hat{\sigma}_1^\dagger) + \frac{\hbar\Omega_2}{2}(\hat{\sigma}_2 + \hat{\sigma}_2^\dagger). \tag{4}$$

$$H_{rot} = i\hbar \dot{\hat{U}} \hat{U}^\dagger + \hat{U} \hat{H} \hat{U}^\dagger$$

$$i\hbar\left(-i\omega_1 |g_1\rangle\langle g_1| - i\omega_2 |g_2\rangle\langle g_2|\right)\left(|e\rangle\langle e| + e^{-i\omega_1 t}|g_1\rangle\langle g_1| + e^{-i\omega_2 t}|g_2\rangle\langle g_2|\right)$$

$$\times\left(|e\rangle\langle e| + e^{i\omega_1 t}|g_1\rangle\langle g_1| + e^{i\omega_2 t}|g_2\rangle\langle g_2|\right)$$

$$= \hbar\omega_1 |g_1\rangle\langle g_1| + \hbar\omega_2 |g_2\rangle\langle g_2|$$

$$\hat{U} H_0 \hat{U}^\dagger = \left(e^{0}|e\rangle\langle e| + e^{-i\omega_1 t}|g_1\rangle\langle g_1| + e^{-i\omega_2 t}|g_2\rangle\langle g_2|\right) \cdot$$

$$\left(-\hbar\omega_{01} |g_1\rangle\langle g_1| - \hbar\omega_{02} |g_2\rangle\langle g_2|\right) \cdot$$

$$\left(|e\rangle\langle e| + e^{+i\omega_1 t}|g_1\rangle\langle g_1| + e^{i\omega_2 t}|g_2\rangle\langle g_2|\right)$$

$$= -\hbar\omega_{01} |g_1\rangle\langle g_1| - \hbar\omega_{02} |g_2\rangle\langle g_2|$$

$$\hat{U} H_{int} \hat{U}^\dagger = \frac{\hbar\Omega_1}{2}(\hat{\sigma}_1 + \hat{\sigma}_1^\dagger) + \frac{\hbar\Omega_2}{2}(\hat{\sigma}_2 + \hat{\sigma}_2^\dagger)$$

from previous part

Thus, $$\tilde{H} = \hbar\Delta_1 |g_1\rangle\langle g_1| + \hbar\Delta_2 |g_2\rangle\langle g_2| + \frac{\hbar\Omega_1}{2}(\hat{\sigma}_1 + \hat{\sigma}_1^\dagger) + \frac{\hbar\Omega_2}{2}(\hat{\sigma}_2 + \hat{\sigma}_2^\dagger)$$

$(\omega_1 - \omega_{01})$

We want to find a state which does not couple to the other states even in the presence of the optical fields. To do so we first make a change of basis for the ground states, one of the new states decouples from the excited state. We define the new ground states $|g_{\pm}\rangle$ as

$$|g_+\rangle = \sin\theta\,|g_1\rangle + \cos\theta\,|g_2\rangle\,, \tag{5}$$

$$|g_-\rangle = \cos\theta\,|g_1\rangle - \sin\theta\,|g_2\rangle\,. \tag{6}$$

3. Obtain the Hamiltonian for the new basis.

$$|g_1\rangle = \sin\theta\,|g_+\rangle + \cos\theta\,|g_-\rangle$$
$$|g_2\rangle = \cos\theta\,|g_+\rangle - \sin\theta\,|g_-\rangle$$

write down this into the H_{rot}

$$|\sigma_+^+\rangle = |e\rangle\langle g_+| = \sin\theta\,\overbrace{|e\rangle\langle g_1|}^{\sigma_1^+} + \cos\theta\,\overbrace{|e\rangle\langle g_2|}^{\sigma_2^+}$$
$$|\sigma_+\rangle = |g_+\rangle\langle e| = \sin\theta\,\underbrace{|g_1\rangle\langle e|}_{\sigma_1} + \cos\theta\,\underbrace{|g_2\rangle\langle e|}_{\sigma_2}$$

?

$$H_{int} = \frac{\hbar\Omega_1}{2}\Big(\overbrace{\sin\theta(\hat\sigma_+ + \hat\sigma_+^+) + \cos\theta(\hat\sigma_- + \hat\sigma_-^+)}^{\hat\sigma_1}\Big)$$
$$+ \frac{\hbar\Omega_2}{2}\Big(\underbrace{\cos\theta(\hat\sigma_+ + \hat\sigma_+^+)}_{\hat\sigma_2} - \underbrace{\sin\theta(\hat\sigma_- + \hat\sigma_-^+)}_{\hat\sigma_2^+}\Big)$$

$$H_0 = \hbar(\sin^2\theta\Delta_1 + \cos^2\theta\Delta_2)|g_+\rangle\langle g_+| + \hbar(\cos^2\theta\Delta_1 + \sin^2\theta\Delta_2)|g_-\rangle\langle g_-|$$
$$+\hbar\sin\theta\cos\theta(\Delta_1 - \Delta_2)\big(|g_+\rangle\langle g_-| + |g_-\rangle\langle g_+|\big)$$

$$\boxed{H = H_0 + H_{int} = \hbar\Delta_+|g_+\rangle\langle g_+| + \hbar\Delta_-|g_-\rangle\langle g_-| + \hbar\Omega_g\big(|g_+\rangle\langle g_-| + |g_-\rangle\langle g_+|\big) + \frac{\hbar\Omega_+}{2}(\hat\sigma_+ + \hat\sigma_+^+) + \frac{\hbar\Omega_-}{2}(\hat\sigma_- + \hat\sigma_-^+)}$$

where

$$\Delta_+ = \sin^2\theta\Delta_1 + \cos^2\theta\Delta_2$$
$$\Delta_- = \cos^2\theta\Delta_1 + \sin^2\theta\Delta_2$$
$$\Omega_g = \sin\theta\cos\theta(\Delta_1 - \Delta_2)$$
$$\Omega_+ = \sin\theta\,\Omega_1 + \cos\theta\,\Omega_2 \qquad \Omega_- = \cos\theta\,\Omega_1 - \sin\theta\,\Omega_2$$
$$\sigma_{\pm} = |g_{\pm}\rangle\langle e|$$

4. What should $\tan\theta$ be for one of the ground states to decouple from the excited state?

$|g_-\rangle$ decouples from $|e\rangle$ when $\Omega_- = 0$

$$\Omega_- = \cos\theta\,\Omega_1 - \sin\theta\,\Omega_2 = 0$$

$$\cos\theta\,\Omega_1 = \sin\theta\,\Omega_1$$

$$\tan\theta = \frac{\Omega_1}{\Omega_2}$$

Now let's take spontaneous emission into account. Supposing that the excited state $|e\rangle$ decays to $|g_{1,2}\rangle$ with the decay rate of $\Gamma_{1,2}$, so that the total decay rate of the excited state is $\Gamma = \Gamma_1 + \Gamma_2$, the Lindblad equation of the system reads

$$\frac{d\hat{\rho}}{dt} = -\frac{i}{\hbar}[\hat{H}, \hat{\rho}] + \Gamma_1 \hat{\mathcal{D}}[\hat{\sigma}_1]\hat{\rho} + \Gamma_2 \hat{\mathcal{D}}[\hat{\sigma}_2]\hat{\rho}, \tag{7}$$

with $\hat{\rho}$ the density matrix. The superoperator $\hat{\mathcal{D}}[\hat{\sigma}]\hat{\rho}$ has the form

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5. Obtain the Lindblad equation in the new basis. To simplify the result use $\Gamma_1 = \Gamma_2 = \Gamma/2$.

6. Which state becomes dark state in which condition?

Plug $|g_{1,2}\rangle$ into Eq (8)

$$\Gamma_1\left(D[\sigma_1]\rho\right) = \Gamma_1\left(\sin^2\theta\, D[\sigma_+]\rho + \cos^2\theta\, D[\sigma_-]\rho + \sin\theta\cos\theta\,(\sigma_+\rho\sigma_-^+ + \sigma_-\rho\sigma_+^+)\right)$$

$$\Gamma_2\left(D[\sigma_2]\rho = \Gamma_2\left(\cos^2\theta\, D[\sigma_+]\rho + \sin^2\theta\, D[\sigma_-]\rho - \sin\theta\cos\theta\,(\sigma_+\rho\sigma_-^+ + \sigma_-\rho\sigma_+^+)\right)\right.$$

$$\frac{d\rho}{dt} = -\frac{i}{\hbar}[H,\rho] + \Gamma_+ D[\sigma_+]\rho + \Gamma_- D[\sigma_-]\rho + \boxed{\sin\theta\cos\theta\,(\Gamma_1 - \Gamma_2)}\,(\sigma_+\rho\sigma_-^+ + \sigma_-\rho\sigma_+^+)$$

with $\Gamma_+ = \sin^2\theta\,\Gamma_1 + \cos^2\theta\,\Gamma_2$

$\Gamma_- = \cos^2\theta\,\Gamma_1 + \sin^2\theta\,\Gamma_2$

correction term to handle asymmetric decay to $|g_1\rangle$ and $|g_2\rangle$?

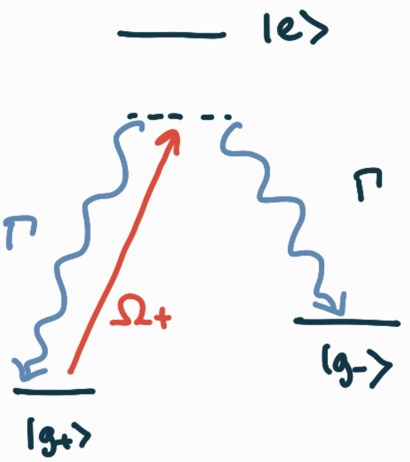
Let $\Gamma_1 = \Gamma_2 = \Gamma = \Gamma_+ = \Gamma_-$

Thus the result became $\frac{d\rho}{dt} = \frac{-i}{\hbar}[H,\rho] + \Gamma D[\sigma^+]\rho + \Gamma D[\sigma_-]\rho$

6) $|g_-\rangle$ decoupled from $|e\rangle$ for $\tan\theta = \frac{\Omega_1}{\Omega_2}$

It also decouples from $|g_+\rangle$ when $\Omega_g = 0$. Therefore $|g_-\rangle$ becomes the DARK STATE when $\Delta_1 = \Delta_2$ (RAMAN condition)

$\Omega_g = \sin\theta\cos\theta\,(\Delta_1 - \Delta_2)$



The possibility to transfer population from one quantum state to another coherently is of crucial importance for precision measurements and quantum information processing. For many applications it is important that the transfer efficiency does not depend sensitively on the experimental parameters such as the laser frequency, intensity and for pulses the precise timing.

This is possible with stimulated Raman adiabatic passage (STIRAP), which we will explore in this exercise. Consider an atom in state $|g_1\rangle$ which should be transferred coherently to state $|g_2\rangle$. To this aim, two laser pulses (of Gaussian envelope $\Omega_{1,2}(t) = \Omega_0 \exp[-(\frac{t-t_{1,2}}{T})^2])$ with frequencies $\omega_1 = \omega_{01} - \Delta$ and $\omega_2 = \omega_{02} - \Delta$ are applied with a certain delay. The basic idea is that a dark state, which is initially identical with $|g_1\rangle$ is adiabatically transferred to $|g_2\rangle$, i.e., the angle $\theta(t)$ changes continuously from 0 to $\pi/2$.

1. Which laser, ω_1 or ω_2, has to be turned on first, and which next in order to achieve this transfer (without populating state $|e\rangle$ at any time)? Provide a schematic plot with intensity of the two Gaussian pulses $\Omega_1(t)$, $\Omega_2(t)$ as a function of time.

$$\left.\begin{array}{l} |g_-\rangle = |g_1\rangle \text{ when } \theta = 0 \\ |g_-\rangle = |g_2\rangle \text{ when } \theta = \frac{\pi}{2} \end{array}\right\}$$

we want $|g_1\rangle \longrightarrow |g_2\rangle$

θ can be controlled via $\tan\theta = \frac{\Omega_1}{\Omega_2}$

* First, ω_2 has to be turned on first and then ω_1.

$|g_2\rangle$ and $|e\rangle$ are first coupled by laser ω_2 making atoms populate in $|g_1\rangle = |g_-\rangle$

Then slowly turn on laser ω_1 so that atoms stay in $|g_-\rangle \neq |g_1\rangle$. Finally slowly turn off laser ω_2, which makes $|g_-\rangle = |g_2\rangle$.

2. Derive the adiabaticity condition $\Omega_0 T \gg 1$ assuming $|t_{1,2}| = T$ in the case $\Delta = 0$. In an experiment in which the states should be transferred adiabatically in an extremely short time, what kind of laser do you need?

Adiabaticity condition is $\dot{\theta}(t) \ll \Omega_+$

$$\Omega_+ = \sin\theta\,\Omega_1 + \cos\theta\,\Omega_2$$

$$\Omega_+ = \sqrt{\Omega_1^2 + \Omega_2^2} \leq \sqrt{2}\,\Omega_0$$

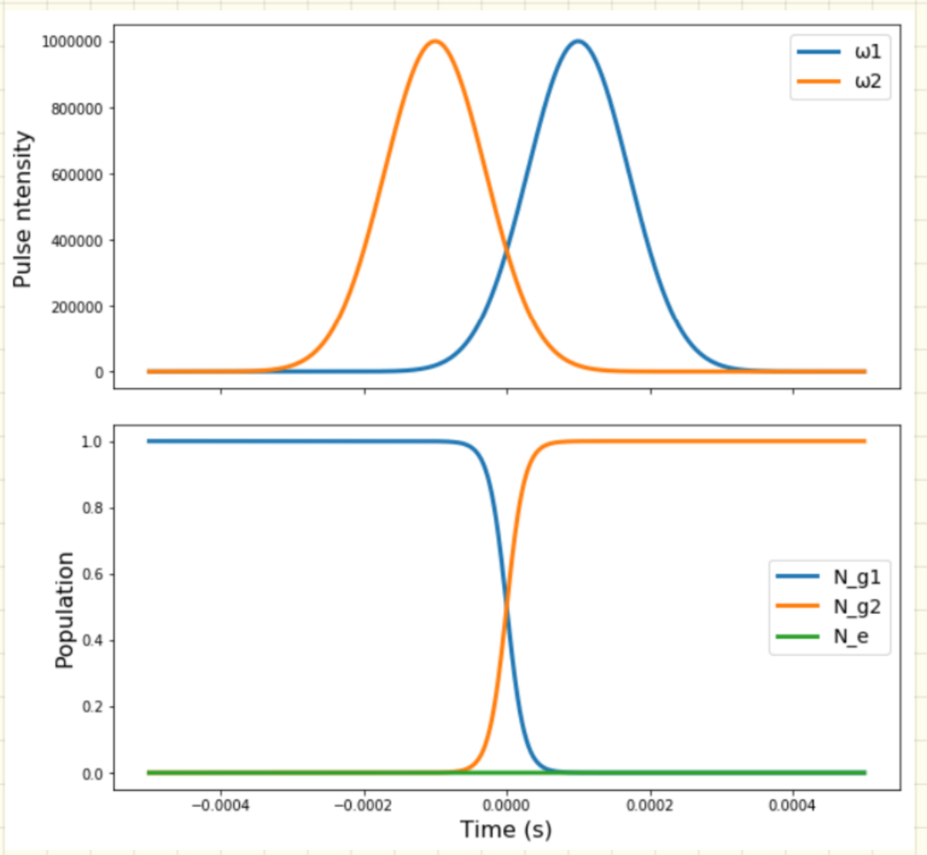
$$\dot{\theta}(t) = \frac{d}{dt}\,\mathrm{Arctan}\left(\frac{\Omega_2}{\Omega_1}\right) = \frac{\dot{\Omega}_2\Omega_1 - \dot{\Omega}_1\Omega_2}{\Omega_1^2 + \Omega_2^2} \ll \sqrt{2}\,\Omega_0$$

$$\frac{1}{\Omega_0 T}\,\frac{4}{\sqrt{2}}\,\frac{\frac{4}{T}\exp\left(-2\,\frac{t^2+T^2}{T^2}\right)}{\exp\left(-2\,\frac{t^2+T^2}{T^2}\right)\left(\exp\left(\frac{4t}{T^2}\right) + \exp\left(-\frac{4t}{T^2}\right)\right)} \ll 1$$

$$\frac{1}{\Omega_0 T} \ll 1$$

For extremely short times (T) provided that the Raman couplings (Ω_0) are large enough. It can be achieved with highly intense pulsed lasers, femtosecond lasers.

3. Using QuTiP plot the populations in the three states as a function of time (download the corresponding jupyter notebook from c4science). Tweak the pulse parameters and assess the adiabaticity condition. You should always find zero population in the state $|e\rangle$.



For the given Raman coupling $\Omega_0 = 1\,\text{MHz}$,

The pulse width has to be sufficiently larger than $1\,\mu s$