9 Atomic Coherence in Three-Level Atoms

9.1 Coherent trapping - dark states

In multi-level systems coherent superpositions between different states (*atomic coherence*) may lead to dramatic changes of light absorption and propagation.

The simplest example is a three level system. In the following a Λ-system with two ground states $|g_1\rangle$, $|g_2\rangle$ and one excited state $|e\rangle$ is depicted:

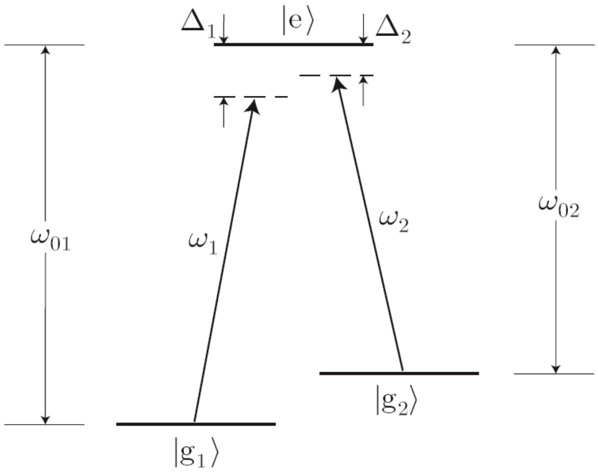

Figure 59: Schematic of a three-level (Λ system.

We assume that two light fields in exact resonance ($\Delta_1 = \Delta_2 = 0$) shine on the system.

We start with the Hamiltonian

$$H_{three} = H_0 + H_1 \tag{422}$$

with

$$H_0 = \hbar\omega_e|e\rangle\langle e| + \hbar\omega_{g1}|g_1\rangle\langle g_1| + \hbar\omega_{g2}|g_2\rangle\langle g_2| \tag{423}$$

$$H_1 = -\frac{\hbar}{2}\left(\Omega_{R1}e^{-i\phi_1}e^{-i\omega_1 t}|e\rangle\langle g_1| + \Omega_{R2}e^{-i\phi_2}e^{-i\omega_2 t}|e\rangle\langle g_2|\right) + H.c. \tag{424}$$

Here, $\Omega_{R1}e^{-i\phi_1}$ and $\Omega_{R2}e^{-i\phi_2}$ are the complex Rabi frequencies associated with the coupling of the field modes of frequency ω_1 and ω_2 to the atomic transition $|e\rangle \rightarrow |g_1\rangle$ and $|e\rangle \rightarrow |g_2\rangle$, repectively.

The most general form of the atomic wave function can then be written as:

$$|\psi\rangle = c_e e^{-i\omega_e}|e\rangle + c_{g1}e^{-i\omega_{g1}}|g_1\rangle + c_{g2}e^{-i\omega_{g2}}|g_2\rangle \tag{425}$$

The equations of motion of the coefficients can be derived from the Schroedinger equation:

$$\dot{c_e} = \frac{i}{2}\left(\Omega_{R1}e^{-i\phi_1}c_{g1} + \Omega_{R2}e^{-i\phi_2}c_{g2}\right) \tag{426}$$

$$\dot{c_{g1}} = \frac{i}{2}\Omega_{R1}e^{i\phi_1}c_e \tag{427}$$

$$\dot{c_{g2}} = \frac{i}{2}\Omega_{R2}e^{i\phi_2}c_e \tag{428}$$

As pointed out above exact resonance ($\omega_e - \omega_{g1} = \omega_1$ and $\omega_e - \omega_{g2} = \omega_2$) is assumed.

We now assume the initial state to be in a superposition of the two ground states:

$$|\psi\rangle = \cos(\theta/2)|g_1\rangle + \sin(\theta_2)e^{-i\psi}|g_2\rangle \tag{429}$$

Then, the following solution for the time evolution of the coefficients can be found:

$$c_e = \frac{i\sin(\Omega t/2)}{\Omega}\left[\Omega_{R1}e^{-i\phi_1}\cos(\theta/2) + \Omega_{R2}e^{-i(\phi_2+\psi)}\sin(\theta/2)\right] \tag{430}$$

$$c_{g1} = \frac{1}{\Omega^2}(\left[\Omega_{R2}^2 + \Omega_{R1}^2\cos(\Omega t/2)\right]\cos(\theta/2) \tag{431}$$

$$- 2\Omega_{R_1}\Omega_{R_2}e^{i(\phi_1-\phi_2-\psi)}\sin^2(\Omega t/4)\sin(\theta/2)) \tag{432}$$

$$c_{g2} = \frac{1}{\Omega^2}(\left[\Omega_{R1}^2 + \Omega_{R2}^2\cos(\Omega t/2)\right]e^{-i\psi}\sin(\theta/2) \tag{433}$$

$$- 2\Omega_{R_1}\Omega_{R_2}e^{-i(\phi_1-\phi_2)}\sin^2(\Omega t/4)\cos(\theta/2)) \tag{434}$$

with $\Omega = (\Omega_{R1}^2 + \Omega_{R1}^2)^{1/2}$

For the special case

$$\Omega_{R1} = \Omega_{R1} \qquad \theta = \pi/2 \qquad \phi_1 - \phi_2 - \psi = \pm\pi \tag{435}$$

the population is *trapped* in the ground states, i.e.:

$$c_e = 0 \tag{436}$$
$$c_{g1} = \frac{1}{\sqrt{2}} \tag{437}$$
$$c_{g2} = \frac{1}{\sqrt{2}} e^{-i\psi} \tag{438}$$

Obviously, there is no absorption even in the presence of resonant fields! This phenomenon is called *coherent population trapping*; the system is said to be in a *dark state*. Quantum mechanically in this situation there is a destructive interference between the two paths that lead to a transition from one of ground states to the excited state.

Even in the more general case:

$$\frac{\sin(\theta/2)}{\cos(\theta/2)} = \tan(\theta/2) = -\frac{\Omega_{R1}}{\Omega_{R2}} e^{-i(\phi_1 - \phi_2 - \psi)} \tag{439}$$

there is no population in the excited state $|e\rangle$.

The generalized dark state

$$|\psi(t)\rangle = \frac{\Omega_{R2}(t) e^{-i\phi_2} |g1\rangle - \Omega_{R1}(t) e^{-i\phi_1} |g2\rangle}{\sqrt{\Omega_{R1}^2 + \Omega_{R2}^2}} \tag{440}$$

can be created, e.g., by starting with the atom in the ground state $|g1\rangle$ with $\Omega_{R1} = 0$ and adiabatically switching on Ω_{R1}.

9.2 Electromagnetically Induced Transparency (EIT)

9.2.1 Basic principles

We now want to discuss how atomic coherence can change the absorption and dispersion of a probe beam of classical light. In order to do that we start with the same three-level scheme as shown in Fig. 59. We now consider a weak *probe beam* $E = E_0 \exp(-i\omega_1 t) + c.c.$ and a strong *pump beam* with frequency ω_p having a complex Rabi frequency $\Omega_p \exp(-i\phi)$. Additionally, we introduce incoherent decay rates γ_e, γ_{g1}, and γ_{g2} from the levels $|e\rangle$, $|g_1\rangle$, and $|g_2\rangle$, respectively.

We are interested in the absorption and dispersion of the weak pump beam. It is related to the atomic polarization P as introduced in the previous chapter:

$$P = \langle e|H_I|g1\rangle\rho_{e,g1} = \langle e|\vec{\mu}\vec{E}|g1\rangle\rho_{e,g1} = \mu_{e,g1}E\rho_{e,g1} \tag{441}$$

In the linear regime P is related to the field amplitude E_0 via the complex susceptibility χ:

$$P = \epsilon_0\chi E_0 \tag{442}$$

Its real and imaginary part determine the probe's dispersion and absorption.

We now have to find $\rho_{e,g1}$. This is done as in subsection 9.1 by solving the Schroedinger equation. Compared (eqn. 424) to the previous notation (and setting $\phi_2 = 0$ without loss of generality) we use:

$$\Omega_{R1}e^{-i\omega_1 t} = \frac{2\mu_{e,g1}E_0}{\hbar}e^{-i\omega_1 t}; \qquad \Omega_{R2}e^{-i\phi_2}e^{-i\omega_2 t} = \Omega_p e^{-i\omega_p t}e^{-i\phi_p} \tag{443}$$

After some calculation we find:

$$\dot{\rho}_{e,g1} = -(i(\omega_e - \omega_{g1}) + \gamma_e)\rho_{e,g1} - \frac{i}{2}\Omega_{R1}e^{-i\omega_1}(\rho_{e,e} - \rho_{g1,g1}) \tag{444}$$

$$+ \frac{i}{2}\Omega_p e^{-i\omega_p t}e^{-i\phi_p}\rho_{g2,g1} \tag{445}$$

$$\dot{\rho}_{g2,g1} = -(i(\omega_{g2} - \omega_{g1}) + \gamma_{g2})\rho_{g2,g1} - \frac{i}{2}\Omega_{R1}e^{-i\omega_1}\rho_{g2,e} + \frac{i}{2}\Omega_p e^{i\omega_p t}e^{i\phi_p}\rho_{e,g1} \tag{446}$$

$$\dot{\rho}_{e,g2} = -(i(\omega_e - \omega_{g2}) + \gamma_{g1})\rho_{e,g2} - \frac{i}{2}\Omega_p e^{-i\omega_p t}e^{-i\phi_p}(\rho_{e,e} - \rho_{g2,g2}) \tag{447}$$

$$+ \frac{i}{2}\Omega_{R1}e^{-i\omega_1}\rho_{g1,g2} \tag{448}$$

As we assumed a weak probe field, a solution for $\rho_{e,g1}$ has to be found in first order only, while the rest has to be treated exactly (due to the strong pump).

With the atom initially in the lowest state (all matrix elements are zero except $\rho_{g1,g1} = 1$) we find with the additional substitution

$$\rho_{e,g1} = \tilde{\rho}_{e,g1}e^{-i\omega_1 t} \tag{449}$$

$$\rho_{g2,g1} = \tilde{\rho}_{g2,g1}e^{-i(\omega_1 + \omega_{g2} - \omega_e)t} \tag{450}$$

the following set of equations:

$$\dot{\tilde{\rho}}_{e,g1} = -(\gamma_e + i\Delta)\tilde{\rho}_{e,g1} + \frac{i}{2}\Omega_{R1} + \frac{i}{2}\Omega_p e^{-i\phi_p}\tilde{\rho}_{g2,g1} \tag{451}$$

$$\dot{\tilde{\rho}}_{g2,g1} = -(\gamma_{g2} + i\Delta)\tilde{\rho}_{g2,g1} + \frac{i}{2}\Omega_p e^{i\phi_p}\tilde{\rho}_{e,g1} \tag{452}$$

where $\Delta = (\omega_e - \omega_{g1}) - \omega_1$ is the detuning of the probe field, and where $\omega_p = \omega_e - \omega_{g2}$ is assumed.
this set of equations can be solved, by first writing it in matrix form,

$$\dot{R} = -MR + A \tag{453}$$

with

$$R = \begin{pmatrix} \tilde{\rho}_{e,g1} \\ \tilde{\rho}_{g2,g1} \end{pmatrix}, \quad M = \begin{pmatrix} \gamma_e + i\Delta & -\frac{i}{2}\Omega_p e^{-i\phi_p} \\ -\frac{i}{2}\Omega_p e^{i\phi_p} & \gamma_{g2} + i\Delta \end{pmatrix}, \quad A = \begin{pmatrix} \frac{i}{2}\Omega_{R1} \\ 0 \end{pmatrix} \tag{454}$$

and then integrating

$$R(t) = \int_{-\infty}^{t} e^{-M(t-t')} A \, dt' = M^{-1} A \tag{455}$$

This yields:

$$\rho_{e,g1} = \frac{i\Omega_{R1} e^{-i\omega_1 t}(\gamma_{g2} + i\Delta)}{2[(\gamma_e + i\Delta)(\gamma_{g2} + i\Delta) + \Omega_p^2/4]} \tag{456}$$

By plugging in the expression for Ω_{R1} and using the relation between the atomic polarization P and $\rho_{e,g1}$ one can identify the real and imaginary part of the susceptibility $\chi = \chi' + i\chi''$:

$$\chi' = \frac{N_a |\mu_{e,g1}|^2 \Delta}{\epsilon_0 \hbar Z} \left[\gamma_{g2}(\gamma_e + \gamma_{g2}) + (\Delta^2 - \gamma_e \gamma_{g2} - \Omega_p^2/4)\right] \tag{457}$$

$$\chi'' = \frac{N_a |\mu_{e,g1}|^2}{\epsilon_0 \hbar Z} \left[\Delta^2(\gamma_e + \gamma_{g2}) - \gamma_{g2}(\Delta^2 - \gamma_e \gamma_{g2} - \Omega_p^2/4)\right] \tag{458}$$

where N_a is the atom number density (if a gas of identical atoms is considered) and

$$Z = (\Delta^2 - \gamma_e \gamma_{g2} - \Omega_p^2/4)^2 + \Delta^2(\gamma_e + \gamma_{g2})^2. \tag{459}$$

9.2.2 Change of index of refraction

The complex susceptibility is directly related to the complex index of refraction $n = n' + in''$:

$$n = \sqrt{1 + \chi} \approx 1 + \frac{\chi}{2} \tag{460}$$

n' determines the dispersion and n'' the absorption of the probe beam.

Figure 60 plots the real and imaginary part of the index of refraction for a weak pump beam. When the pump is present, there is distinct dip (*EIT window*) in the absorption. This allows using an atomic resonance, e.g. to modulate the index of refraction, without (!) significant absorption. This is not possible for a two-level atom.

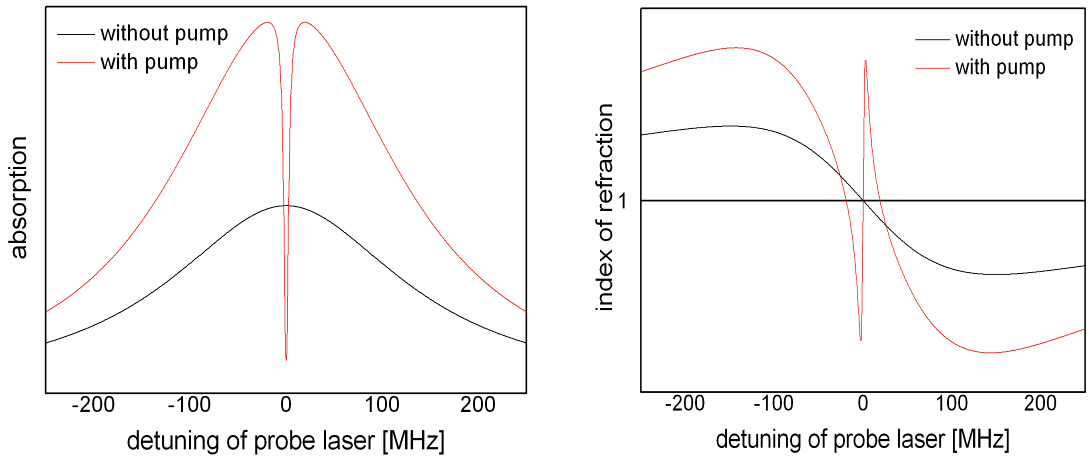

Figure 60: Imaginary (left) and real (right) part of the index of refraction for a probe beam with and without a strong pump beam.

Compare to the dark state describe in the previous subsection the spontaneous emission pumps the system automatically in a state, where there is no absorption due to interference of different excitation paths.

Another interpretation of the EIT effect is provided by the dressed states picture. It was shown in the previous chapter that a coupled system consisting of a two-level atom and a resonant light field has novel eigenstates (dressed states). The dressed states are split by the Rabi frequency Ω which is related to the field strength of the coupling laser. A weak beam probes this new energy level structure and will see a doublet when tuned across the resonance. This is depicted in Fig. 61.

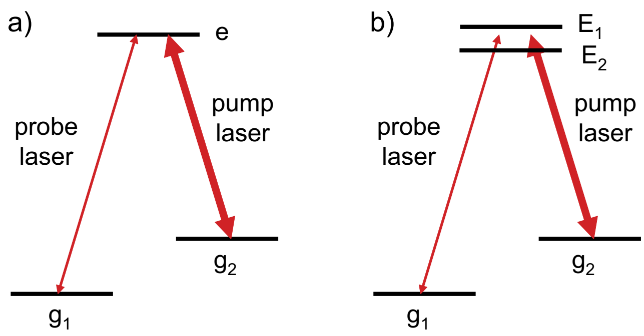

Figure 61: Interpretation of the EIT effect in terms of the dressed states picture: a) Energy levels in the bare state picture, b) Energy levels in the dressed state picture.

As can be seen a strong pump seems to split the excited state, so that a probe is not resonant to any transition anymore. In Fig. 62 the energy level diagram of ^{133}Cs is shown together with a schematic of an experimental setup to study the EIT effect. Figure 63 shows the result of the experiment.

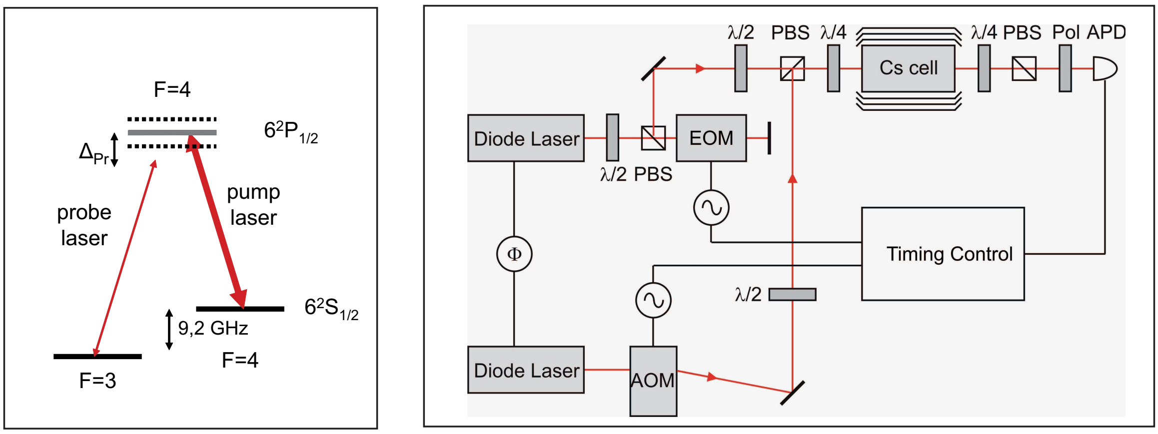

Figure 62: Left: Energy level diagram of ^{133}Cs; Right: schematic of an experimental setup.

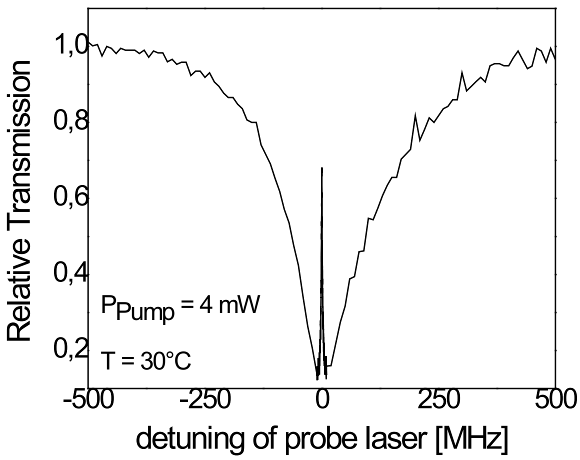

Figure 63: Experimental result of an EIT experiment: The transmission of a weak probe beam is increased when a pump beam is present.

9.3 Slow Light

EIT allows to modify the propagation of light dramatically. The narrow EIT resonance leads to a large gradient of the index of refraction close the atomic resonance. Compared to a two-level atom this is accompanied by negligible absorption. For pulse propagation this has dramatic consequences: The speed of propagation of a pulse with frequency ω is determined by the group velocity v_g. It is:

$$n_g = n' + \omega \frac{dn'}{d\omega} \tag{461}$$

Obviously, n_g can be very large at the EIT resonance!

A prominent experiment has been performed by Lene Hau and co-workers [Nature 397, 594 (1999)] where light could be decelerated to the speed of a bicyclist (see Fig. 64). A requirement is that the light pulse spectrally fits in the EIT window.

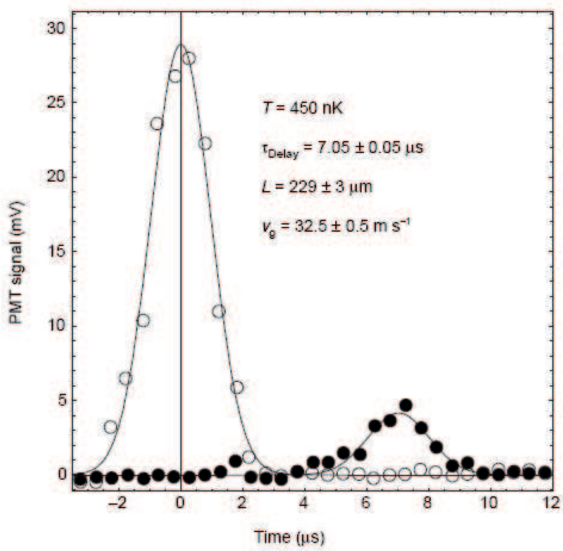

Figure 64: Measured time delay of a light pulse transmitted through an EIT medium. The calculated velocity in the medium was only $32,5\,m/s$, from [Hau et al., Nature 397, 594 (1999)].

More resent experiments could even bring a light pulse to a complete stop in an EIT medium. In this case a light pulse was sent in an EIT cell. Due to the very slow propagation it is compressed and at a certain time completely fits in the cell. Then, the pump beam is switched off. During switch-off the atomic state remains in a dark state. The light pulse is in fact transfered into a coherent superposition of the ground states of many atoms. As these states are typically hyperfine states, it is then referred to as *spin wave*. There it can be stored for some time. In order to release the light the pump beam has to be switched on again, and the pulse continues to travel.

Figure 65 shows a numerical simulation of such a light storage process.

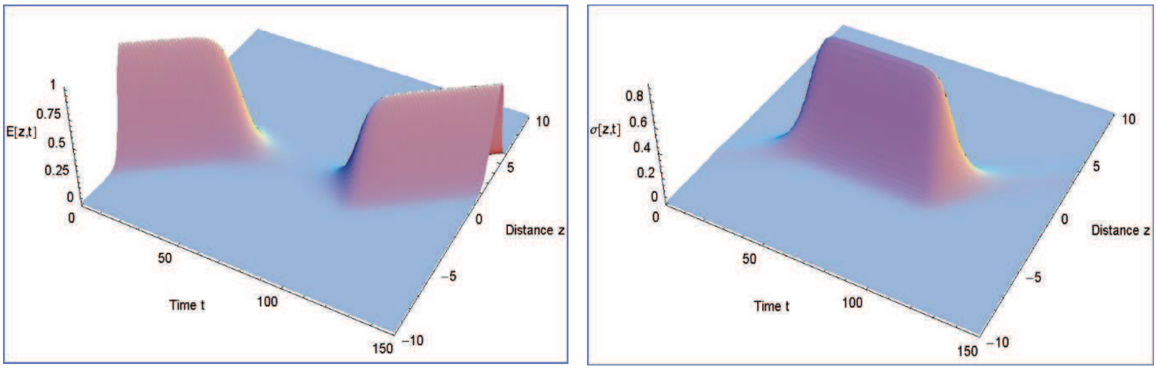

Figure 65: Simulation of light storage: The left plot shows the expectation value of the electric field of a light pulse entering the EIT medium. On the right there is a plot of the population of the atomic ground states. At a time between 50 and 100 (arbitrary units) the pump pulse is switched off.

Experimentally storage of light pulses has been demonstrated with storage times up to $> 200\,\mu s$ in atomic gas cells and up to a second in the solid state at ultra-low temperatures. Figure 66 shows the experimental results from the Walsworth/Lukin groups in Harvard [Phillips et al., Phys. Rev. Lett. 86, 783 (2001)].

Important applications of EIT are possible coherent interfaces between light and matter, e.g. for quantum information processing. Another interesting aspect is that rather small optical delay lines can be fabricated. Also due to the very slow propagation of the light non-linear effects are dramatically enhanced.

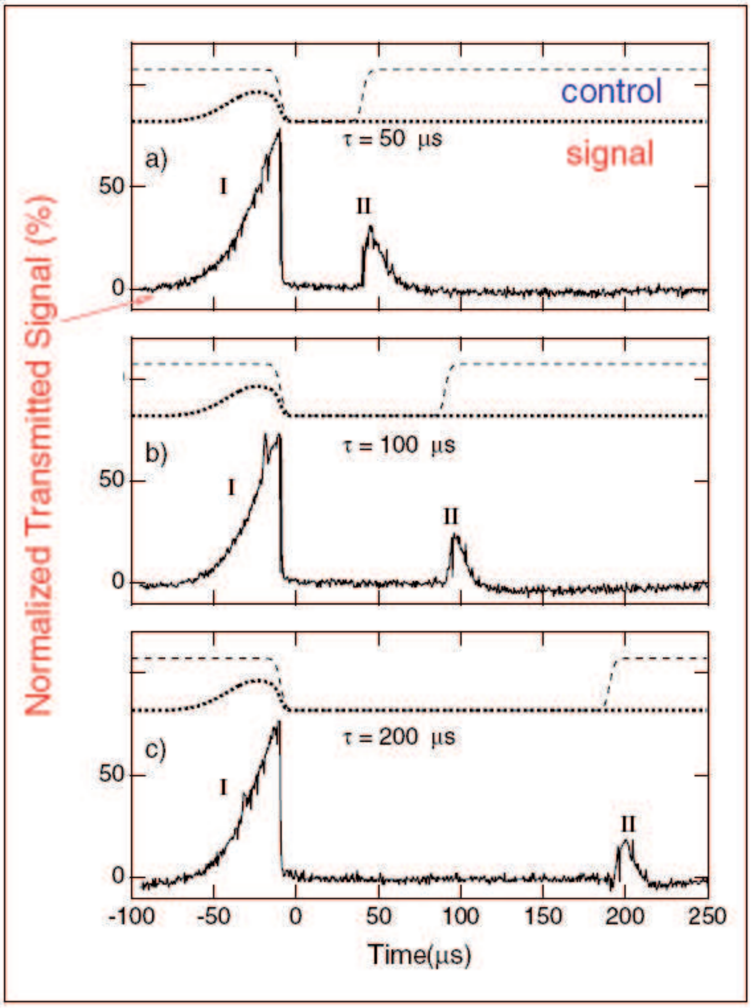

Figure 66: Storage of a light pulse for different storage times, from [Phillips et al., Phys. Rev. Lett. 86, 783 (2001)].

9.4 Stimulated Raman Adaiabtic passage (STIRAP)

A similar process as the one utilized in light storage is *stimulated Raman adabatic passage (STIRAP)*. It is a very efficient way to move all the population from one ground state to another in a three-level (Λ) system. Consider the following system:

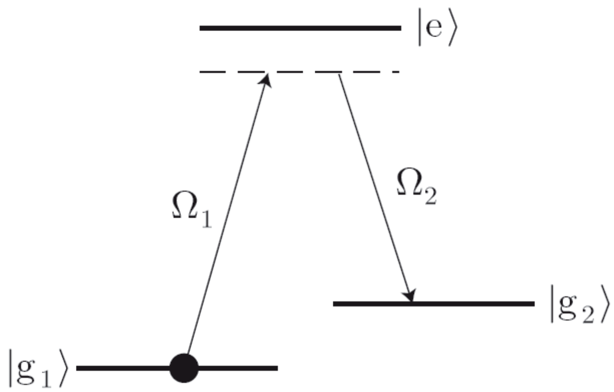

Figure 67: Schematic of a STIRAP in a Λ system.

Instead of using an exact π-pulse driving the transition between the two ground states one can make use again of the dark state. As show above the dark state is:

$$|\psi(t)\rangle = \frac{\Omega_{R2}(t)e^{-i\phi_2}|g1\rangle - \Omega_{R1}(t)e^{-i\phi_1}|g2\rangle}{\sqrt{\Omega_{R1}^2 + \Omega_{R2}^2}} \tag{462}$$

That means an adiabatic switching between the two extremal cases of the dark state, i.e. from atom in $|g_1\rangle$ and $\Omega_{R1} = 0$ to atom in state $|g_2\rangle$ and $\Omega_{R2} = 0$ is possible.

The somewhat counter-intuitive sequence of pulses shown in Fig. 68 has to be applied.

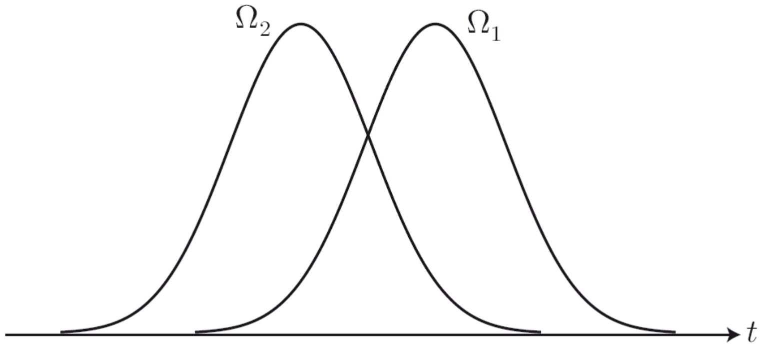

Figure 68: "Counter-intuitive" pulse sequence for a STIRAP process

Then, the atom will always follow the dark state until it reaches the final state $|g_2\rangle$. Compared to applying a π-pulse directly, the method is very forgiving concerning the exact shape and intensity of the pulses.