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Phase Control of Medium Gain in a Double Two-level System: From Ultrashort to Long Pulse Regime

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We consider a double two-level system in which each single two-level system is driven by a strong pulse. The light shifts induced by the strong field are probed by a weak pulse that connects the two sub-system in a crossed manner– connecting the ground state of one system with the excited state of the other and vice versa. We are interested in coherent control of medium gain for the weak pulse and we demonstrate the conditions on laser and atomic parameters that render this control possible. Both ultra-short and long pulse regimes are discussed.

Keywords: Coherent control, propagation effects.

1 Introduction

The idea of coherent control emerged a few years ago when it was demonstrated that accurate control of the dynamics of a physical system, interacting with light, was possible [1]. Laser field interacting with quantum mechanical systems gives rise to different quantum paths. The idea of coherent control is to make use of interference between these different paths to suppress or favor one specific channel. Several mechanisms in this regard have been explored. The combination of a fundamental frequency and its harmonics [2] and excitation by time delayed coherent pulses [3] are some examples. The control comes from the relative optical phase difference between different excitation fields. The attractive aspect in these experiments is the ease and the versatility in the control of this latter parameter. For example, for a sequence of pulses, it can be realized by modifying the relative delay between the pulses [4].

In a series of papers, we have applied coherent control ideas to the control of propagation effects of ultrashort pulses [5], [6], [7], [8]. An original situation arises in a double two-level system where each single two-level sub-system is driven by a strong pulse. The light shifts induced in this manner are probed by a weak pulse that connects cross transitions (the pulses are mutually coherent). One example of this situation is $(J = 1/2 \rightarrow 1/2)$ transition excited by a sequence of π-polarized strong pulse and σ-polarized weak pulse. In such system, strong interference effects take place when the relative optical phase is varied. For ultrashort pulses, we have shown that light-shifts effects can be controlled and a very efficient control of the amplification of weak pulse is obtained [5]. The scope of present paper is to investigate the general conditions and constraints, on atomic system and laser pulses, in order to achieve the phase-control of the medium gain, both in the ultrashort and long pulse regimes. We consider $(J = 1/2 \rightarrow 1/2)$ transition excited by a sequence of strong π-polarized pulse and an elliptically-polarized weak pulse. The excited states are assumed to be damped with relaxation rates that may differ. The excitation is resonant and we assume that light-shifts are significant (i.e. larger than the absorption line-width and the weak-pulse spectrum bandwidth). We will discuss situation in which the radiating coherence is phase-dependent and thus coherent phase control of medium gain is possible.

2 Theoretical Model

Consider a double two-level system consisting of left and right sub-systems $(|a\rangle, |c\rangle)$ and $(|b\rangle, |d\rangle)$ as shown in fig[1]. The excited levels $|c\rangle$ and $|d\rangle$ relax into their respective ground states with the rates Γ_π and $\Gamma_{\acute{\pi}}$ and into the crossed ground states with the rates Γ_{σ_+} and Γ_{σ_-}. The total relaxation rates from $|c\rangle$ and $|d\rangle$ are respectively $\Gamma = \Gamma_\pi + \Gamma_{\sigma_+}$ and $\acute{\Gamma} = \Gamma_{\acute{\pi}} + \Gamma_{\sigma_-}$. Each sub-system is driven by a π polarized pulse $\vec{E}_\pi$. Another pulse with elliptic polarization and zero inclination, $\vec{E}_{ellip}$ couples these systems in a crossed manner–connecting the ground state of the left sub-system to the excited state of the right and vice versa (fig[2.1,2.2]). Both these pulses are resonant with resonance frequency being ω_0. The expressions for the two pulses are given below:

$$\vec{E}_\pi(\vec{r}, t) = \vec{e}_\pi A_\pi(\vec{r}, t) e^{-i\omega_0 t} + c.c. \tag{2.1}$$

$$\vec{E}_{ellip}(\vec{r}, t) = \left(\vec{e}_+ A_{\sigma_+} + \vec{e}_- A_{\sigma_-}\right)(\vec{r}, t) e^{-i\omega_0 t} e^{-i\phi} + c.c. \tag{2.2}$$

Here A_π and $\left(A_{\sigma_+}, A_{\sigma_-}\right)$ determine the amplitude of the two pulses and ϕ is the phase difference between the two pulses (that may vary spatially). Polarization axis are given as:

$$\vec{e}_\pi = \vec{e}_z \tag{2.3}$$

$$\vec{e}_\pm = \mp \frac{\vec{e}_x \pm i\vec{e}_y}{\sqrt{2}} \tag{2.4}$$

The Hamiltonian of the system within Rotating Wave Approximation can be expressed

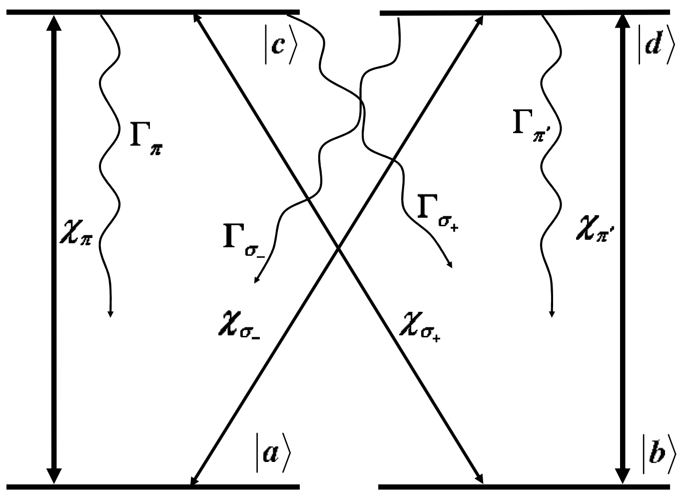

Figure 2.1: A double two level system. Field $\vec{E}_\pi$ couples $|a\rangle$ and $|c\rangle$ in the left sub-system and $|b\rangle$ and $|d\rangle$ in the right sub-system. Field $\vec{E}_{ellip}$ couples the two sub-systems in a crossed manner, connecting $|a\rangle$ to $|d\rangle$ and $|b\rangle$ to $|c\rangle$. See text for the definitions of the parameters.

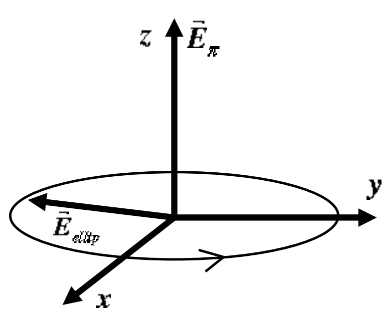

Figure 2.2: Polarization configuration for excitation fields.

in $(|a\rangle, |b\rangle, |c\rangle, |d\rangle)$ basis as:

$$H = \begin{pmatrix} 0 & 0 & -d_\pi A_\pi & -d_{\sigma_-} A_{\sigma_-} e^{i\phi} \\ 0 & 0 & -d_{\sigma_+} A_{\sigma_+} e^{i\phi} & -d_{\acute{\pi}} A_\pi \\ -d_\pi A_\pi & -d_{\sigma_+} A_{\sigma_+} e^{-i\phi} & 0 & 0 \\ -d_{\sigma_-} A_{\sigma_-} e^{-i\phi} & -d_{\acute{\pi}} A_\pi & 0 & 0 \end{pmatrix} \tag{2.5}$$

here d_j and $\chi_j = d_j A_j/\hbar$ with $j = (\pi, \acute{\pi}, \sigma_+, \sigma_-)$ are different dipole matrix elements and corresponding Rabi frequencies. We have $d_{\acute{\pi}} = -d_\pi$ and $d_{\sigma_+} = -d_{\sigma_-}$ from $3j$ relations.

The evolution of the system is governed by the equation

$$\partial_t \rho = -\frac{i}{\hbar}[H, \rho] + Relaxation \tag{2.6}$$

where the density matrix is:

$$\rho = \begin{pmatrix} \rho_{aa} & \rho_{ab} & \rho_{ac} & \rho_{ad} \\ \rho_{ba} & \rho_{bb} & \rho_{bc} & \rho_{bd} \\ \rho_{ca} & \rho_{cb} & \rho_{cc} & \rho_{cd} \\ \rho_{da} & \rho_{db} & \rho_{dc} & \rho_{dd} \end{pmatrix} \tag{2.7}$$

We can write the density matrix equations in two sets. The first set consists of populations and the coherences between states connected directly by $\vec{E}_\pi$ field. It is given as:

$$\dot{\rho_{aa}} = i\chi_\pi (\rho_{ca} - \rho_{ac}) + i\chi_{\sigma_-} \left(e^{i\phi}\rho_{da} - e^{-i\phi}\rho_{ad}\right) + (\Gamma_\pi \rho_{cc} + \Gamma_{\sigma_-}\rho_{dd}) \tag{2.8a}$$

$$\dot{\rho_{bb}} = i\chi_{\acute{\pi}} (\rho_{db} - \rho_{bd}) + i\chi_{\sigma_+} \left(e^{i\phi}\rho_{cb} - e^{-i\phi}\rho_{bc}\right) + (\Gamma_{\sigma_+} \rho_{cc} + \Gamma_{\acute{\pi}}\rho_{dd}) \tag{2.8b}$$

$$\dot{\rho_{cc}} = i\chi_\pi (\rho_{ac} - \rho_{ca}) + i\chi_{\sigma_+} \left(e^{-i\phi}\rho_{bc} - e^{i\phi}\rho_{cb}\right) - \Gamma\rho_{cc} \tag{2.8c}$$

$$\dot{\rho_{dd}} = i\chi_{\acute{\pi}} (\rho_{bd} - \rho_{db}) + i\chi_{\sigma_-} (e^{-i\phi}\rho_{ad} - e^{i\phi}\rho_{da}) - \acute{\Gamma}\rho_{dd} \tag{2.8d}$$

$$\dot{\rho_{ac}} = i\chi_\pi (\rho_{cc} - \rho_{aa}) + ie^{i\phi} \left(\chi_{\sigma_-}\rho_{dc} - \chi_{\sigma_+}\rho_{ab}\right) - \Gamma\rho_{ac}/2 \tag{2.8e}$$

$$\dot{\rho_{bd}} = i\chi_{\acute{\pi}} (\rho_{dd} - \rho_{bb}) + ie^{i\phi} \left(\chi_{\sigma_+}\rho_{cd} - \chi_{\sigma_-}\rho_{ba}\right) - \acute{\Gamma}\rho_{bd}/2 \tag{2.8f}$$

The other set consists of Zeeman coherences and the coherences between states connected directly by $\vec{E}_{ellip}$ field.

$$\dot{\rho_{ab}} = i\chi_\pi \rho_{cb} - i\chi_{\acute{\pi}}\rho_{ad} + i\chi_{\sigma_-} e^{i\phi}\rho_{db} - i\chi_{\sigma_+} e^{-i\phi}\rho_{ac} \tag{2.8g}$$

$$\dot{\rho_{cd}} = i\chi_\pi \rho_{ad} - i\chi_{\acute{\pi}}\rho_{cb} + i\chi_{\sigma_+} e^{-i\phi}\rho_{bd} - i\chi_{\sigma_-} e^{i\phi}\rho_{ca} - \left(\Gamma + \acute{\Gamma}\right)\rho_{cd}/2 \tag{2.8h}$$

$$\dot{\rho_{ad}} = i\chi_\pi \rho_{cd} - i\chi_{\acute{\pi}}\rho_{ab} + i\chi_{\sigma_-} e^{i\phi} (\rho_{dd} - \rho_{aa}) - \acute{\Gamma}\rho_{ad}/2 \tag{2.8i}$$

$$\dot{\rho_{cb}} = i\chi_\pi \rho_{ab} - i\chi_{\acute{\pi}}\rho_{cd} + i\chi_{\sigma_+} e^{-i\phi} (\rho_{bb} - \rho_{cc}) - \Gamma\rho_{cb}/2 \tag{2.8j}$$

Here we can make a distinction between the ultra-short pulse regime and long pulse regime. We will show that the physics of coherent control is very different in these two situations. We are interested in the qualitative behavior of the medium gain seen by the $\vec{E}_{ellip}$ field in presence of $\vec{E}_{\pi}$ field. We consider the radiating coherences ρ_{cb} and ρ_{da} that are responsible for the modification of $\vec{E}_{ellip}$ field.

2.1 Ultra-short pulse regime

With ultra-short pulses, relaxation processes become negligible. The analytical solution of the density matrix equations (2.8) is possible using perturbative development. However the physics of coherent control is better highlighted in adiabatic states of the atom dressed by the $\vec{E}_{\pi}$ field. We define the rotation matrix as:

$$R = \frac{1}{\sqrt{2}} \begin{pmatrix} 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & -1 \\ -1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 \end{pmatrix} \tag{2.9}$$

The dressed hamiltonian is given by the transformation:

$$\begin{aligned} H_{dressed} &= R \cdot H.R^{\dagger} \\ &= \begin{pmatrix} -\chi_{\pi} & C_{\alpha\beta} & 0 & C_{\alpha\delta} \\ C_{\beta\alpha} & \chi_{\acute{\pi}} & C_{\beta\gamma} & 0 \\ 0 & C_{\gamma\beta} & \chi_{\pi} & C_{\gamma\delta} \\ C_{\delta\alpha} & 0 & C_{\delta\gamma} & -\chi_{\acute{\pi}} \end{pmatrix} \end{aligned} \tag{2.10}$$

here $(|\alpha\rangle, |\beta\rangle, |\gamma\rangle, |\delta\rangle)$ are transformed $(|a\rangle, |b\rangle, |c\rangle, |d\rangle)$ states and $C_{ij} = C^*_{ji}$ are coupling elements between these states due to $\vec{E}_{ellip}$ field. In this representation, the energies of states $|\alpha\rangle$ and $|\beta\rangle$ are down shifted by χ_{π}(note that$\chi_{\acute{\pi}} = -\chi_{\pi}$) whereas the energies of states $|\gamma\rangle$ and $|\delta\rangle$ are up shifted by the same quantity. The key point in this representation is the differentiation between parallel and anti-parallel transition as shown in (fig[2.3]). The anti-parallel transitions take place between states that are oppositely light shifted i.e. $|\alpha\rangle \leftrightarrow |\delta\rangle$ and $|\beta\rangle \leftrightarrow |\gamma\rangle$. The coupling elements for these transitions are given by:

$$\begin{pmatrix} C_{\alpha\delta} \\ C_{\beta\gamma} \end{pmatrix} = -\frac{1}{2} \begin{pmatrix} \chi_{\sigma_+} e^{-i\phi} + \chi_{\sigma_-} e^{i\phi} \\ \chi_{\sigma_+} e^{i\phi} + \chi_{\sigma_-} e^{-i\phi} \end{pmatrix} \tag{2.11}$$

For important light shifts such as $\chi_{\pi} T >> 1$ where T is the pulse duration of $\vec{E}_{ellip}$ field, the interaction of the $\vec{E}_{ellip}$ field is not resonant and one expects a vanishing contribution to the radiating coherences. The parallel transitions occur between states that are identically light shifted i.e. $|\alpha\rangle \leftrightarrow |\beta\rangle$ and $|\gamma\rangle \leftrightarrow |\delta\rangle$. The coupling elements are:

$$C_{\alpha\beta} = C_{\gamma\delta} = \frac{-\chi_{\sigma_+} e^{-i\phi} + \chi_{\sigma_-} e^{i\phi}}{2} \tag{2.12}$$

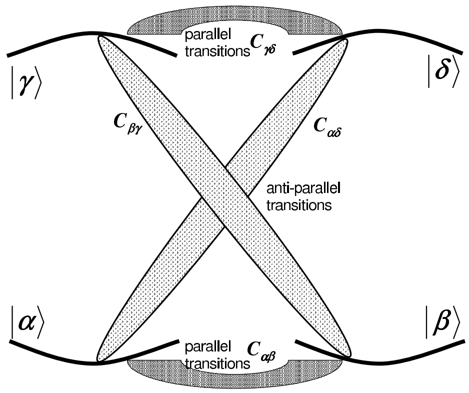

Figure 2.3: Dressed state representation of $\vec{E}_{ellip}$ interaction with the system. $\vec{E}_{ellip}$ can induce parallel and anti-parralel transitions. The relaxation paths are not shown. See text for the definitions of the parameters.

The interaction for this latter case is always resonant, leading to a non-vanishing contribution to the radiating coherences. Phase control of the medium gain is thus possible. The optimal control is obtained when $\left|\chi_{\sigma_+}\right| = \left|\chi_{\sigma_-}\right|$, for which the maximum contrast is obtained for the coupling elements (2.12) when varying the relative phase ϕ. Moreover if $\chi_{\sigma_+} = \chi_{\sigma_-}$ (x-polarized field), we get $C_{\alpha\beta} = C_{\gamma\delta} = i\chi_{\sigma_+}\sin\phi$. The medium is transparent only for $\phi = 0$ (and $\chi_\pi T >> 1$). If $\chi_{\sigma_+} = -\chi_{\sigma_-}$ (y-polarized field), we get $C_{\alpha\beta} = C_{\gamma\delta} = -\chi_{\sigma_+}\cos\phi$. The medium is transparent only for $\phi = \pi/2$ (and $\chi_\pi T >> 1$).

2.2 Long pulse regime

In continuous regime the relaxation processes overshadow the parallel/anti-parallel description of interaction and we have to go back to the bare state. Analytical solution for the stationary regime of (2.8) can be worked out. The coherences ρ_{cb} and ρ_{da} responsible for the modification of $\vec{E}_{ellip}$ field are given at the first order (with respect to $\vec{E}_{ellip}$ field) as:

$$\rho_{cb}^{(1)} = 2i \frac{\left[\left(\chi_{\acute{\pi}}^2 + \frac{\acute{\Gamma}^2}{4}\right) - \frac{\Gamma_{\sigma_-}}{\Gamma_{\sigma_+}}\left(\chi_{\acute{\pi}}^2 + \frac{\Gamma\acute{\Gamma}}{4}\right)\right]\chi_{\sigma_+} e^{i\phi} + \left[\frac{\Gamma_{\sigma_-}}{\Gamma_{\sigma_+}}\left(\chi_\pi^2 + \frac{\Gamma^2}{4}\right) + \left(\frac{\acute{\Gamma}^2}{4} + \chi_\pi \chi_{\acute{\pi}}\right)\right]\chi_{\sigma_-} e^{-i\phi}}{\chi_{\acute{\pi}}^2 \Gamma + \chi_\pi^2 \acute{\Gamma}} \rho_{dd}^{(0)} \tag{2.13}$$

$$\rho_{da}^{(1)} = 2i \frac{\left[\frac{\Gamma_{\sigma_-}}{\Gamma_{\sigma_+}}\left(\chi_\pi^2 + \frac{\Gamma^2}{4}\right) - \left(\chi_\pi^2 + \frac{\Gamma\acute{\Gamma}}{4}\right)\right]\chi_{\sigma_-} e^{i\phi} + \left[\chi_{\acute{\pi}}^2 + \frac{\acute{\Gamma}^2}{4} + \frac{\Gamma_{\sigma_-}}{\Gamma_{\sigma_+}}\left(\frac{\Gamma^2}{4} + \chi_\pi \chi_{\acute{\pi}}\right)\right]\chi_{\sigma_+} e^{-i\phi}}{\chi_{\acute{\pi}}^2 \Gamma + \chi_\pi^2 \acute{\Gamma}} \rho_{dd}^{(0)} \tag{2.14}$$

with

$$\rho_{dd}^{(0)} = \frac{(\chi_{\acute{\pi}})^2 (\chi_\pi)^2 \Gamma_{\sigma_+}}{(\chi_\pi)^2 \left(+\frac{\acute{\Gamma}^2}{4} + 2(\chi_{\acute{\pi}})^2\right)\Gamma_{\sigma_+} + (\chi_{\acute{\pi}})^2 \left(+\frac{\Gamma^2}{4} + 2(\chi_\pi)^2\right)\Gamma_{\sigma_-}} \tag{2.15}$$

We can see that for arbitrary driving field strength the possibility of phase control is not obvious. However if the driving field $\vec{E}_\pi$ is strong enough ($\chi_\pi >> (\Gamma, \acute{\Gamma})$), it saturates the system. In this limit, we obtain:

$$\rho_{cb}^{(1)} = \frac{2i}{\Gamma + \acute{\Gamma}} \left(\Gamma_{\sigma_+} - \Gamma_{\sigma_-}\right) \left(\chi_{\sigma_+} e^{i\phi} - \chi_{\sigma_-} e^{-i\phi}\right) \tag{2.16a}$$

$$\rho_{da}^{(1)} = -\frac{2i}{\Gamma + \acute{\Gamma}} \left(\Gamma_{\sigma_+} - \Gamma_{\sigma_-}\right) \left(\chi_{\sigma_-} e^{i\phi} - \chi_{\sigma_+} e^{-i\phi}\right) \tag{2.16b}$$

We see that we have the possibility of coherent control of the medium gain for $\vec{E}_{ellip}$ field provided that $\Gamma_{\sigma_+} - \Gamma_{\sigma_-}$ does not vanish. For the system where relaxation processes favor one channel over the other (Γ_{σ_+} over Γ_{σ_-} or vice verse), the control becomes possible. The maximum control is again exerted for the case when $\left|\chi_{\sigma_-}\right| = \left|\chi_{\sigma_+}\right|$. If $\chi_{\sigma_-} = \chi_{\sigma_+}$ (x-polarized field), we have

$$\rho_{cb}^{(1)} = -\frac{4}{\Gamma + \acute{\Gamma}} \left(\Gamma_{\sigma_+} - \Gamma_{\sigma_-}\right) \chi_{\sigma_+} \sin\phi \tag{2.17a}$$

$$\rho_{da}^{(1)} = \frac{4}{\Gamma + \acute{\Gamma}} \left(\Gamma_{\sigma_+} - \Gamma_{\sigma_-}\right) \chi_{\sigma_+} \sin\phi \tag{2.17b}$$

and if $\chi_{\sigma_-} = -\chi_{\sigma_+}$ (y-polarized field) we get:

$$\rho_{cb}^{(1)} = \frac{4i}{\Gamma + \acute{\Gamma}} \left(\Gamma_{\sigma_+} - \Gamma_{\sigma_-}\right) \chi_{\sigma_+} \cos\phi \tag{2.18a}$$

$$\rho_{da}^{(1)} = -\frac{4i}{\Gamma + \acute{\Gamma}} \left(\Gamma_{\sigma_+} - \Gamma_{\sigma_-}\right) \chi_{\sigma_+} \cos\phi \tag{2.18b}$$

Two important remarks can be made here. First, the bleaching of the atomic system by the strong field $\vec{E}_\pi$ doesn't induce transparency in the medium if $\Gamma_{\sigma_+} \neq \Gamma_{\sigma_-}$. This is because in the saturation limit, the populations are equally distributed in the ground and excited levels for both left and right sub-system but not between crossed levels. Transitions between these latter still lead to the emission of radiation. Secondly, in the adiabatic description, when ($\chi_\pi >> (\Gamma, \acute{\Gamma})$) no anti-parallel transitions occur. This is because the light shifts exceeds largely the absorption line width. Only parallel transitions are efficient leading to radiating coherence proportional to the parallel coupling element (2.12).

3 Conclusion

We have shown in this paper, the possibility to achieve phase control of the interaction between an elliptically-polarized probe pulse and a double two-level system driven by a strong π-polarized pulse. In the ultra-short regime, the control is always possible and the optimum control is obtained for $\chi_{\sigma_-} = \pm\chi_{\sigma_+}$ where χ_{σ_+} and χ_{σ_-} are the Rabi frequencies of σ_+ and σ_- components of probe pulse. This control can be understood in dressed regime as an interplay between parallel and anti parallel-transitions between the states that are light shifted in the same sense or in opposite sense respectively. For the long pulse regime, and in the saturation limit, the control is not possible except in a particular situation where the population damping rate from excited state $|J = 1, M_J = 1/2\rangle$ to the ground state $|J = 1, M_J = -1/2\rangle$ differs from the population damping rate from excited state $|J = 1, M_J = -1/2\rangle$ to $|J = 1, M_J = 1/2\rangle$. This situation may occur when the atomic system is embedded in a polarized buffer gas leading to spin-dependence of the collision cross-sections [9]. The observation of phase-dependence of the medium gain in such sample may become a sensitive tool to measure the polarization of the buffer gas.

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