Electromagnetically induced transparency and two-photon Raman resonances

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1 Dark state

There are several ways to explain EIT, but probably the most intuitive one involves introduction of a so-called "dark state" - a quantum superposition of the atomic levels that is decoupled from the interacting laser fields. A traditional EIT arrangement includes a three-level atomic (or atom-like) system, in which two of the levels are coupled to the common third level via two near-resonant electromagnetic fields. Assuming that each optical field interacts with only its corresponding transition, the interaction Hamiltonian for such system can be written as:

$$\hat{H} = \begin{pmatrix} -\hbar\omega_{13} & 0 & -\wp_{13}E_1 \\ 0 & -\hbar\omega_{23} & -\wp_{23}E_2 \\ -\wp_{13}E_1 & -\wp_{23}E_2 & 0 \end{pmatrix}, \tag{1}$$

where ω_{13} and ω_{23} are the frequency of the corresponding atomic transitions (here we place zero evenrgy level at the state $|3\rangle$), $E_{1,2}exp(-i\nu_{1,2}t) + c.c.$ are the electromagnetic fields interacting with each atomic transition $\wp_{ij}$ is the dipole moment of a corresponding atomic transition. To simplify this discussion, we for now consider resonant conditions $\nu_1 = \omega_{13}$ and $\nu_2 = \omega_{23}$. In this case it is easy to show that one of the eigenstates $|D\rangle$ of such interaction Hamiltonian has zero eigenvalue, such that $\hat{H}|D\rangle = 0$. Thus, any atom in such state is effectively decoupled from either laser fields, rendering such state "non-interacting". Moreover, such a state contains only two of three states, involved in the interaction:

$$|D\rangle = \mathcal{N}\left(\Omega_1|2\rangle - \Omega_2|1\rangle\right), \tag{2}$$

where $\mathcal{N} = \sqrt{\Omega_1^2 + \Omega_2^2}$ is the normalization factor. Note, that such dark state is universal, and does not depend on either relative energies of the chosen atomic states or the strengths of the optical fields, down to single-photon fields in a fully quantum EIT treatment.

In a typical EIT arrangement, the levels are chosen such that states $|1\rangle$ and $|2\rangle$ have longer lifetime than the state $|3\rangle$. Especially in the Λ configuration, the first two states are chosen among the ground states sublevels. In this case, the atoms in the dark state cannot be excited into the electronic excited states, prohibiting the fluorescence and making the atom "dark" for the external observer. At the same time, the absence of the spontaneous emission removes the dominant optical loss mechanism, so the laser fields can now propagate through the resonant atomic medium without any absorption. By analogy, we can introduce the orthogonal "bright" superposition $|B\rangle = \mathcal{N}(\Omega_1|1\rangle + \Omega_2|2\rangle)$, such that is coupled to the excited state $|3\rangle$, and the interaction Hamiltonian may be written as

$$\hat{H} = \hbar\sqrt{\Omega_1^2 + \Omega_2^2}\,(|3\rangle\langle B| + |B\rangle\langle 3|) \tag{3}$$

It is easy to see that the relative phase of the atomic superposition is critical to ensure the non-interaction condition for the dark state. For example, if the phase of one of the fields is suddenly flipped by π, thus changing the minus sign in the dark state to plus, the atoms temporarily become absorbing, until the atomic coherence is adjusted to the new conditions. By using such arguments, we can qualitatively explain the spectral width of the EIT resonance, although accurate derivation require using density matrix formalism, described in the next subsection.

For now, we specifically assumed that frequencies of each optical field match exactly the frequencies of the corresponding transitions. One can show that even for non-zero laser detunings, the steady-state dark state of Eq.(1) exists for the zero two-photon detuning $\delta = 0$, where we define $\delta = \nu_1 - \nu_2 - (\omega_{13} - \omega_{23}$ as the mismatch between the two-photon transition, formed by the two lasers, and the frequency difference between states $|1\rangle$ and $|\rangle$. If a small non-zero detuning δ is introduced, the state of atoms initially prepared in $|D\rangle$ is going to evolve in time as:

$$|D_\delta(t)\rangle = \mathcal{N}\left(\Omega_1|2\rangle - e^{i\delta\cdot t}\Omega_2|1\rangle\right), \tag{4}$$

causing the sign of the dark state phase to slowly change. Since in reality atoms cannot maintain their coherence forever, the dark state can exist only for a finite lifetime τ_{coh}. So if the two-photon detuning is small, such that the accumulated phase $\delta \cdot \tau_{coh} \ll 1$ is negligible, the dark state stays largely non-interacting and EIT is preserved. But as detuning increases, the effect of the phase evolution becomes more pronounced. in fact, we can roughly estimate the spectral EIT width to be inversely proportional to the dark state lifetime using $\delta \cdot \tau_{coh} \approx \pi/2$. Such estimate is quite accurate in the limit of very weak optical fields. We can also use this model to qualitatively explain the power broadening: the increase of the EIT linewidth with the power of the optical fields. Eq.(1) assumes free evolution of the atomic state; however, the stronger are the optical fields, the larger is the probability to the evolving state to be rephased by the repeated interaction, shortening the free phase evolution and thus increasing the transparency tolerance to the non-zero detuning.

2 Note on terminology: CPT vs EIT vs Raman

One of the difficulties of the literature search about EIT-related research is difference in terminology different scientists use: for example, the two-photon transmission resonances can be referred to as electromagnetically-induced transparency (EIT), coherent population trapping (CPT), dark resonances, and Raman resonances. Moreover, different people sometimes put slightly different differentiation in each of this term, so here we outline what we perceive as most common definitions.

CPT is often referred to the experimental arrangement involving a Λ-system with two long-lived energy levels (typically two hyperfine or Zeeman ground state sublevels) and two optical fields of comparable strength. In this case atoms "trapped" in the quantum superposition with close to maximum coherence, and can be thought as a generalization of the optical pumping process.

Such configuration is most common for metrology applications, such as CPT-based atomic clocks, magnetometers, etc.

EIT is a more general case in which the transmission for a resonant optical field is enhanced by means of another optical field, particularly without reduction of the atomic population on the corresponding transition. This effect can happen in any three-level system (Λ, V or ladder configuration), and, in principle, for arbitrary values of the optical fields. However, most often EIT experiments imply one strong ("pump" or "control") and one "weak" ("probe" or "signal") optical fields. In this arrangement the EIT looks the most "counterintuitive", especially in the ladder system, where adding a strong pump field between nominally empty excited states changes probe absorption dramatically without noticeably changing atomic populations. Indeed, according to the dark state Eq.(1), for $\Omega_1 \ll \Omega_2$ the population of the state $|1\rangle$, coupled to the weaker optical field, is $|\Omega_2/\sqrt{\Omega_1^2+\Omega_2^2} \approx 1$. Such regime is also most relevant to quantum information applications, in which EIT is used for realization of the strong coupling between quantum optical fields and long-lived atomic states, in particular for the slow light and quantum memory.

EIT is of course a small subset of general two-photon Raman processes. However, in the context of light-atom interaction, often Raman resonances are referred to the case of narrow absorption resonances appearing in a Λ system when the two optical fields are detuned from the excited state, while maintaining the two-photon resonance (we will consider the effect of the laser detuning of EIT below). In the last decade Raman resonances became a viable alternative to EIT for quantum memory applications.

3 Density matrix description of the EIT

While the concept of the dark state provides an intuitive insight into the nature of EIT, the accurate description of this process requires proper account of the decoherence processes for both optical transitions and, even more importantly, the atomic coherence associated with the dark state. Since the wave function formalism is not adequate for describing quantum systems in the presence of decoherence, we have to utilize the density matrix formalism. Since we will be mostly interested in the steady-state or slowly varying atomic evolution, we can apply the rotating wave approximation and removing all fast oscillating terms. In this case the Hamiltonian $\hat{H}$, given by Eq.(1 can be rewritten as:

$$\frac{\hat{H}_{RWA}}{\hbar} = \begin{pmatrix} -\Delta_1 & 0 & -\Omega_1 \\ 0 & -\Delta_2 & -\Omega_2 \\ -\Omega_1^* & -\Omega_2^* & 0 \end{pmatrix}. \tag{5}$$

Here, $\Omega_{1,2}$ are the Rabi frequencies, associated with each slowly varying amplitudes $\tilde{E}_{1,2}$ of the electromagnetic field, $\Omega_i = \wp_{i3}\tilde{E}/2\hbar$, $\Delta_i = \nu_i - \omega_{i,3}$ is the one-photon detuning of each laser from its corresponding transition. In this case the evolution of the atomic state matrix $\hat{\rho}$ under the action of the , is described by the following Maxwell-Block equation:

$$i\hbar\frac{d\rho}{dt} \hat{=} \left[\hat{H}_{RWA}, \hat{\rho}\right] + \frac{1}{2}\left\{\hat{\Gamma}, \hat{\rho}\right\}, \tag{6}$$

where the matrix $\hat{\Gamma}$ contains information about all the decoherence effects. We will discuss the specific effects of various aspects of the environment further in the manuscript, at that point we

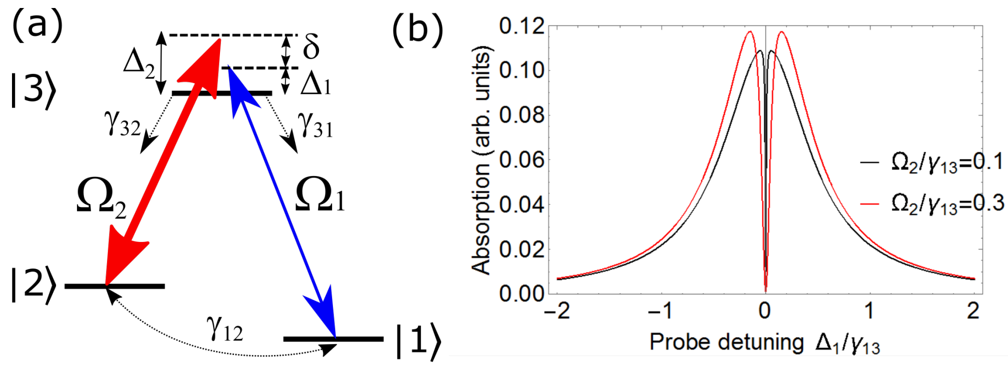

Figure 1: (a) A three-level Λ system, considered in the calculations below. (b) Example of the narrow transmission resonance within the probe field homogeneous absorption profile due to EIT for two different values of the control field. In this example $\gamma_{12} = 0.001\gamma_{13}$,$\Delta_2 = 0$, and $\Omega_1 = 0.001\gamma_{13}$.

will introduce these decoherence rates phenomenologically: γ_i is the population decay rate of the ith state (in case of a state having more than one decay channel, r_{ij} is the branching ratio to the state j), and γ_{ij} is the decoherence rate of the corresponding off-diagonal matrix element ρ_{ij}. Here we will also assume a closed system (i.e., there is no population exchange to the outside of the three atomic levels), although it has been shown that the corresponding calculations for the open system result in very similar outcome. Finally, since the Λ system, shown in Fig. 2 is by far more common EIT configuration, in the following we will assume that the states $|1\rangle$ and $|2\rangle$ are sublevels of the ground electronic state and experience no spontaneous emission, coupled to the common excited electron state $|3\rangle$. In this case, the time evolution equations for the density matrix elements are:

$$
\begin{aligned}
\dot{\rho}_{11} &= r_{31}\gamma_3 - i\Omega_1\rho_{13} + i\Omega_1^*\rho_{31} \\
\dot{\rho}_{22} &= r_{32}\gamma_3 - i\Omega_2\rho_{23} + i\Omega_2^*\rho_{32} \\
\dot{\rho}_{21} &= -(\gamma_{12} - i\delta)\rho_{21} - i\Omega_1\rho_{23} + i\Omega_2^*\rho_{31} \\
\dot{\rho}_{31} &= -(\gamma_{13} - i\Delta_1)\rho_{31} + i\Omega_2\rho_{21} + i\Omega_1(\rho_{11} - \rho_{33}) \\
\dot{\rho}_{32} &= -(\gamma_{23} - i\Delta_2)\rho_{32} + i\Omega_1\rho_{12} + i\Omega_2(\rho_{22} - \rho_{33})
\end{aligned}
\tag{7}
$$

These equations can provide the exact solutions for any values of parameters *and hopefully we will have some numerical tool to explore these solutions.* In this section we will consider only the steady-state solutions to analyze the characteristics of the EIT transmission resonances. In this case, even though Eqs.(7) becomes a system of linear equations and can be solved analytically, the resulting expressions are rather cumbersome. So here we analyze the most common case of the strong pump - weak probe regime ($\Omega_1 \ll \Omega_2$), in which the system response to the probe field is linear, and thus we can simplify the solution, keeping only the linear terms in Ω_1.

In this approximation it is convenient to use the perturbative approach to the solution. If there is no probe field $\Omega_1 = 0$, all atomic population is optically pumped into the $|2\rangle$ state (assuming that the pump field is sufficiently strong to provide efficient optical pumping): $\rho_{11}^{(0)} = 1$, and $\rho_{22}^{(0)} = \rho_{33}^{(0)} = 0$. Also $\rho_{23}^{(0)} = 0$, since this is the coherence between two empty states. Substituting these values into the right hand side of Eqs.(7) and keeping only linear terms in Ω_1, we can substantially simplify the solution, since only two equations remain:

$$\begin{aligned} 0 &= -\Gamma_{12}\rho_{21} + i\Omega_2^*\rho_{31} \\ 0 &= -\Gamma_{13}\rho_{31} + i\Omega_2\rho_{21} + i\Omega_1, \end{aligned} \tag{8}$$

where we use $\Gamma_{12} = \gamma_{12} - i\delta$ and $\Gamma_{13} = \gamma_{13} - i\Delta_1$. This leads to very simple and elegant expressions for the ground state and optical coherences:

$$\rho_{21} = -\frac{\Omega_1\Omega_2^*}{\Gamma_{12}\Gamma_{13} + |\Omega_2|^2} \tag{9}$$

$$\rho_{31} = i\Omega_1\frac{\Gamma_{12}}{\Gamma_{12}\Gamma_{13} + |\Omega_2|^2}, . \tag{10}$$

Then the probe linear susceptibility χ_p is:

$$\chi_p(\Delta_1, \delta) = \frac{\wp_{13}^2}{\hbar\epsilon_0}\frac{\rho_{31}}{\Omega_1} = i\frac{\wp_{13}^2}{\hbar\epsilon_0\Gamma_{13}}\frac{\Gamma_{12}\Gamma_{13}}{\Gamma_{12}\Gamma_{13} + |\Omega_2|^2}. \tag{11}$$

It is easy to see that in the ideal case of no decoherence between the states $|1\rangle$ and $|2\rangle$ $\gamma_{12} = 0$ and zero two-photon detuning $\delta = 0$ (i.e., for $\Gamma_{12} = 0$, the susceptibility completely vanishes, resulting is 100% transparency for the probe field. This result is particularly counter-intuitive when the lasers are tune close to atomic resonance, where we would expect strong resonant absorption for the probe field due to large atomic population in the state $|1\rangle$. This potential elimination of resonant absorption by applying a strong additional electromagnetic field on a different transition is the origin of the name for electromagnetically-induced transparency.

Resonant EIT; power broadening Let us fist consider the case of the probe laser tuned exactly to the atomic resonance $\Delta_1 = 0$, but allow a small two-photon detuning $\delta \ll \gamma_{13}$. In this case we can derive the canonical expression for the EIT resonance susceptibility by subsituting $\Gamma_{13} = \gamma_{13}$ and $\Gamma_{12} = \gamma_{13} - i\delta$ in Eq.(11):

$$\chi_p(\delta) = i\frac{\wp_{13}^2}{\hbar\epsilon_0\gamma_{13}}\frac{\left[\gamma_{12}\gamma_{EIT} + \delta^2\right] - i\delta|\Omega_2|^2/\gamma_{13}}{\gamma_{EIT}^2 + \delta^2}, \tag{12}$$

where $\gamma_{EIT} = \gamma_{12} + |\Omega_2|^2/\gamma_{13}$. The probe absorption coefficient $\alpha = k/2Im(\chi_p)$ in this case is:

$$\alpha_p(\delta) = \alpha_0\frac{\gamma_{12}\gamma_{EIT} + \delta^2}{\gamma_{EIT}^2 + \delta^2}, \tag{13}$$

where $\alpha_0 = \frac{k\wp_{13}^2}{2\hbar\epsilon_0\gamma_{13}}$ is the unsaturated absorption for the $|1\rangle - |3\rangle$ optical transition (without the control field). It is easy to see that at exact two-photon resonance $\delta = 0$ the probe absorption is suppressed by the factor $\frac{\alpha_{EIT}}{\alpha_0} = \frac{\gamma_{12}}{\gamma_{EIT}} = \frac{\gamma_{12}}{\gamma_{12}+|\Omega_2|^2/\gamma_{13}}$. Realistically, we can approach high transparency in the limit of the strong pump field $|\Omega_2| \gg \sqrt{\gamma_{12}\gamma_{13}}$, resulting in the vanishing absorption suppression factor $\frac{\alpha_{EIT}}{\alpha_0} = \frac{\gamma_{12}\gamma_{13}}{|\Omega_2|^2} \to 0$.

Eq.(13 also predicts the linewidth of the EIT transmission resonance $\gamma_{EIT} = \gamma_{12} + |\Omega_2|^2/\gamma_{13}$. It is easy to see that for the very weak control field the resonance width is limited by the decoherence rate $\gamma_1 2$, which can in principle be very small, especially in the case of a Λ interaction system.

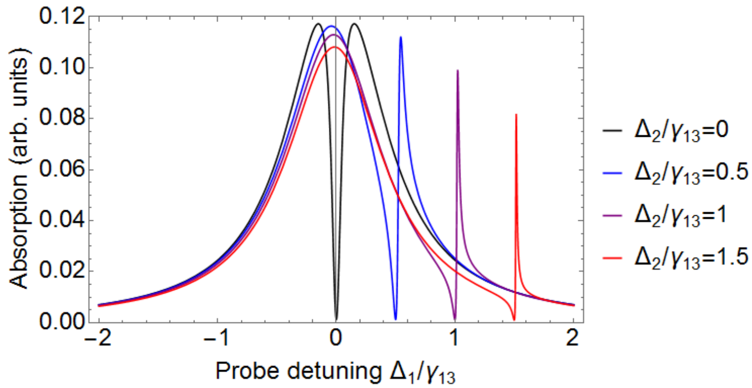

Figure 2: Evolution of the two-photon resonance lineshape for different control field detunings Δ_2. In this example $\gamma_{12} = 0.001\gamma_{13}$, $\Omega_1 = 0.001\gamma_{13}$, and $\Omega_2 = 0.3\gamma_{13}$.

As the control power increases, the EIT resonance broadens proportionally. For most practical applications the balance between such power broadening (a narrow resonance needs lower control power) and the absorption suppression factor (higher transmission needs higher control power) determines the optimal control field parameters.

Two-photon resonances in case of non-zero laser detuning

Let us now consider another limiting case in which the probe field is detuned relatively far away from the corresponding atomic transition, such that $\Delta_1 \gg \gamma_{13}$. To analyze the probe absorption it is convenient to rewrite Eq.(11) as:

$$\chi_p(\Delta_1, \delta) = i\frac{\wp_{13}^2}{\hbar\epsilon_0\Gamma_{13}} - i\frac{\wp_{13}^2}{\hbar\epsilon_0\Gamma_{13}}\frac{|\Omega_2|^2}{\Gamma_{12}\Gamma_{13} + |\Omega_2|^2}. \tag{14}$$

In such form we can easily identify the first term as a resonant probe interaction, while the second term describes the control field effect. One can check that the largest relative contribution from the second term happens near the two-photon resonance $\delta \ll \Delta_1$. Then, taking into account $\Delta_1 \gg \gamma_{13}, \gamma_{12}$, we can simplify the expression for the off-resonant probe susceptibility as:

$$\chi_p(\Delta_1, \delta) = i\alpha_0\frac{\gamma_{13}}{\gamma_{13} - i\Delta_1} + i\alpha_0\frac{|\Omega_2|^2/\Delta_1^2}{\gamma_R - i(\delta - \delta_R)}. \tag{15}$$

Here again the first term is the residual linear susceptibility, while the second term corresponds to a two-photon Raman absorption resonance with the width $\gamma_R = \gamma_{12} + \gamma_{13}|\Omega_2|^2/\Delta^2 \ll \gamma_{13}$, shifted from the exact two-photon resonance by $\delta_R = |\Omega_2|^2/\Delta$.