Electromagnetically induced transparency and coherent-state preparation in optically thick media

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Abstract: The preparation of an optically dense ensemble of three-level systems in dark states of the interaction with coherent radiation is discussed. It is shown that methods involving spontaneous emissions of photons such as Raman optical pumping fail to work beyond a critical density due to multiple scattering and trapping of these photons and the associated decay of the dark state(s). In optically thick media coherent-state preparation is only possible by entirely coherent means such as stimulated Raman adiabatic passage (STIRAP). It is shown that STIRAP is the underlying physical mechanism for electromagnetically induced transparency (EIT).

OCIS codes: (020.1670) Coherent optical effects; (020.7010) Trapping;

References

1. for a recent review on EIT see: S. E. Harris, "Electromagnetically induced transparency," Phys. Today **50**, 36 (1997) and references therein.
2. for a recent review see: E. Arimondo, "Coherent population trapping in laser spectroscopy," Prog. Opt. **35**, 259 (1996).
3. S. E. Harris and Zhen-Fei. Luo, "Preparation Energy for Electromagnetically Induced Transparency," Phys. Rev. A **52**, R928 (1995).
4. for a recent review on STIRAP see: K. Bergmann, H. Theuer, and B. W. Shore, "Coherent Population Transfer Among Quantum States of Atoms and Molecules," Rev. Mod. Phys. **70**, 1003 (1998).
5. T. Holstein, "Imprisonment of resonance radiation in gases," Phys. Rev. A **72**, 1212 (1947).
6. Michael Fleischhauer, "Optical pumping in dense atomic media: Limitations due to reabsorption of spontaneously emitted photons," http://xxx.lanl.gov/abs/quant-ph/9811017.
7. Michael Fleischhauer and Aaron S. Manka, "Propagation of laser pulses and coherent population transfer in dissipative three-level systems: An adiabatic dressed-state picture," Phys. Rev. A. **54**, 794 (1996).
8. Michael Fleischhauer and Susanne F. Yelin, "Radiative atom-atom interactions in optically dense media: Quantum corrections to the Lorentz-Lorenz formula," http://xxx.lanl.gov/abs/quant-ph/9809087.
9. C. M. Bowden and J. Dowling, "Near-dipole-dipole effects in dense media: Generalized Maxwell-Bloch equations," Phys. Rev. A **47**, 1247 (1993).
10. A. Rahman, R. Grobe, and J. H. Eberly, "Two-Photon Beer's Law for Coherently Prepared Three-Level Media," *Coherence and Quantum Optics VII* p.449 (Plenum, New York, 1996).

1. Introduction

The possibility to render optically thick media transparent for coherent laser radiation via electromagnetically induced transparency (EIT) [1] gained considerable interest over the last years. This is due to its potentials to utilise the large dispersive and nonlinear response close to atomic resonances without suffering from linear absorption. Many interesting applications have been proposed and in part experimentally realized.

Examples are resonantly enhanced nonlinear processes which do not require external phase-matching tools, new mechanisms for laser operation, novel spectroscopic applications and optical magnetometer, quantum noise reduction, applications in sub-recoil laser cooling, cavity QED, and quantum-non-demolition measurements.

All these implemented or potential applications are based on the cancellation of the linear absorption due to trapping of population in a decoupled or dark state [2] which is a coherent superposition of meta-stable atomic states. An important question is thereby how the system is prepared in such a state. The most obvious mechanism resulting into the creation of a trapping state is Raman optical pumping. In Ref.[3] an alternative mechanism for establishing EIT for pulses, namely stimulated Raman adiabatic passage (STIRAP) [4] was discussed.

In the present paper it will be shown that in optically thick media where EIT and its applications are particularly interesting, there is a qualitative difference between the two mechanisms. It will be argued that Raman optical pumping fails to work beyond some critical density and only the entirely coherent mechanism of STIRAP is able to prepare the system in a dark state. This has important consequences for some applications of coherent population trapping, such as sub-recoil laser cooling or coherent information storage in optically dense media.

The essential difference between EIT in optically thick and dilute media results from the different role of spontaneous emission. In a dilute medium spontaneously emitted photons simply escape and lead to an irreversible loss of energy. In an optically thick medium these photons get re-absorbed or scattered by other atoms, which leads to a phenomenon known as radiation trapping [5]. As a consequence the rate of optical pumping is substantially slowed down which can make population transfer by this mechanism entirely impossible [6]. On the other hand STIRAP is based on a slow (adiabatic) rotation of the dark state from the initially populated ground state into the coherent superposition which does not involve spontaneous emissions [4].

2. Radiation trapping in optically thick three-level systems

We here consider a dense ensemble of three level atoms driven by two coherent fields as shown in Fig. 1a. For simplicity we assume equal but orthogonal dipole moments of the two optical transitions and correspondingly equal radiative decay rates. Both fields are co-propagating, have fixed relative phases and are in two-photon resonance. Levels $|b\rangle$ and $|c\rangle$ are assumed to be metastable, but we take into account a (e.g. collisional) de-phasing of the $|b\rangle - |c\rangle$ coherence with rate γ_0.

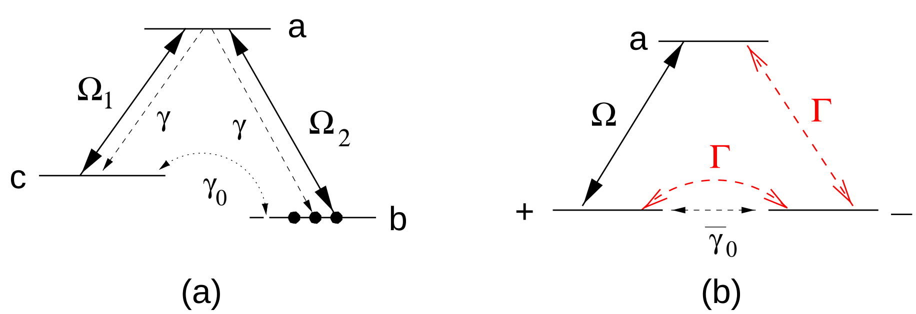

Fig.1: Three level system driven by two coherent fields with real Rabi frequencies Ω_1 and Ω_2 in (a) bare atomic basis and (b) basis of dark ($|-\rangle$) and bright ($|+\rangle$) states.

Initially, i.e. without the fields, all population is in state $|b\rangle$. We consider the case of large Doppler-broadening of the two optical transitions with the (same) Gaussian

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distribution of width Δ_D. Two-photon Doppler-broadening of the $|b\rangle - |c\rangle$ coherence is neglected.

The interaction of the fields with the atoms is most conveniently described in a rotating frame using the so-called dark–bright basis, consisting of the excited bare state $|a\rangle$, the decoupled or dark state $|-\rangle$, and the orthogonal bright state $|+\rangle$:

$$|-\rangle \equiv \frac{1}{\Omega(t)}\big(\Omega_2(t)\,|c\rangle - \Omega_1(t)\,|b\rangle\big), \qquad |+\rangle \equiv \frac{1}{\Omega(t)}\big(\Omega_2(t)\,|b\rangle + \Omega_1(t)\,|c\rangle\big). \tag{1}$$

This is illustrated in Fig. 1b. $\Omega(t) = \sqrt{\Omega_1^2(t) + \Omega_2^2(t)}$ is the effective total Rabi-frequency, where $\Omega_{1,2}(t)$ are the slowly-varying Rabi-frequencies of the laser fields, and $\Omega_1(t) = \Omega(t)\,\cos\theta(t)$ and $\Omega_2(t) = \Omega(t)\,\sin\theta(t)$.

In the slowly-varying amplitude and phase approximation (SVEA) the total Rabi-frequency Ω, which couples to the $|a\rangle - |+\rangle$ transition, obeys the equation [7]

$$\left(\frac{\partial}{\partial t} + c\frac{\partial}{\partial z}\right)\Omega(z,t) = -g^2 N\,\mathrm{Im}[\rho_{a+}] \tag{2}$$

where g is the effective coupling strength proportional to the dipole moment and N is the number density of atoms. When the mixing angle $\theta(t)$ is time-dependent, there is an additional coupling between the dark and bright states not shown in Fig. 1b. The effective Rabi-frequency of this (non-adiabatic) coupling is $2\dot\theta(t)$. The energy loss of the pulses due to absorption is characterised by the evolution of $\Omega(z,t)$. Reshaping of the pulses due to mutual photon exchange via Raman scattering is described by the evolution of $\dot\theta(z,t)$. We are here interested only in the energy loss and will therefore not consider the propagation of $\dot\theta(z,t)$. Since furthermore for strong fields and in the adiabatic limit $\dot\theta$ is much smaller than Ω we can ignore the non-adiabatic coupling in the present discussion altogether.

The coherence decay γ_0 in the bare-state basis corresponds to a population exchange between the dark and bright state with rate $\overline{\gamma}_0(t) = \gamma_0\, 2\sin^2\theta(t)\cos^2\theta(t)$. (In an asymmetric case, $\Omega_1 \neq \Omega_2$, it also leads to some cross-coupling rates, which are however of no importance here and are neglected.)

We now discuss the effect of radiation trapping in an optically thick sample of three-level atoms. As shown in [8], the reabsorption of spontaneous photons in an in-homogeneously broadened system leads to additional nonlinear relaxation and pump terms in the single-atom Bloch equations. For the case of strong fields, such that the total Rabi-frequency exceeds the one-photon Doppler-width (which is a necessary condition for EIT), there are essentially two independent spontaneous emission channels corresponding to the Autler-Townes transitions in a fully diagonalized basis. The corresponding rates can be calculated along the lines of Refs.[6, 8]. In the dark–bright-state basis they correspond to an incoherent population exchange of states $|a\rangle$ and $|-\rangle$ and $|-\rangle$ and $|+\rangle$ with the rate

$$\Gamma = \frac{\gamma}{2}\frac{1}{\sqrt{\pi}}\int_{-\infty}^{\infty}\mathrm{d}y\,\mathrm{e}^{-y^2}\Big[1 - \exp\big\{-K\,\mathrm{e}^{-y^2}\big\}\Big]\frac{1-\rho_{--}}{3\rho_{--}-1} \tag{3}$$

where ρ_{--} is the dark-state population and

$$K = N\lambda^2 d\frac{\gamma}{\sqrt{8\pi}\Delta_D}\frac{1}{2}\big(3\rho_{--} - 1\big) = \frac{K_0}{2}\big(3\rho_{--} - 1\big). \tag{4}$$

$\lambda = \lambda_1 \approx \lambda_2$ is the laser wavelength. d is the smallest effective escape distance of the incoherent photons. If the laser beams are for example cylindrical with a homogeneous transversal intensity profile, d is the radius of the cylinder.

Taking into account the coherent coupling of the total Rabi-frequency and the collective rates, averaging over the Doppler-distribution and adiabatically eliminating the coherences, we find the Bloch equations

$$\dot{\rho}_{aa} = -\big(2\gamma+\Gamma\big)\rho_{aa} + \Gamma\rho_{--} - R\big(\rho_{aa}-\rho_{++}\big), \quad (5)$$

$$\dot{\rho}_{++} = -\big(\overline{\gamma}_0+\Gamma\big)\rho_{++} + \big(\overline{\gamma}_0+\Gamma\big)\rho_{--} + \gamma\rho_{aa} + R\big(\rho_{aa}-\rho_{++}\big). \quad (6)$$

Furthermore $\rho_{a+} = -i\Omega/\Delta_D\sqrt{\pi/8}\,\big(\rho_{aa}-\rho_{++}\big)$. Here $R(z,t) = \Omega^2(z,t)/(\Delta_D\sqrt{8/\pi})$ is the effective optical pump rate. Equations (5) and (6) are highly nonlinear since Γ is a functional of ρ_{--}. We have not taken into account Lorentz-Lorenz local field corrections [9] in these equations. Radiation trapping of spontaneous photons becomes important when the smallest escape distance is of the order of the absorption length, i.e. if $N\lambda^2 d \sim 1$. Lorentz-Lorenz local field corrections, on the other hand, require densities of at least one atom per cubic wavelength, i.e. $N\lambda^3 \sim 1$. Thus radiation trapping is usually relevant for much smaller densities then necessary for local field effects.

3. EIT with cw-fields

We first discuss the stationary properties of EIT. In this case the only mechanism for creating a trapping state is Raman optical pumping. As discussed in [6] the trapping of spontaneous photons leads to a substantial decrease of the pumping rate. This reduction can be so severe that a very small population exchange rate between dark and bright states is sufficient to balance out optical pumping. As a result the maximum achievable stationary population of the dark state can be well below unity. This is illustrated in Fig. 2, where we have plotted the stationary population of the dark state for different values of γ_0 as a function of the density parameter $K_0 \equiv N\lambda^2 d\gamma/(\Delta_D\sqrt{8\pi})$. One recognises that beyond a certain critical density, which depends on the de-phasing rate γ_0, a considerable amount of population remains in other states.

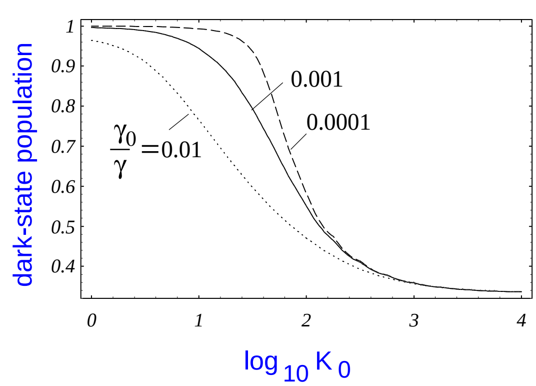

Fig.2: Stationary population of the dark state, ρ_{--}, as function of optical depth K_0 for different values of the de-phasing rate γ_0. The two fields have equal Rabi-frequencies and $R = 100$.

Introducing the normalised propagation distance $\xi = z/z_0$, where z_0 corresponds to the absorption length for a single laser field, $z_0 \equiv \left[\frac{3}{4}N\lambda^2\gamma/(\sqrt{2\pi}\Delta_D)\right]^{-1}$, we find the propagation equation

$$\frac{\mathrm{d}}{\mathrm{d}\xi}R(\xi) = \big(\overline{\rho}_{aa} - \overline{\rho}_{++}\big)\; R(\xi). \quad (7)$$

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$\overline{\rho}_{aa}$ and $\overline{\rho}_{++}$ are the stationary populations in the excited and bright state respectively.

In Fig. 3 we have plotted the normalised effective pump rate R, which is proportional to the average intensity of the fields as function of the normalised propagation distance. One recognises a linear decrease of the intensity in the initial phase.

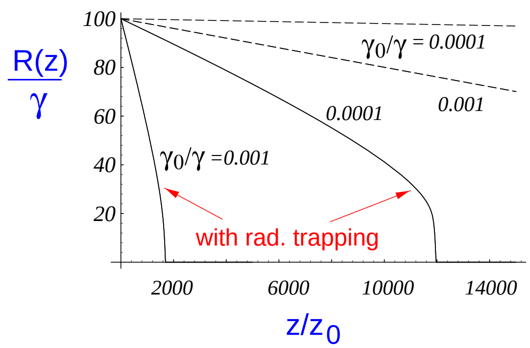

Fig.3: EIT for cw-fields. Plotted is the stationary pump rate, $R(z)$, which is a measure for the average intensity of the fields as function of the propagation distance for different values of $\overline{\gamma}_0 = \gamma_0$. $K_0 = 10$ and $\Omega_1 = \Omega_2$. The dashed lines correspond to the case when radiation trapping is ignored.

This non-exponential behaviour is typical for EIT [10], since for strong fields the absorption coefficient is inversely proportional to the intensity. Although transparency can be maintained over several thousands one-photon absorption length, it is substantially reduced as compared to the case without radiation trapping.

4. Pulsed EIT and stimulated Raman adiabatic passage

As can be seen from Fig. 3, a mechanism that avoids the generation of spontaneous photons in the creation of the dark state could lead to a substantial increase of transparency. This is clearly possible only in the non-stationary regime. Since the characteristic times for reaching the stationary state are rather long (on the order of the decay time of the dark state), techniques of adiabatic transfer can be applied. When the fields Ω_1 and Ω_2 are applied in counterintuitive order, i.e. if Ω_1 is switched on first, the dark state coincides initially with the bare atomic state $|b\rangle$. Switching on the second field at a rate small compared to the total Rabi-frequency adiabatically rotates the dark state into the coherent superposition of $|b\rangle$ and $|c\rangle$ without significantly populating the excited state, i.e. without spontaneous emission. To ensure adiabaticity, the time T of creating the dark state has to be large compared to the Rabi-oscillation period, $T \gg \Omega^{-1}$ and thus can still be much smaller than the time for reaching the stationary state.

This is illustrated in Fig. 4. Here we have shown the absorption as function of time if both pulses are applied at the same time (upper dashed curve) and for a counterintuitive application, i.e. for Ω_2 switched on after Ω_1 (lower dashed and dashed-dotted curve).

For a counterintuitive application of the pulses, the dark state corresponds initially to the bare state $|b\rangle$. Thus for small times all population is hidden from the interaction and the absorption is close to zero. When the second pulse (Ω_2) is turned on, the dark state rotates from the bare state $|b\rangle$ to the superposition of $|b\rangle$ and $|c\rangle$ according to Eq.(1) and all population remains in this state (adiabatic population transfer). When both fields are on, the de-phasing (γ_0) of the $|b\rangle - |c\rangle$ coherence leads however to a population exchange ($\overline{\gamma}_0$) between bright and dark state and an increase of absorption.

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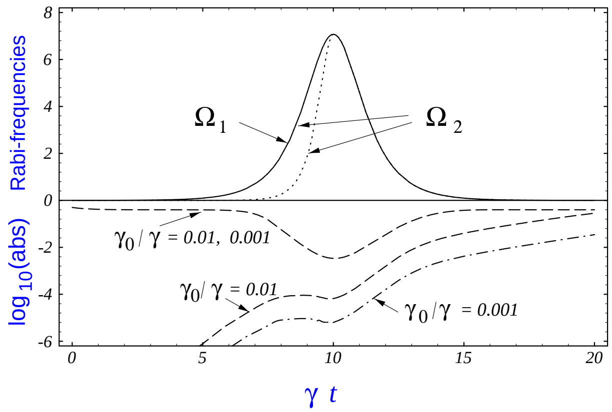

Fig.4: Absorption in arbitrary units and log-scale as function of time for simultaneous pulses ($\Omega_1 = \Omega_2$) (upper dashed curve) and for counterintuitive pulses (lower dashed and dashed-dotted curve). The upper half of the box shows corresponding Rabi-frequencies. Here $R_{\max}/\gamma = 100$ and $K_0 = 10$.

This is in contrast to the case of identical pulses, where initially a substantial amount of the population (50 % in the above example) is in the bright state. Thus there is a rather large absorption at the front end of the pulses. When the strength of the fields increases this population is optically pumped into the bright state and the absorption decreases. Due to radiation trapping this pump process is however not efficient and the absorption remains two orders of magnitude larger as compared to the case of counterintuitive pulses.

It is interesting to note that initially matched pulses ($\Omega_2(t)/\Omega_1(t) = \text{const.}$) evolve into a counterintuitive pair of pulses in the curse of propagation due to Raman scattering [3]. In the initial phase of propagation a positive value of $\dot{\theta}$ is built up and a counterintuitive pulse sequence established.

5. Summary

In the present paper we have discussed electromagnetically induced transparency and the generation of coherent superposition states (dark states) in optically thick three-level media. It was shown that reabsorption and multiple scattering of spontaneously emitted photons leads to an substantial enhancement of absorption and thus disfavours Raman optical pumping as mechanism of dark state creation. This sets rather strong bounds to the ability of achieving stationary EIT or to prepare coherent superposition states in the stationary regime. On the other hand, the non-stationary coherent population transfer with a counterintuitive sequence of pulses (STIRAP) avoids radiation trapping and thus leads to a significantly enhanced transparency. Since the time of establishing the stationary state is rather long (dark states can have lifetimes on the order of microseconds) pulse length in the nanosecond or microsecond regime can be used.

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