Raman adiabatic transfer of optical states in multilevel atoms

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We analyze electromagnetically induced transparency and light storage in an ensemble of atoms with multiple excited levels (multi-Λ configuration) which are coupled to one of the ground states by quantized signal fields and to the other one via classical control fields. We present a basis transformation of atomic and optical states which reduces the analysis of the system to that of electromagnetically induced transparency in a regular three-level configuration. We demonstrate the existence of dark state polaritons and propose a protocol to transfer quantum information from one optical mode to another by an adiabatic control of the control fields.

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I. INTRODUCTION

Electromagnetically induced transparency (EIT) is a quantum interference effect occurring when a weak signal light field and a stronger control field both interact with an ensemble of atoms with Λ-shaped energy level configuration [1,2]. The quantum probabilities for an excitation of the atoms by both light fields interfere destructively, so that no excitation takes place and the normally highly opaque medium becomes transparent for the signal field.

EIT in atomic media attracts great interest due to its possible applications in nonlinear optics and quantum information processing. In particular, high sensitivity to the two-photon resonance condition leads to a steep dispersion for the signal field which therefore experiences a greatly reduced group velocity. The demonstration of such an effect in an ultracold atomic gas [3] and hot atomic vapor [4] and the subsequent stopping of light [5,6] by an adiabatic process make this system appealing as a candidate for a quantum optical memory device.

Of further interest are double- and multi-Λ configurations that contain two or more excited levels and are excited by several control fields. Nonlinear effects such as four-wave mixing [7,8], phase conjugation [9], and amplification without inversion [10,11] have been investigated for strong fields applied to both sides of the Λ [12]. If, on the other hand, the control fields couple to the same ground state, and the signal fields to the other (Fig. 1), the behavior of the system with respect to the signal fields is analogous to regular EIT, but with given control fields EIT is experienced by only one specific linear combination of signal modes [12–16] whereas others get absorbed. The action of the atomic sample on the signal fields is analogous to that of an interferometer followed by absorption of all but one output modes. Raczynski and Zaremba [17,18] investigated formation of dark-state polaritons [19] as well as storage of light in a double-Λ system.

Most of the existing work on EIT in multilevel systems was done with classical fields. An expansion into the quantum domain was undertaken by Liu et al. who derived an expression for a dark state with multiple quantum signal fields in stationary modes [16,20]. However, to our knowledge, no full quantum EIT and/or light storage formalism has been developed for propagating optical fields in this system. In the present paper, we bridge this gap by elaborating a basis transformation for both atomic and optical states which reduces multilevel EIT to the well investigated EIT in a regular Λ scheme. In addition, we show that by an adiabatic change of the control fields, a transfer of quantum optical states between different signal modes or their linear combinations can be implemented. This procedure resembles stimulated Raman adiabatic passage (STIRAP) [21], but applies to optical rather than atomic states and can be useful for routing and distribution of optically encoded information in classical and quantum communication.

II. DISCRETE FIELD MODES

In order to better understand EIT in a multi-Λ ensemble, we first focus on a simplified system with discrete (non-propagating, such as in a cavity) quantized field modes before we generalize our treatment to propagating wave packets.

We consider an ensemble of $N$ multi-Λ-configuration atoms (Fig. 1). Each of the excited states $\{ |A_q\rangle \}_{q=1,\ldots,Q}$ is coupled to the two ground states $|B\rangle$ and $|C\rangle$ by a quantized

![Figure 1](http://www.iqis.org/)

FIG. 1. Multi Λ-system; $Q$ excited states $|A_q\rangle$ are each coupled by a classical control field $\Omega_q$ with detuning $\Delta$ to the ground state $|C\rangle$ and by a quantized field $\hat{a}_q$ with detuning $\delta$ to another ground state $|B\rangle$. 

*http://www.iqis.org*/
signal field $a_q$ and a classical control field $\Omega_q$, respectively. All the signal beams are detuned from the optical resonance by the same amount $\delta = E_q - E_R - h \nu_q$; the detuning of the control beams is $\Delta = E_q - E_C - h \omega_q$, where $\nu_q$ and $\omega_q$ are the respective laser frequencies. The detunings $\Delta$ and $\delta$ are assumed to be small compared to the energy difference of the excited states so that each field $\Omega_q$, $a_q$ essentially only couples to its corresponding level $|A_q\rangle$.

Let $\hat{\sigma}_{ij} = \sigma_{ij} / \sqrt{|\sigma_{ij}|}$ be the flip operator between the states $|B\rangle$ and $|\alpha\rangle$ of the $j$th atom. When all fields are resonant ($\delta = \Delta = 0$) the dynamics of this system is described by the interaction Hamiltonian

$$\hat{H}_\text{int} = -\hbar \sum_{j=1}^{N} \sum_{q=1}^{Q} \left[ g_j \hat{a}_q \hat{\sigma}_{aj} + \Omega_q(t) \hat{\sigma}_{aj} \right] + \text{H.a.} \quad (1)$$

in the corotating frame. Here $\hat{a}_q$ is the photon annihilation operator of the $q$th mode and $g_q$ describes the vacuum Rabi frequency of that transition which is assumed to be equal for all the atoms (Dicke limit). $\Omega_q(t)$ is the slowly varying Rabi frequency of the according classical control field.

Let $|C^k\rangle$ denote the totally symmetric state with $k$ atoms in state $|C\rangle$ and all others in state $|B\rangle$

$$|C^k\rangle = \frac{1}{\sqrt{N_N^k}} \sum_{i,j_1<\cdots<j_k<N} |B_{i1}, \ldots, C_{j1}, \ldots, C_{j_k}, \ldots, B_N\rangle. \quad (2)$$

By analogy to Ref. [20], it then can be shown that the states

$$|D,n\rangle = \left[ \sum_{j=1}^{N} \hat{\sigma}_{CB} - \sum_{q=1}^{Q} \left( \frac{\Omega_q(t)}{g_q} \hat{a}_q \right) \right]^n |C^0, (0, \ldots, 0)\rangle \quad (3)$$

are dark states: they are eigenstates of the interaction Hamiltonian with zero eigenvalue. Here the $(n_1, \ldots, n_q)$ part denotes the state of the quantized light field in Fock representation.

### Adiabatic transfer of optical states

If one of the control fields is strong ($\Omega_i \gg g_i / \sqrt{N}$) while others vanish, the dark state takes the form

$$|D,n\rangle \xrightarrow{\Omega_i \rightarrow \Delta \rightarrow 0} |C^0, (0, \ldots, n, \ldots, 0)\rangle; \quad (4)$$

all photons gather in the according signal field mode. If all controls are slowly switched off the dark state adiabatically changes to

$$|D,n\rangle \xrightarrow{\Omega_1 \rightarrow \cdots \Omega_Q \rightarrow 0} |C^0, (0, \ldots, 0)\rangle, \quad (5)$$

so the quantum optical state carried by the $i$th mode is converted to a coherent collective ground state superposition [19].

Suppose now that while the system is in the state (4), another control field $\Omega_j$ is turned on. In this case, by adiabatic following, the state of the system will convert to

$$|D, n\rangle \xrightarrow{\frac{\Omega_{k+\Delta}}{g_k} + \frac{\Omega_{j-\Delta}}{g_j}} \left( \frac{\Omega_j}{g_j} \hat{a}_j + \frac{\Omega_k}{g_k} \hat{a}_k \right)^n |C^0, (0, \ldots, 0)\rangle. \quad (6)$$

If $\Omega_i$ is then slowly turned off, the quantum state of the $i$th optical mode will be transferred to the $j$th mode completely

$$|D,n\rangle \xrightarrow{\Omega_i \rightarrow 0} |C^0, (0, \ldots, n, \ldots, 0)\rangle. \quad (7)$$

We see that, by varying the control fields, the quantum state can be transferred to any other optical mode or their coherent superposition. We call this procedure Raman adiabatic transfer of optical states (RATOS) by analogy to the well known STIRAP technique which permits transfer of population between atomic states by means of adiabatic interaction with light [21]. In RATOS, on the other hand, quantum states are transferred between optical states by adiabatic interaction with atoms.

The above treatment is valid for the case of discrete, nonpropagating modes, e.g., in a cavity. In the practical case of a propagating field, photons first travel through an atom-free environment, then couple into an EIT medium, experience RATOS while in transfer, and finally leave the medium. In order to understand the propagation dynamics, the theory must be reformulated in terms of dark-state polaritons akin to Ref. [19]. This is our task for the remainder of the paper.

One specific question that needs to be addressed is whether RATOS can be applied to optical fields that are initially (prior to coupling into an EIT system) not in a dark state in the sense of Eq. (3). An example is both modes $i$ and $j$ containing one photon while the remaining modes are in the vacuum state. Can one choose control fields in such a way that these photons are losslessly coupled into an EIT medium, and if not, what minimum loss can one expect? More generally, what are the possibilities of quantum optical state engineering in a multi-level EIT environment?

### III. MAPPING TO A SINGLE-Λ SYSTEM

Our approach is to develop a basis transformation of the atomic and optical states that will map a multi-Λ-system to a normal EIT (single-Λ) scheme, thus providing an intuitive understanding for the optical properties of the system.

#### A. Changing the atomic basis

Consider one atom with a multi-Λ level structure as in Fig. 1. In the rotating wave frame the Hamiltonian reads

$$\hat{H}(t) = -\frac{\Delta}{2} |B\rangle\langle C| + \Omega_q(t) |A_q\rangle\langle C| + \text{H.a.}, \quad (8)$$

which in the absence of the quantized fields reduces to

$$\hat{H}_0 = -\frac{\Delta}{2} |B\rangle\langle B| - \Delta |C\rangle\langle C| + \Omega |E\rangle\langle B| + \text{H.a.,} \quad (9)$$

where
\[ |EB\rangle = \sum_{q=1}^{Q} \frac{\Omega_q}{\Omega} |A_q\rangle \]  

(10)

and

\[ \Omega = \sqrt{\sum_{q=1}^{Q} |\Omega_q|^2}. \]  

(11)

\(\hat{H}_0\) possesses \(Q\) eigenstates of zero eigenvalue, one of them obviously being \(|B\rangle\). The others are superpositions of excited states \(|A_q\rangle\) that are orthogonal to the “excited bright state” \(|EB\rangle\) and thus not coupled by the control fields.

A basis spanning this subspace can be explicitly constructed by an unitary Householder reflection [22]

\[ \hat{U} = \sigma |u\rangle\langle u| - 1 \quad \text{with} \quad \sigma = \langle A_0 | EB \rangle + 1 = \frac{\Omega_0}{\Omega} + 1 \]

and \(|u\rangle = \frac{1}{\sigma} (|A_0\rangle + |EB\rangle)\)

(12)

so that \(|EB\rangle = \hat{U}|A_0\rangle\) and

\[ |ED_q\rangle = \hat{U}|A_q\rangle = \frac{\Omega_0^*}{\Omega_0^* + \Omega} (|A_0\rangle + |EB\rangle) - |A_q\rangle \]

for \(q = 1, \ldots, Q - 1\).

In this basis the interaction Hamiltonian then reads

\[ \hat{H} = -\hbar \frac{\Lambda}{2} |C\rangle\langle C| - \hbar \delta |B\rangle\langle B| - \hbar \Omega |EB\rangle\langle C| \]

\[ -\hbar \sum_{q=1}^{Q} \sum_{r=1}^{Q-1} g_{qr} |ED_q\rangle\langle A_r| \hat{a}_q |ED_r\rangle\langle B| \]

\[ -\hbar \sum_{q=1}^{Q} g_{q0} |EB\rangle\langle A_q| \hat{a}_q |EB\rangle\langle B| + \text{H.a.} \]

(13)

As expected, the states \(|ED_q\rangle\) do not undergo any interaction with the classical fields \(\Omega_q\) at all.

This can be interpreted physically by understanding that the phases and amplitudes of the excited states \(|A_q\rangle\) are such that the probability amplitudes for a transition from the states \(|ED_q\rangle\) to \(|C\rangle\) interfere destructively, akin to dark states in a normal EIT scheme, hence, we call the \(|ED_q\rangle\) “excited dark states.”

Also in close analogy to EIT, the ground state \(|C\rangle\) is coupled to only one particular superposition \(|EB\rangle\) of the excited states (the excited bright state), where the transition probabilities interfere constructively.

However, each of the weak quantized optical modes \(\hat{a}_q\) couples the ground state \(|B\rangle\) to all of the states \(|EB\rangle, |ED_1\rangle, \ldots, |ED_{Q-1}\rangle\) (see Fig. 2). If the excited states \(|A_q\rangle\) have a short lifetime, so do the states \(|ED_q\rangle\). Hence, in general, light in the modes \(\hat{a}_q\) would not experience EIT; the photons would get absorbed, exciting the atom to the \(|ED_q\rangle\) levels which decay due to spontaneous emission. In Sec. III B we show, however, that there exists a linear superposition of signal states which does not couple to \(|ED_q\rangle\)’s, thus enabling EIT in this system.

As is expected, the Hamiltonian is

\[ \hat{H} = -\hbar \left( \frac{\Delta}{2} |C\rangle\langle C| + \Omega |EB\rangle\langle C| \right) - \hbar \left( \frac{\delta}{2} |B\rangle\langle B| + g \hat{b}_q |EB\rangle\langle B| \right) \]

\[ -\hbar \sum_{q=1}^{Q-1} \hat{b}_q \left( g_{q0} |EB\rangle\langle B| + \sum_{r=1}^{Q-1} g_{r0} |ED_r\rangle\langle B| \right) + \text{H.a.} \]

(18)

The first two terms correspond exactly to the Hamiltonian of a traditional \(\Lambda\) system (\(|B\rangle \leftrightarrow |EB\rangle \leftrightarrow |C\rangle\)). The quantized field mode

\[ |ED_{Q-1}\rangle \]

\[ |ED_1\rangle \]

\[ \hat{a}_q \]

\(q = 1, \ldots, Q\)

\[ |EB\rangle \]

\[ \delta \]

\[ \Omega \]

\[ |C\rangle \]

\[ |B\rangle \]

\[ \Delta \]

\[ |ED_0\rangle \]
be decomposed into components each interacting only with their respective transition
\[ \hat{E}(z,t) = \sum_{q=1}^{Q} \hat{E}_q(z,t), \quad \hat{E}_q \text{ coupling } |B\rangle \leftrightarrow |A_q\rangle. \] (22)

We now define the slowly varying field operators \( \tilde{a}_q(z,t) \) by the positive frequency parts of our field components
\[ \hat{E}_q^+(z,t) = \tilde{a}_q(z,t) \frac{\hbar \nu}{2\omega_0 \nu} \exp \left[ \frac{\nu}{c} (z-ct) \right]. \] (23)

To describe the evolution of the atomic variables, we can assume that the quantum amplitude of the atomic variables does not depend strongly on the position. By introducing a “smearing kernel” \( s \) with \( \int_0^L s(z) dz = L/N \) and a zero-centered support with a width that is large compared to the average distance of two atoms but small in relation to the medium length \( L \), we obtain the mean-field operators
\[ \bar{s}_{a,b}(z) = \sum_{j=1}^{N} s(z-z_j) \bar{s}_{a,b}^{(j)}. \] (24)
so that, assuming \( \Delta = \delta = 0 \),
\[ \dot{\hat{H}}(t) = -\frac{\hbar N}{L} \int_0^L dz \left[ g \bar{s}_{EB,B} \hat{b}_Q + \bar{s}_{EB,C} \right. \]
\[ + \sum_{q=1}^{Q-1} \hat{b}_q \left( g_{EB} \bar{s}_{EB,B} + \sum_{r=1}^{Q-1} g_{ED} \bar{s}_{ED,C} \right) + \text{H.a.} \] (25)

Performing mappings \( \hat{U} \) and \( \hat{W} \) on atoms and light, the Hamiltonian transforms as follows:
\[ \hat{H}_{\text{int}} = -\frac{\hbar N}{L} \int_0^L dz \left[ g \bar{s}_{EB,B} \hat{b}_Q + \bar{s}_{EB,C} \right. \]
\[ + \sum_{q=1}^{Q-1} \hat{b}_q \left( g_{EB} \bar{s}_{EB,B} + \sum_{r=1}^{Q-1} g_{ED} \bar{s}_{ED,C} \right) + \text{H.a.} \]. (26)

The Maxwell-Bloch equations for the individual fields are
\[ \left( \frac{\partial}{\partial t} + c \frac{\partial}{\partial z} \right) \tilde{a}_q = iN g^* \bar{s}_{B,A_q}. \] (27)
Performing summation of Eqs. (27) over \( q \)'s with weights \( \Omega_{q}^f g^* \) and utilizing relations (10) and (19), we find
\[ \left( \frac{\partial}{\partial t} + c \frac{\partial}{\partial z} \right) \hat{b}_Q = iN g \bar{s}_{EB}. \] (28)

In other words, if there is no light in the modes \( \hat{b}_{q+Q} \) and no atoms are in the excited states, the propagation of mode \( \hat{b}_Q \) in a multilevel EIT setting is fully equivalent to that in a single-\( \Lambda \) system defined by Fig. 3.

Similarly to Ref. [19], one can define the dark-state polariton for this system. Upon entering the medium an incoming light pulse in the EIT mode forms a polariton \( \Psi \), a superposition of an electromagnetic wave in the \( \hat{b}_Q \) mode and a collective atomic excitation \( \bar{s}_{EB,C} \) which generates an eigen-
state of eigenvalue zero of the interaction Hamiltonian
\[ \Psi = \cos \theta(t) \vec{b}_Q - \sin \theta(t) \sqrt{N} \vec{a}_{R,C}, \]  
\[ \tan \theta(t) = \frac{\sqrt{N}}{R(t)}. \]

By changing the classical control fields’ parameter \( R \) the character of this polariton [whether it is more optical \((\theta = 0)\) or has a stronger atomic component \((\theta = \pi/2)\)] can be changed.

### B. Incoupling and slowdown

The susceptibility for the EIT mode \([23]\) is proportional to
\[ \chi_Q \approx N g^2 \frac{\Delta - \delta}{\sqrt{\delta^2 + \gamma^2}} + \Omega^2, \]

where \( \gamma \) is the spontaneous decay rate of the excited bright-state \(|EB\). So for a signal beam in precise two-photon resonance \((\Delta = \delta)\) the refractive index for the signal field is\(\frac{(\Delta - \delta)}{\delta + \frac{\gamma}{2}} + \Omega^2\) where \( \gamma \) is the spontaneous decay rate of the light of interest. For a signal beam in precise two-photon resonance \((\Delta = \delta)\) the refractive index for the signal field is
\[ \chi_Q \approx N g^2 \frac{\Delta - \delta}{\sqrt{\delta^2 + \gamma^2}} + \Omega^2, \]

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\[ \chi_Q \approx N g^2 \frac{\Delta - \delta}{\sqrt{\delta^2 + \gamma^2}} + \Omega^2. \]

If the effective Rabi frequency \( \Omega \) is small compared to \( \gamma \), the transparency window is narrow and Eq. (32) predicts a strong dispersion. This leads to a strongly reduced group velocity \( v_g \) for the polariton wave
\[ v_g = \frac{c}{1 + n_g}, \quad n_g = \frac{N}{R^2}. \]

### V. RATOS

**The procedure**

Based on this formalism we now describe a protocol for transfer of quantum information between optical modes (Raman adiabatic transfer of optical states, RATOS).

If the intensities of the control fields are changed slowly, the eigenstates follow the new conditions adiabatically \([28]\). The dark-state polariton as eigenstate of zero interaction energy is thus preserved; however, its mode composition and propagation velocity can be controlled by the parameters \( \{\Omega_q\} \) of the strong control fields according to Eq. (19).

This allows for transfer of quantum information from an optical mode \( \vec{a}_i \) to another mode \( \vec{a}_j \):

1. First only one strong control field \( \Omega_i \) is switched on. The medium then exhibits electromagnetically induced transparency for the \( \vec{b}_Q = \vec{a}_i \) mode.

2. An incoming quantum pulse in the \( \vec{a}_i \) mode can enter the EIT medium without absorption or reflection since at two-photon resonance the refractive index for the signal field is

3. One. The pulse experiences a reduction of the group velocity according to Eq. (33).

4. This slowdown also leads to a spatial compression: the pulse gets shorter in length, which helps in keeping the size of the medium reasonably small.

5. Once the pulse is completely inside the medium, the control field \( \Omega_i \) is replaced by another field \( \Omega_j \), adiabatically. Assuming the mixing angle \( \theta \) is kept constant, the polariton changes its characteristics as follows:

\[ \Psi_{\text{RATOS}} = \cos \theta \vec{a}_i - \sqrt{N} \sin \theta \vec{a}_{C,EB} \]

and all photons are now in the \( j \)th mode.

6. A pulse with a different frequency but in the same optical quantum state as the original pulse exits the medium in mode \( \vec{a}_j \).

RATOS might find applications as an optical switch to route optical quantum information. If in the end not one but several control fields are present, the incoming pulse is split into optical modes with different frequencies.

We now review a few recently published procedures for transferring optical information via atomic transitions that are related to the one developed above. Zibrov et al. \([24]\) used the double \( \Lambda \) system formed by the fine structure splitting of \( ^{87}\text{Rb} \). They first couple in a light pulse resonant to one of the fine transition lines and store it via an adiabatic turn off of the control field. Later on they retrieve it with a control field tuned to the other fine structure transition. Matsko et al. and Peng et al. \([25,26]\) investigate transferring a light pulse to another mode using storage in a single-\( \Lambda \) system. By inverting the roles of the control and signal modes in the retrieving process, the pulse is retrieved at the frequency of the original control field. The main difference of RATOS with respect to these proposals is that it offers a way to extend this transfer to multiple modes (and even to their coherent superpositions) and that an intermediate storing of the pulse is not necessary.

### VI. QUANTUM STATE ENGINEERING

Now we also can answer the question to which extent RATOS can be used for engineering of optical quantum states. The only mode that can losslessly enter a multilevel EIT sample is that associated with the operator \( \vec{b}_Q \) which is a linear combination of individual mode operators \( \{\vec{a}_q\} \). However, by choosing amplitudes and phases of the control fields one can adjust the coefficients of the linear combination.

The linear transformation \( \mathcal{W}(\{\Omega_1, \ldots, \Omega_Q\}) : \vec{a}_q \rightarrow \vec{b}_q \) of the fields at the cell entrance can be visualized as an interferometer, i.e., a sequence of linear optical elements such as beam splitters and mirrors (Fig. 4). While this transformation does not by itself represent any physical process, the modes \( \vec{b}_q \) do have a physical meaning as only one of them is able to propagate through the cell due to EIT; the rest get absorbed.

While the mode \( \vec{b}_Q \) is traversing the cell, the control fields may change adiabatically so at the cell output, when the propagating modes convert back to \( \vec{a}_q \)'s, the interferometer,
For this reason, only with 50% probability will both photons $a\bar{q}$ and $b\bar{q}$ be coupled into the EIT mode $b\bar{q}$, with equal probability they will experience absorption. So in this setup the double-$\Lambda$ medium does not perform better than an ordinary beam splitter: here the Hong-Ou-Mandel effect [27] also provides a 50% probability for the two photons to fuse into a specific mode. Furthermore, it is clear that no combination of control fields would make the atomic system fully transparent for the state (35), so this state cannot be coupled into the EIT medium without loss.

In summary, a multi-$\Lambda$ medium is equivalent to a linear optical system with a built-in storage device and with multiple input and output modes which differ in frequency (Fig. 4).

VII. CONCLUSIONS

We have extended a full quantum treatment of the electromagnetically-induced transparency to multi-$\Lambda$ systems. An explicit form of an unitary mapping is presented that relates the dark states to the effects observed in a standard EIT scheme. Most of the properties of this well investigated system can be transferred and extended to systems with multiple excited levels.

The mapping provides a physical explanation for the existence of the decay sensitive $|ED_i\rangle$ states and the according bright-state modes $b_{1,2}\rangle$.

EIT in a multi-$\Lambda$ scheme might be useful for multiplexing and routing of optical quantum information as well as for the preparation of multimode entangled quantum states. Its application to quantum-optical engineering is however limited by its equivalence to a linear-optical setup with a built-in storage capability.

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[28] Strictly speaking, time variation of $W_{\phi^k}$ leads to a geometric phase which we will explore in a subsequent publication.