Coherent preparation by laser light of the quantum states of atoms and molecules can lead to quantum interference in the amplitudes of optical transitions. In this way the optical properties of a medium can be dramatically modified leading to electromagnetically induced transparency and related effects. These phenomena have placed gas phase systems at the centre of recent advances in the development of media with radically new optical properties. We will discuss these advances, and the new possibilities that arise in non-linear optics and quantum information science, when the optical properties of a medium are modified by coherent preparation. We will develop the theory of electromagnetically induced transparency by considering the atomic dynamics and the optical response first for a CW laser. The pulse propagation and the adiabatic evolution of the field coupled states is then developed. From this we will see how coherently prepared media can be used to improve frequency conversion in non-linear optical mixing experiments. The extension of these concepts to very weak optical fields in the few photon limit will then be examined. We will conclude by discussing future prospects and potential new applications.
2003). It should be emphasized that the modification of atomic properties due to quantum interference has been studied extensively for 25 years, see e.g. Arimondo (1996). In particular the phenomenon of coherent population trapping (CPT) observed by Alzetta et al. (1976) is closely related to EIT. In contrast to CPT which is a "spectroscopic" phenomenon that involves only modifications to the material states in an optically thin sample, EIT is a phenomenon specific to optically thick media where both the optical fields and the material states are modified.

The optical properties of atomic and molecular gases are fundamentally tied to their intrinsic energy level structure. The linear response of an atom to resonant light is described by the first-order susceptibility $\chi^{(1)}$. The imaginary part of this susceptibility $\text{Im}[\chi^{(1)}]$ determines the dissipation of the field by the atomic gas (absorption), whilst the real part $\text{Re}[\chi^{(1)}]$ determines the refractive index. The form of $\text{Im}[\chi^{(1)}]$ at a dipole-allowed transition as a function of frequency is that of a Lorentzian function with a width set by the damping. The refractive index $\text{Re}[\chi^{(1)}]$ follows the familiar dispersion profile, with anomalous dispersion (decrease in $\text{Re}[\chi^{(1)}]$ with field frequency) in the central part of the absorption profile within the line-width. Figure 1 illustrates both the conventional form of $\chi^{(1)}$ and the modified form that results from EIT as will be discussed shortly.

![Graph of susceptibility as a function of frequency](image)

**FIG. 1** Susceptibility as a function of the frequency $\omega_p$ of the applied field relative to the atomic resonance frequency $\omega_{31}$, for a radiatively broadened two-level system with radiative width $\gamma_{31}$ (dashed lines) and an EIT system with resonant coupling field (full line). *top*: Imaginary part of $\chi^{(1)}$ characterizing absorption, *bottom*: Real part of $\chi^{(1)}$ determining the refractive properties of the medium.

In the case of laser excitation where the magnitude of the electric field can be very large we reach the situation where the interaction energy of the laser coupling divided by $\hbar$ exceeds the characteristic linewidth of the bare atom. In this case the evolution of the atom-field system requires a description in terms of state-amplitude or density-matrix equations. In such a description we must retain the phase information associated with the evolution of the atomic state amplitudes, and it is in this sense that we refer to atomic coherence and coherent preparation. This is of course in contrast to the rate equation treatment of the state populations often appropriate when the damping is large or the coupling is weak for which the coherence of the states can be ignored. For a full account of the coherent excitation of atoms the reader is recommended to consult Shore (1990).

For a 2-level system, the result of coherent evolution is characterized by oscillatory population transfer (Rabi flopping). The generalisation of this coherent situation to driven 3-level atoms leads to many new phenomena some of which, such as Autler-Townes splitting (Autler and Townes, 1955), dark-states and EIT, will be the subject of this review. These phenomena can be understood either within the basis of bare atomic states or new eigenstates which diagonalize the complete atom-field interaction Hamiltonian. In both cases we will see that interference between alternative excitation pathways between atomic states lead to modified optical response.

The linear and non-linear susceptibilities of a Λ-type 3-level system driven by a coherent coupling field will be derived in section III. Figures 1a and b show the imaginary and real part of the linear susceptibility for the case of a resonant coupling field as a function of the probe field detuning from resonance. Figure 2 shows the corresponding third-order non-linear susceptibility. Inspection of these frequency dependent dressed susceptibilities reveal immediately several important features. One recognizes that $\text{Im}[\chi^{(3)}]$ undergoes destructive interference in the region of resonance, i.e. the coherently driven medium is transparent to the probe field. The fact that transparency of the sample is attained at resonance is not in itself of great importance as the same degree of transparency can be obtained simply by tuning sufficiently away from resonance. What is important is that in the same spectral region as there is a high degree of transmission the non-linear response $\chi^{(3)}$ displays constructive interference, i.e. its value at resonance is larger than expected from a sum of two split Lorentzian lines. Furthermore the dispersion variation in the vicinity of the resonance differs markedly from the steep anomalous dispersion familiar at an undressed resonance (see Fig. 1). Instead there is a normal dispersion in a region of low absorption the steepness of which is controlled by the coupling-laser strength (i.e. very steep for low values of the drive laser coupling). Thus despite the transparency the transmitted laser pulse can still experience strong dispersive and non-linear effects. It is most significant that the refractive index passes through the vacuum value and the dispersion is steep and linear exactly where absorp-
**ELECTROMAGNETICALLY INDUCED TRANSPARENCY**

**A. Interference between excitation pathways**

To understand how quantum interference may modify the optical properties of an atomic system it is informative to follow the historical approach and turn first to the subject of photoionisation of multi-electron atoms. Due to the existence of doubly excited states the photoionisation spectrum of multi-electron atoms can show a rich structure of resonances. These resonances are broadened due to relatively rapid decay (caused by the interaction between the excited electrons of these states) to the degenerate continuum states, with lifetimes in the picosecond to sub-picosecond range. For the reason that they decay naturally to the continuum, these states are called auto-ionising states. Fano introduced to atomic physics the concept of the interference between the excitation channels to the continuum that exist for a single auto-ionising resonance coupled to a flat continuum (Fano, 1961) Figure 3(a). In the vicinity of the auto-ionising resonance the final continuum state can be reached via either direct excitation (channel 1) or through the resonance with the configuration interaction leading to the decay that provides a second channel to the final continuum state. The interference between these channels can be constructive or destructive and leads to frequency dependent suppression or enhancement in the photoionisation cross-section.

**FIG. 2** Absolute value of nonlinear susceptibility for sum-frequency generation $|\chi^{(3)}|$, as a function of $\omega_p$, in arbitrary units. The parameters are identical to those used in Fig. 1.

Coherent preparation techniques are most effective, and have been most studied, in atomic and molecular samples that are in the gas phase. The reason for this is that the coherent evolution of the states will in general be inhibited by dephasing of the complex state amplitudes, but the coherence dephasing rates for gases are relatively small when compared to those in solid state media. Nevertheless several workers have recently made progress in applying these techniques to a variety of solid-state systems (Faist et al., 1997; Ham et al., 1997a; Schmidt et al., 1997; Serapaglia et al., 2000; Wei and Manson, 1999; Zhao et al., 1997). Atomic gases remain very important in optical applications for several key reasons; for example they are often transparent in the vacuum UV and infra-red, and there exist techniques to cool them to ultra-cold temperatures thus eliminating inhomogeneous broadening. They can also tolerate high optical intensities. Gas phase media are for instance necessary for frequency conversion beyond the high and low frequency cut-off of solid state materials.

The dramatic modifications of the optical properties that are gained through coherent preparation of the atoms or molecules in a medium have ensured a renaissance in activity in the area of gas phase non-linear optics. New opportunities arise also due to the extremely spectrally narrow features that can result from coherent preparation; in particular this gives ultra-low group velocities and ultra-large non-linearities. These are of potential importance to new techniques in optical information storage and also of non-linear optics at the few photon level both of which may be important to quantum information processing.

Throughout this review we will try to point out the important applications that arise as a result of EIT and describe in outline some of the seminal experiments.

We attempt to cover comprehensively the alteration of linear and non-linear optical response due to electromagnetically induced transparency and related phenomena. We do not, however, cover the related topic of lasing without inversion (Gornyi et al., 1989; Harris, 1989; Kocharovskyakaya and Khanin, 1986; Scully et al., 1989) and the interested reader is advised to look elsewhere for reviews on that subject (Knight, 1990; Kocharovskyakaya, 1992; Mandel, 1993; Scully and Zubairy, 1997). In the following section we will introduce the underlying physical concepts of EIT through the coupling of near resonant laser fields with the states of a 3-level system. In section III we discuss in detail the dynamics in 3-level atoms coupled to the applied laser fields and determine the optical response. In section IV we treat the topic of pulse propagation in EIT and review the developments that have culminated in the demonstration of ultra-slow group velocities down to a few metres per second and even of “stopping” and “storing” of a pulse of light. The utility of coherent preparation in enhancing the efficiency of frequency conversion processes is discussed in section V. Then in section VI we will turn to the treatment of EIT in the few photon limit where it is necessary to apply a fully quantum treatment of the fields. Finally we draw conclusions and discuss the prospects in quantum optics and atom optics that may arise as a result of EIT.

**TABLE 1**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\omega$</td>
<td>1.5</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>0.3</td>
</tr>
</tbody>
</table>

**FIG. 1** Example of the suppressed transmission in EIT. The parameters are identical to those used in Table 1.
Experiments by Madden and Codling (1965) showed the resulting transparency windows in the auto-ionising spectrum of helium. Sometime later the auto-ionising interference structures in strontium were used by Armstrong and Wynne to enhance sum-frequency mixing in a frequency up-conversion experiment to generate light in the vacuum UV (Armstrong and Wynne, 1974). In this wave-mixing experiment the absorption was eliminated in spectral regions where the non-linear response remained large, hence an improved efficiency for frequency conversion was reported.

During the 1960’s auto-ionising spectra were much studied (see for example (Garton, 1966)). Several authors addressed the issue of interaction between two or more spectral series in the same frequency range where the interference between closely spaced resonances needs to be considered (Fano and Cooper, 1965; Shore, 1967). Shapiro provides the first explicit analysis of the case of interference between two or more resonances coupled to a single continuum (Shapiro, 1970). In this case the interference is mainly between the two transition pathways from the ground state to the final state via each of the two resonances (Figure 3(b)). Naturally interference will be significant only if the spacing between these resonances is comparable to or less than their widths.

Hahn, King and Harris (Hahn et al., 1990) showed how this situation could be used to enhance four-wave mixing. In experiments in zinc vapour that showed significantly increased non-linear mixing one of the fields was tuned to a transition between an excited bound state and a pair of closely spaced auto-ionising states, that had a frequency separation much less than their decay widths.

The case of interference between two closely spaced lifetime broadened resonances, decaying to the same continuum, was further analyzed by Harris (1989). He pointed out that this will lead to lasing without inversion since the interference between the two decay channels eliminates absorption whilst leaving stimulated emission from the states unchanged. Although we will not discuss the subject of lasing-without inversion further in this review, this work was an important step in the story with which we are concerned. A breakthrough was then made in the work of Imamoglu and Harris (1989) where it was realised that the pair of closely spaced lifetime broadened resonances were equivalent to dressed states created by coupling a pair of well separated atomic bound levels with a resonant laser field (Figure 4). They thus proposed that the energy level structure required for quantum interference could be engineered by use of an external laser field. Harris et al. (1990) then showed how this same situation can be extended to frequency-conversion in a four-wave mixing scheme among atomic bound-states with the frequency conversion hugely enhanced. This is achieved through the cancellation of linear-susceptibility at resonance as shown in Figure 1, whilst the non-linear susceptibility is enhanced through constructive interference. The latter paper was the first appearance of the term electromagnetically induced transparency (EIT) which was used to describe this cancellation of the linear response by destructive interference in a laser dressed medium.

Boller et al. (1991), in discussing the first experimental observation of EIT in Sr vapour point out that there are two physically informative ways that we can view EIT. In the first we use the picture that arises from the work of Imamoglu and Harris where the dressed states can be viewed as simply comprising two closely spaced resonances effectively decaying to the same continuum (Boller et al., 1991; Zhang et al., 1995). If the probe field is tuned exactly to the zero field resonance frequency then the contributions to the linear susceptibility due to the two resonances, which are equally spaced but with opposite signs of detuning, will be equal and opposite and thus lead to the cancellation of the response at this frequency due to a Fano-like interference of the decay channels. An alternative and equivalent picture is to consider the bare rather than the dressed atomic states. In this view EIT can be seen as arising through different pathways between the bare states. The effect of the fields is to transfer a small, but finite amplitude, into state |2⟩. The amplitude for |3⟩ which is assumed to be the only decaying state and thus the only way to absorption is thus driven by two routes; directly via the |1⟩ – |3⟩ pathway, or indirectly via |1⟩ – |3⟩ – |2⟩ – |3⟩ pathway (or

\[ |3⟩ \rightarrow |2⟩ \rightarrow |3⟩ \]
by higher order variants). Because the coupling field is much more intense than the probe, this indirect pathway has a probability amplitude that is in fact of equal magnitude to the direct pathway, but for resonant fields it is of opposite sign.

B. Dark state of the 3-level \( \Lambda \)-type atom

The use in laser spectroscopy of externally applied electromagnetic fields to change the system Hamiltonian of course pre-dates the idea of using this in non-linear optics or in lasing without inversion. We must mention the enormous body of work treating the effects of static magnetic fields (Zeeman effect) and static electric fields (Stark effect). The case of strong optical fields applied to an atom began to be extensively studied following the invention of the laser in the early 1960’s. Hänisch and Toschek (1970) recognized the existence of these type of interference processes for three-level atoms coupled to strong laser fields in computing the system susceptibility from a density matrix treatment of the response. They identified terms in the off-diagonal density matrix elements indicative of the interference, although they did not explicitly consider the optical and non-linear optical effects in a dense medium.

Our interest is in the case of electromagnetic fields, in the optical frequency range, applied in resonance to the states of a 3-level atom. We illustrate the three possible coupling schemes in Figures 5 and 6. For consistency schemes we will be most concerned with the Lambda configuration Fig. 5, since the Ladder and Vee configurations illustrated in Fig. 6 are of more limited utility for the applications that will be discussed later.

![FIG. 5 Generic system for EIT: Lambda-type scheme with probe field of frequency \( \omega_p \) and coupling field of frequency \( \omega_c \). \( \Delta_1 = \omega_31 - \omega_p \) and \( \Delta_2 = \omega_32 - \omega_p \) denote field detunings from atomic resonances and \( \Gamma_{31}, \Gamma_{32} \) radiative decay rates from state |1\> to state |k\>.

The physics underlaying the cancellation of absorption in EIT is identical to that involved in the phenomena of dark-state and coherent population trapping (Lounis and Cohen-Tannoudji, 1992). We will therefore review briefly the concept of dark-states. Alzetta et al. (1976) made the earliest observation of the phenomenon of coherent population trapping (CPT) shortly followed by Whitley and Stroud (1976). Gray et al. (1978) explained these observations using the notion of coherent population trapping in a dark eigen-state of a three level Lambda medium (see Figure 5). In this process a pair of near resonant fields are coupled to the Lambda system and result in the Hamiltonian \( H = H_0 + H_{int} \), where the Hamiltonian for the bare atom is \( H_0 \) and that for the interaction with the fields is \( H_{int} \). The Hamiltonian \( H \) has a new set of eigenstates when viewed in a proper rotating frame (see below) one of which has the form \( |a^0\> = \alpha|1\> - \beta|2\> \) which contains no amplitude of the bare state |3\> and is therefore effectively decoupled from the light fields. In the experiments of Alzetta population was pumped into this state via spontaneous decay from the excited states and then remains there since the excitation probability of this dark-state is cancelled via interference. A very informative review of the applications of dark-states and the coherent population trapping that accompanies them in spectroscopy has been provided by Arimondo (1996).

We would now like to look a bit more closely at the structure of the laser dressed eigen-states of a 3-level atom illustrated in Fig. 5. This discussion is intended to provide a simple physical picture that establishes the connection between the key ideas of EIT and that of maximal coherence.

Within the dipole approximation the atom-laser interaction \( H_{int} = \mu \cdot E \) is often expressed in terms of the Rabi coupling (or Rabi frequency) \( \Omega = \mu \cdot E_0/h \), with \( E_0 \) being the amplitude of the electric field \( E \), and \( \mu \) the transition electronic dipole moment. After introducing the rotating-wave approximation, we can represent the Hamiltonian of the 3-level atom interacting with a coupling laser with real Rabi-frequency \( \Omega_p \) and a probe laser with Rabi-frequency \( \Omega_p \) (Fig. 5), in a rotating frame as

\[
H_{int} = \frac{\hbar}{2} \begin{bmatrix}
0 & 0 & \Omega_p \\
0 & -2(\Delta_1 - \Delta_2) & \Omega_p \\
\Omega_p & \Omega_c & -2\Delta_1
\end{bmatrix}.
\]
Here $\Delta_1 = \omega_{31} - \omega_p$ and $\Delta_2 = \omega_{32} - \omega_c$ are the detunings of the probe and coupling laser frequencies $\omega_p$ and $\omega_c$ from the corresponding atomic transitions.

A succinct way of expressing the eigenstates of the interaction Hamiltonian Eq.(1) is in terms of the “mixing angles” $\theta$ and $\phi$ that are dependent in a simple way upon the Rabi couplings as well as the single-photon ($\Delta_1 = \Delta$) and two-photon ($\delta = \Delta_1 - \Delta_2$) detunings (see Figure 5). For two-photon resonance ($\Delta_2 = \Delta_1$, or $\delta = 0$) the mixing angles are given by:

$$\tan \theta = \frac{\Omega_p}{\Omega_c},$$

(2)

$$\tan 2\phi = \frac{\sqrt{\Omega_p^2 + \Omega_c^2}}{\Delta}.$$  

(3)

The eigenstates can then be written in terms of the bare atom states:

$$|a^+\rangle = \sin \theta \sin \phi |1\rangle + \cos \phi |3\rangle + \cos \theta \sin \phi |2\rangle,$$  

(4)

$$|a^0\rangle = \cos \theta |1\rangle - \sin \theta |2\rangle,$$  

(5)

$$|a^-\rangle = \sin \theta \cos \phi |1\rangle - \sin \phi |3\rangle + \cos \theta \cos \phi |2\rangle.$$  

(6)

The readers attention is drawn to the following features: Whilst the state $|a^0\rangle$ remains at zero energy the pair of states $|a^+\rangle$ and $|a^-\rangle$ are shifted up and down by an amount $\hbar \omega^\pm$

$$\hbar \omega^\pm = \frac{\hbar}{2}(\Delta \pm \sqrt{\Delta^2 + \Omega_p^2 + \Omega_c^2}).$$  

(7)

The states $|a^\pm\rangle$ retain a component of all of the bare atomic states, but in contrast state $|a^0\rangle$ has no contribution from $|3\rangle$ and is therefore the dark-state since if the atom is formed in this state there is no possibility of excitation to $|3\rangle$ and subsequent spontaneous emission.

It should be noted that the dark-state will always be one of the possible states of the dressed system, but that the details of the evolution of the fields can determine whether the atom is in this state $|a^0\rangle$, in $|a^\pm\rangle$ or an admixture. Evolution into the dark-state via optical pumping (through spontaneous decay from $|3\rangle$) is one way to trap population in this state that is well known in laser spectroscopy and laser-atom manipulation. EIT offers an alternative, adiabatic, and much more rapid route to evolve into this state.

To see the origin of EIT using the dressed state picture above consider the case of a weak probe, that is $\Omega_p \ll \Omega_c$. In this case $\sin \theta \to 0$ and $\cos \theta \to 1$ which results in the dressed eigenstates shown in Figure 4. The ground state becomes identical to the dark-state $|a^0\rangle = |1\rangle$ from which excitation cannot occur. Furthermore, when the probe is on resonance ($\Delta = 0$) then $\tan \phi \to 1$ (i.e. $\phi = \pi/2$), and then $|a^+\rangle = 1/\sqrt{2}(|2\rangle + |3\rangle)$ and $|a^-\rangle = 1/\sqrt{2}(|2\rangle - |3\rangle)$; these are the usual dressed states relevant to EIT in the limit of a strong coupling field and a weak probe. Conversely the picture of dark-states show us that the EIT interference will survive in a Lambda system in the limit of a pair of strong fields and so is relevant to the situation of maximal coherence.

To ensure that a dark-state is formed an adiabatic evolution of the field is often adopted (Oreg et al., 1984). The physically most interesting example of this involves a so called counter-intuitive pulse sequence. This was first demonstrated by Bergmann and his co-workers (Ganbay et al., 1990; Kuklinski et al., 1989) who termed this Stimulated Raman Adiabatic Passage (STIRAP). With the system starting with all the amplitude in the ground-state $|1\rangle$ the laser field at $\omega_c$ is first applied. This is like the EIT situation since $\Omega_p \ll \Omega_c$ and the dark-state indeed corresponds exactly to $|1\rangle$ with negligible population either in $|2\rangle$ or $|3\rangle$. But now $\Omega_p$ is gradually (i.e. adiabatically) increased and at the same time $\Omega_c$ is gradually decreased so that eventually $\Omega_c \ll \Omega_p$ and the dark-state becomes $|a^0\rangle = -|2\rangle$. This population transfer is attained via the “maximally coherent” dark-state when $\Omega_p = \Omega_c$, i.e. $\theta = \pi/4$ in which the state takes the form $|a^0\rangle = 1/\sqrt{2}(|1\rangle - |2\rangle)$. A full description of this process and the conditions for adiabaticity are given in the next section.

III. ATOMIC DYNAMICS AND OPTICAL RESPONSE

The essential features of EIT and many of its applications can be quantitatively described using a semiclassical analysis: this will be the focus of this section where we will assume continuous-wave (cw) classical fields interacting with a single atom that can be modelled as a lambda system (Figure 5). For the derivation of linear and non-linear susceptibilities, we concentrate on the steady-state solution of the atomic master equation. While a master equation analysis is general and can be used for the treatment of non-perturbative field effects, a single-atom wave-function approach is simpler and more illustrative: we will therefore use the latter to discuss the dressed-state interpretation of EIT. As many of the central features of EIT are related to optically thick media we will discuss the light transmission of such a medium. The process of establishing EIT will then be examined and the connection to Raman adiabatic passage will be established. Finally, we will discuss EIT inside optical resonators and briefly review some applications of EIT that will not be expanded upon in the following sections of the review.

A. Master equation and linear susceptibility

We consider an ensemble of identical atoms whose dynamics can be described by taking into account only three of its eigenstates. In the absence of electromagnetic fields, all atoms are assumed to be in the lowest energy state $|1\rangle$ (Fig. 5). State $|2\rangle$ has the same parity as $|1\rangle$ and is assumed to have a very long coherence time. The highest energy state $|3\rangle$ is of opposite parity and has
a non-zero electric-dipole coupling to both $|1\rangle$ and $|2\rangle$. A (near) resonant non-perturbative electromagnetic field of frequency $\omega_c$, termed the coupling field, is applied on the $|2\rangle$-$|3\rangle$ transition. A probe field of frequency $\omega_p$ is applied on the $|1\rangle$-$|3\rangle$ transition: EIT is primarily concerned with the modification of the linear and nonlinear optical properties of this - typically perturbative - probe field. We emphasize that most atomic systems have a subspace of their state-space that can mimic this simplified picture when driven by near resonant polarized electromagnetic fields.

The time-dependent interaction Hamiltonian in the interaction picture that describes the atom-laser coupling is:

$$H_{int} = -\frac{\hbar}{2} \left[ \Omega_p(t) \hat{\sigma}_{31} e^{i\Delta_1 t} + \Omega_c(t) \hat{\sigma}_{52} e^{i\Delta_2 t} + h.c. \right]$$

where $\Omega_c(t)$ and $\Omega_p(t)$ denote the Rabi frequency associated with the coupling and probe fields. $\hat{\sigma}_{ij} = |i\rangle \langle j|$ is the atomic projection operator ($i,j = 1,2,3$). This semiclassical Hamiltonian can be obtained from the fully quantized interaction-picture interaction Hamiltonian by replacing annihilation and creation operators by $c$-numbers.

The dynamics of the laser driven atomic systems is governed by the master equation for the atomic density operator

$$\frac{d\rho}{dt} = \frac{1}{i\hbar} \left[ H_{int}\rho \right] + \Gamma_{31} \left[ 2\sigma_{13} \rho \sigma_{31} - \sigma_{33} \rho - \rho \sigma_{33} \right] + \frac{\Gamma_{32}}{2} \left[ 2\sigma_{23} \rho \sigma_{32} - \sigma_{33} \rho - \rho \sigma_{33} \right] + \frac{\gamma_{2dep}}{2} \left[ 2\sigma_{22} \rho \sigma_{22} - \sigma_{22} \rho - \rho \sigma_{22} \right] + \frac{\gamma_{3dep}}{2} \left[ 2\sigma_{33} \rho \sigma_{33} - \sigma_{33} \rho - \rho \sigma_{33} \right],$$

where the second and third terms on the right hand side describe spontaneous emission from state $|3\rangle$ to states $|1\rangle$ and $|2\rangle$, with rates $\Gamma_{31}$ and $\Gamma_{32}$, respectively. We assume here that the thermal occupancy of the relevant radiative modes are completely negligible for optical frequencies, this approximation is easily justified. A detailed derivation of this master equation is given in (Cohen-Tannoudji et al., 1992). We have also introduced energy conserving dephasing processes with rates $\gamma_{3dep}$ and $\gamma_{2dep}$, the latter determines one of the fundamental time-scales for practical EIT systems as we will see shortly. For convenience, we define the total spontaneous emission rate out of state $|3\rangle$ as $\Gamma_3 = \Gamma_{31} + \Gamma_{32}$. The coherence decay rates are defined as $\gamma_{31} = \Gamma_3 + \gamma_{3dep}$, $\gamma_{32} = \Gamma_3 + \gamma_{3dep} + \gamma_{2dep}$, and $\gamma_{21} = \gamma_{2dep}$.

The polarization generated in the atomic medium by the applied fields is of primary interest since it acts as a source term in Maxwell's equations and determines the electromagnetic field dynamics. The expectation value of the atomic polarization is

$$\bar{P}(t) = -\frac{1}{V} \sum_i \langle cr_i \rangle / V$$

To obtain the expression in the second line, we assumed that all $N_{atom}$ atoms contained in the volume $V$ couple identically to the electromagnetic fields. The time-dependent exponentials arise from the conversion to the Schrödinger picture. Assuming $\mu_{13} = \mu_{13} \mathbf{z}$ and $\mu_{23} = \mu_{23} \mathbf{z}$, we let $g = N_{atom}/V$ and obtain $P_c(t) = P(t)$ as

$$P(t) = g \left[ \mu_{13} \rho_{31} e^{-i\omega_{31} t} + \mu_{23} \rho_{32} e^{-i\omega_{32} t} + c.c. \right].$$

We now focus on the perturbative regime in the probe field and evaluate the off-diagonal density matrix elements $\rho_{31}(t), \rho_{32}(t)$, and $\rho_{12}(t)$ to obtain $P(t)$, or equivalently, the linear susceptibility $\chi^{(1)}(-\omega_p, \omega_p)$. Taking $\rho_{11} \simeq 1$ and using a rotating frame to eliminate fast exponential time-dependences, we find

$$\rho_{32} = -\frac{i \Omega_p e^{i\Delta_1 t}}{(\gamma_{21} + i2\Delta_2)\rho_{13}},$$

$$\rho_{12} = -\frac{i \Omega_p e^{i\Delta_2 t}}{(\gamma_{21} + i2\Delta_2 - \Delta_1)\rho_{13}},$$

$$\rho_{31} = \frac{i \Omega_p e^{i\Delta_1 t}}{(\gamma_{31} + i2\Delta_2)\rho_{21}} + \frac{i \Omega_p e^{i\Delta_2 t}}{(\gamma_{31} + i2\Delta_1)\rho_{21}}.$$  

As in Sec. I we define the single-photon detuning as $\Delta = \omega_{31} - \omega_p$ and two-photon detuning as $\delta = \omega_{31} - \omega_{21} - (\omega_p - \omega - e)$. Keeping track of the terms that oscillate with exp$[-i\omega_p t]$, we obtain

$$\chi^{(1)}(-\omega_p, \omega_p) = \frac{|\mu_{13}|^2 g}{\epsilon_0 \hbar} \times$$

$$\times \left[ \frac{4\delta (|\Omega_c|^2 - 4\delta \Delta) - 4\Delta \gamma_{21}^2}{(|\Omega_c|^2 + (\gamma_{31} + i2\Delta)(\gamma_{21} + i2\delta))^2} \right.$$  

$$+ i \frac{8\delta^2 \gamma_{31} + 2\gamma_{21} (|\Omega_c|^2 + 2\gamma_{21} \gamma_{31})}{(|\Omega_c|^2 + (\gamma_{31} + i2\Delta)(\gamma_{21} + i2\delta))^2} \right].$$

The linear susceptibility given in Eq. (13) contains many of the important features of EIT. First, it must be mentioned that Eq. (13) predicts Autler-Townes splitting of an atomic resonance due to the presence of a non-perturbative field. There is more however in this expression than the modification of absorption due to the appearance of dressed atomic-states: in particular, for two-photon Raman resonance ($\delta = 0$), both real and imaginary parts of the linear susceptibility vanish in the ideal limit of $\gamma_{21} = 0$. As depicted in Figure 7, this result is independent of the strength of the coherent coupling field. It should be noted that changing the Rabi-frequency of the coupling laser only changes the spectral profile of absorption, the integral of Im$[\chi]$ over $\Delta$ is conserved as $\Omega_c$ is varied. In the limit $|\Omega_c| > \gamma_{31}$, the absorption profile carries the signatures of an Autler-Townes doublet. Important features in absorption, such as vanishing loss at $\delta = 0$ and enhanced absorption on the low and high
energy sides of the doublet, become evident on a closer inspection. For \( \Omega_c \ll \gamma_{31} \), we obtain a sharp transmission window with a linewidth much narrower than \( \gamma_{31} \). In this latter case, it becomes apparent that the modifications in the linear susceptibility call for an explanation based on quantum interference phenomena rather than a simple line splitting. Before proceeding with this discussion, we highlight some of the other important properties that emerge in Eq. (13).

First and foremost it must be noted that the possibility of eliminating absorption in an otherwise optically thick medium has been demonstrated in several laboratories in systems ranging from hot atomic vapor cells to magnetically trapped Bose-condensed atoms. Figure 8 shows the absorption profile obtained in the first experiment to demonstrate the effect, carried out by Boller and co-workers (Boller et al., 1991). The application of a coupling field, opens up a transparency window in an otherwise completely opaque atomic cell. This experiment was carried out using an autonionizing state \( |3\rangle \) of strontium with \( \gamma_{31} \sim 2 \times 10^{11} \text{ s}^{-1} \). The decay rate of the lower state coherence \( \gamma_{21} \sim 4 \times 10^9 \text{ s}^{-1} \) is determined by the collisional broadening for an atomic density of \( g \sim 5 \times 10^{15} \text{ cm}^{-3} \).

The transparency obtained at two-photon resonance is independent of the detuning of the probe field from the bare \( |1\rangle\rightarrow|3\rangle \) transition \( (\Delta) \). As \( \Delta \) increases however, the distance between the frequencies where one obtains transparency and maximum absorption becomes smaller, thereby limiting the width of the transparency window. At the same time, the transmission profile and the associated dispersion becomes highly asymmetric for \( \Delta > \gamma_{31} \). Figure 9 shows contour plots of \( \text{Im}[\chi^{(1)}] \) as a function of the detunings \( \Delta_1 \) and \( \Delta_2 \) of the two fields as well as the single \( (\Delta) \) and two \( (\delta) \) photon detunings. It is evident that for large detuning \( \Delta_2 \) of the coupling field the absorption spectrum is essentially that of a two-level system with an additional narrow Raman peak close to the two-photon resonance. Exactly at the two-photon resonance point \( \Delta_1 = \Delta_2 \) the absorption vanishes independent of the single-photon detuning.

In the limit \( \Delta_2 = 0 \) and for \( \Omega_c \ll \gamma_{31} \), the real part of the susceptibility seen by a probe field varies rapidly at resonance \( (\Delta \sim 0) \). In contrast to the well-known (single) atomic resonances, the enhanced dispersion in the EIT system is associated with a vanishing absorption coefficient, implying that ultra-slow group velocities for light pulses can be obtained in transparent media which will be discussed extensively in the following section.

Typically, observation of coherent phenomena in atomic gases is hindered by dephasing due to collisions and laser fluctuations as well as inhomogeneous broadening due to the Doppler effect. While recent EIT experiments are carried out using ultra-cold atomic gases driven by highly coherent laser fields where Eq. (9) provides a perfect description of the atomic dynamics, it is important to analyze the robustness of EIT against these non-ideal effects.

In actual atomic systems, the dephasing rate of the forbidden \( |1\rangle\rightarrow|2\rangle \) transition is non-zero due to atomic collisions. All the important features of EIT remain observable even when \( \gamma_{21} \neq 0 \), provided that the coupling field...
This implies that EIT effects can be observed in dense atomic gases, or even solids, provided that there is a metastable transition with a relatively long dephasing lifetime. Collisional broadening dominates over lifetime broadening. We first note from Fig. 10b that the absorption profile appears virtually unchanged with respect to the ideal case, provided that the inequality (14) is satisfied. The transparency is no longer perfect however, and the residual absorption due to $\gamma_{21}$ provides a fundamental limit for many of the EIT applications. For the $\gamma_{21} \gg \gamma_{31}$ case depicted in Fig. 10c, the absorption minimum is absent: in this limit a constructive interference enhances the absorption coefficient in between the two peaks.

The linear susceptibility given in Eq. 13 depends only on the total coherence decay rates, and not on the population decay rates of the atomic states. As a consequence, the strong quantum interference effects depicted in Figs. 3a-c are observable even in systems where collisional broadening dominates over lifetime broadening. This implies that EIT effects can be observed in dense atomic gases, or even solids, provided that there is a metastable transition with a relatively long dephasing rate that satisfies $\gamma_{21} \ll \gamma_{31}$. EIT in such a collisionally broadened atomic gas was first reported in 1991 (Field et al., 1991).

Amplitude or phase fluctuations of the nonperturbative coupling laser can have a detrimental effect on the observability of EIT. Instead of trying to present a general analysis of the effect of coupling laser linewidth on linear susceptibility, we discuss several special cases which are of practical importance. When the laser linewidth is due to phase fluctuations that can be modeled using the Wiener-Levy phase diffusion model, it can be shown that the resulting coupling laser linewidth directly contributes to the coherence decay rates $\gamma_{21}$ and $\gamma_{32}$ (Imamoglu, 1991). In contrast, for a coupling field with large amplitude fluctuations, the absorption profile could be smeared out. In applications where a non-perturbative probe field is used, fluctuations in both laser fields are important. If however, the two lasers are obtained from the same fluctuating laser source using electro-optic or acousto-optic modulation, then to first order EIT is preserved.

Doppler broadening is ubiquitous in hot atomic gases. If the Lambda system is based on two hyperfine-split metastable states $|1\rangle$ and $|2\rangle$, then the Doppler broadening has no adverse effect on EIT, provided that one uses co-propagating probe and coupling fields. In other cases, the susceptibility given in Eq. (13) needs to be integrated over a Gaussian density of states corresponding to the Gaussian velocity distribution of the atoms. Qualitatively, the presence of two-photon Doppler broadening with width $\Delta \nu_{dopp} \gg \Omega_c$ will wash out the level-splitting and the interference. EIT can be recovered by increasing the coupling field intensity so as to satisfy $\Omega_c > \Delta \nu_{dopp}$; in this limit we obtain

$$\text{Im} \left[ \chi^{(1)} (-\omega_p, \omega_p) \right]_{\omega_p=\omega_{31}} \propto \frac{\Delta \nu_{dopp}^2 \gamma_{31}}{\Omega_c^4}. \quad (15)$$

This strong dependence on reciprocal $\Omega_c$ is a direct consequence of robust quantum interference. We emphasize that in this large coupling field limit, we can consider $\Omega_c$ as the effective detuning from the dressed-state resonances. Thanks to quantum interference, this effective detuning can be used to suppress absorption much more...
strongly in comparison to non-interfering systems where the absorption coefficient is only proportional to the inverse detuning squared.

B. Effective Hamiltonian and dressed-state picture

It is well known in quantum optics that for a given master equation in the so-called Lindblad form, one can obtain an equivalent stochastic wave-function description of the dynamics of the system based on the evolution via a non-Hermitian effective Hamiltonian and quantum jump processes (Carmichael, 1993; Dalibard et al., 1992; Gardiner et al., 1992). Further simplification of the description of the dynamics is obtained when the probability of a quantum jump process is negligibly small: in this limit, the non-unitary wave-function evolution induced by the effective Hamiltonian captures most of the essential physics and can be used to calculate linear and nonlinear susceptibilities. This simplification applies for an atomic system where the ground-state (which has near-unity occupancy) is coupled to excited states via weak electromagnetic fields. In the case of EIT, this is exactly the case when atoms in state $|1\rangle$ are driven by a weak probe field, independent of the strength of the coupling field.

Our starting point is the Hamiltonian of Eq. (8): to incorporate the effect of radiative decay out of state $|3\rangle$, we introduce an anti-Hermitian term, $-i\hbar\Gamma_3\sigma_{33}/2$ which reproduces the correct decay terms in the master equation. Conversely, the physics that this combined non-Hermitian Hamiltonian fails to capture is the re-population of atomic states due to spontaneous emission down to states $|1\rangle$ and $|2\rangle$. Since we cannot describe pure dephasing processes using a wave-function model, we can use population decay of state $|2\rangle$ (at rate $\Gamma_2$) to mimic dephasing: we therefore introduce an additional anti-Hermitian decay term to obtain an effective Hamiltonian in the rotating frame

$$H_{eff} = \frac{\hbar}{2}[\Omega_p \sigma_{11} + \Omega_c \sigma_{32} + h.c.] + \hbar(\Delta - \frac{i}{2}\Gamma_3)\sigma_{33} + \hbar(\delta - \frac{i}{2}\Gamma_2)\sigma_{22}. \quad (16)$$

We reemphasize that for $\Gamma_2 = 0$ and $\delta = 0$, one of the eigenstates of $H_{eff}$ is

$$|a^0\rangle = \frac{1}{\Omega_c^2 + \Omega_p^2}[\Omega_c|1\rangle - \Omega_p|2\rangle], \quad (17)$$

with an eigenfrequency $\hbar\omega_0 = 0$, independent of the values of the Rabi frequencies and the single-photon detuning. We here have assumed real-valued Rabi-frequencies, which is always possible as long as propagation effects are not considered. In the time-dependent response, we will see that the emergence of EIT can be understood as a result of the system moving adiabatically from bare state $|1\rangle$ in the absence of fields to $|a^0\rangle$ in the presence of the fields.

The structure of this dark eigenstate provides us the simplest picture with which we can understand coherent population trapping and EIT: the atomic system under the application of the two laser fields satisfying $\delta = 0$ move into $|a^0\rangle$, which has no contribution from state $|3\rangle$. Since the population in state $|3\rangle$ is zero, there is no spontaneous emission or light scattering and hence no absorption. In the limit of a perturbative probe field where $|a^0\rangle \sim |1\rangle$, we have an alternative way of understanding the vanishing amplitude in $|3\rangle$: the atom has two ways of reaching the dissipative state $|3\rangle$: either directly from state $|1\rangle$ or via the path $|1\rangle - |3\rangle - |2\rangle - |3\rangle$. The latter has comparable amplitude to the former since $\Omega_c$ is non-perturbative: as a result, the amplitudes for these paths can exhibit strong quantum interference.

We can alternatively view the atomic system in a different basis - one that would have yielded a diagonal Hamiltonian matrix had the dissipation and the probe coupling been vanishingly small. This dressed-state basis is obtained by applying the unitary transformation on the (bare-basis) state-vector $|\Psi(t)\rangle$

$$|\Psi_d(t)\rangle = U|\Psi(t)\rangle, \quad (18)$$

where

$$U = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos \phi & \sin \phi \\ 0 & -\sin \phi & \cos \phi \end{bmatrix}. \quad (19)$$

Here $\phi$ is defined by $\tan(2\phi) = \Omega_c/\Delta_2$. Application of this transformation to $H_{eff}$ yields

$$\tilde{H}_{eff} = -\frac{\hbar}{2} \begin{bmatrix} 0 & \Omega_p \sin \phi & \Omega_p \cos \phi \\ \Omega_p \sin \phi & -2\Delta_- + i\Gamma_- & -\frac{i}{2}\Gamma_+ + \frac{i}{2}\Gamma_- \\ \Omega_p \cos \phi & -\frac{i}{2}\Gamma_- + \frac{i}{2}\Gamma_+ & -2\Delta_+ + i\Gamma_+ \end{bmatrix}. \quad (20)$$

Here, $\Delta_-$ and $\Delta_+$ denote the detunings of the probe field from the dressed resonances, $\Gamma_- = (\Gamma_2 \cos^2 \phi + \Gamma_3 \sin^2 \phi)$, $\Gamma_+ = (\Gamma_2 \sin^2 \phi + \Gamma_3 \cos^2 \phi)$, and $\Gamma_{-+} = (\Gamma_2 - \Gamma_3) \sin 2\phi$. We can calculate the absorption rate of the probe photons by determining the eigenvalues of $\tilde{H}_{eff}$ in the limit of perturbative $\Omega_p$. The imaginary part of the eigenvalue $\text{Im}(-\lambda_1)$ that corresponds to $|1\rangle$ as $\Omega_p \to 0$ for $\Gamma_2 = 0$ is

$$\text{Im}(\lambda_1) = \frac{-\Omega_c^2 \Gamma_3 (\Delta_+ \sin^2 \phi + \Delta_- \cos^2 \phi)^2}{4\Delta_-^2 \Delta_+^2 + (\Delta_+ \Gamma_3 \sin^2 \phi + \Delta_- \Gamma_3 \cos^2 \phi)^2} \propto \text{Im} \left[ \chi^{(1)}(-\omega_p, \omega_p) \right]. \quad (21)$$

While this expression contains the same information as Eq. (13), appearance of the square of the sum of two amplitudes in the numerator of Eq. (21) makes the role of quantum interference clearer.

$\tilde{H}_{eff}$ given in Eq. (20) has exactly the same form as the one describing Fano interference of two autoionizing states (Fano, 1961; Harris, 1989). It is the presence of imaginary off-diagonal terms $\kappa_{23} = \kappa_{32} = 0.5(\Gamma_2 - \Gamma_3) \sin 2\phi$ that gives rise to quantum interference in absorption. When $\Gamma_2 = 0$, we find that $\kappa_{23} = \sqrt{\Gamma_2 \Gamma_3}$.
where \( \Gamma_{2d} = \Gamma_2 \cos^2 \phi + \Gamma_3 \sin^2 \phi \) and \( \Gamma_{3d} = \Gamma_2 \sin^2 \phi + \Gamma_3 \cos^2 \phi \). In this case, we find that the interference in absorption is destructive when the probe field is tuned in between the two dressed-state resonances, with a perfect cancellation for \( \delta = 0 \). When \( \Gamma_3 = 0 \), we have \( \kappa_{23} = -\sqrt{\Gamma_{2d} \Gamma_{3d}} \); in this case, the quantum interference is constructive for probe fields tuned in between the two dressed-states and destructive otherwise. Even though absorption is suppressed for \( |\delta| \gg \Omega_c \), we never obtain complete transparency or a steep dispersion in this \( \Gamma_3 = 0 \) case. Finally, for \( \Gamma_2 = \Gamma_3 \), the imaginary off-diagonal terms vanish; in this limit, there are no interference effects. These results were already depicted in Fig. 10.

The complete elimination of absorption for \( \delta = 0 \) in the limit \( \Gamma_2 = 0 \) can therefore be understood by invoking the arguments used by Fano (Fano, 1961); let’s assume that the decay of state \( |3 \rangle \) is due to spontaneous emission into state \( |f \rangle \). For an atom initially in state \( |1 \rangle \), a probe photon absorption event results in the generation of a (Raman) scattered photon and leaves the atom in state \( |j \rangle \).

To obtain the nonlinear susceptibility, we assume both additional driving term \( \Omega_{eff} \) that couples states \( |1 \rangle \) and \( |2 \rangle \) coherently. Since we have assumed this transition to be dipole-forbidden, we envision that this effective coupling is mediated by two laser fields (at frequencies \( \omega_a \) and \( \omega_b \)) and a set of intermediate (non-resonant) states \( |i \rangle \)

\[
\Omega_{eff} = \sum_{i} \Omega_a \Omega_b \left( \frac{1}{\omega_{11} - \omega_a} + \frac{1}{\omega_{11} - \omega_b} \right). \tag{22}
\]

Here \( \Omega_a = \mu_{1a} E_a / \hbar \) and \( \Omega_b = \mu_{1b} E_b / \hbar \).

We can now rewrite the effective Hamiltonian for sum frequency generation as

\[
H_{eff}^{(3)} = -\frac{\hbar}{2} \left[ \Omega_{eff} \hat{\sigma}_{21} + \Omega_p \hat{\sigma}_{31} + \Omega_c \hat{\sigma}_{32} + h.c. \right] + \hbar(\Delta - \frac{i}{2} \Gamma_3) \hat{\sigma}_{33} + h(\delta - \frac{i}{2} \Gamma_2) \hat{\sigma}_{32}. \tag{23}
\]

To obtain the nonlinear susceptibility, we assume both \( \Omega_{eff} \) and \( \Omega_p \) to be perturbative. The generated polarization at \( \omega_p \) is given by

\[
P(t) = g p_{13} a_3(t) \exp(-i \omega_p t) + c.c., \tag{24}
\]

where \( a_3(t) \) is the probability amplitude for finding a given atom in state \( |3 \rangle \); this amplitude is calculated using \( H_{eff}^{(3)} \) and has two contributions: the first contribution proportional to \( \Omega_p \) simply gives us the same linear susceptibility we calculated in the previous subsection using the master equation approach. The second contribution proportional to \( \Omega_{eff} \Omega_c \propto \Omega_p \Omega_b \Omega_c \) gives us the nonlinear susceptibility

\[
\chi^{(3)}(-\omega_p, \omega_a, \omega_b, \omega_c) = \frac{2 \mu_{23} \mu_{31} \theta}{3 \epsilon_0 \hbar^3} \frac{1}{\Omega_c^2 + (\gamma_{31} + i 2 \Delta)(\gamma_{21} + i 2 \delta)} \times \sum_i \mu_{1i} \mu_{2i} \left[ \frac{1}{\omega_{11} - \omega_a} + \frac{1}{\omega_{11} - \omega_b} \right]. \tag{25}
\]

This third-order nonlinear susceptibility that describes sum-frequency generation highlights one of the most important properties of EIT: namely, that the nonlinear susceptibilities need not vanish when the linear susceptibility vanishes due to destructive quantum interference. In fact, for \( \Omega_c^2 < \Gamma_3^2 / 2 \), we notice that \( \chi^{(3)}(-\omega_p, \omega_a, \omega_b, \omega_c) \) has a maximum at \( \delta = 0 \) where \( \chi^{(3)}(-\omega_p, \omega_p) = 0 \).

It is perhaps illustrative to compare \( \chi^{(3)} \) obtained for the EIT system with the usual third-order nonlinear susceptibility for non-resonant sum-frequency generation. As expected, the expressions in the two limits have the same form, with the exception of detunings: the two and three photon detunings in the case of non-resonant susceptibility \( 1/(\Delta \omega_1 \Delta \omega_3) \) is replaced by \( 1/(\Omega_c^2 + \gamma_{21} \gamma_{31}) \) in the case of EIT with \( \delta = \Delta = 0 \).

C. Transmission through a medium with EIT

Many of the interesting applications of EIT stem from the spectroscopic properties of dark resonances, i.e. narrow transmission windows, in otherwise opaque media. It is therefore worthwhile to consider the specifics of the EIT transmission profile in more detail. In an optically thick medium it is not the susceptibility \( \chi^{(1)}(-\omega_p, \omega_p) \) itself that governs the spectroscopic properties but the collective response of the entire medium characterized by the amplitude transfer function

\[
T(\omega_p, z) = \exp \left\{ ikz \chi^{(1)}(-\omega_p, \omega_p)/2 \right\}. \tag{26}
\]

with \( k = 2\pi / \lambda \) the resonant wavenumber and \( z \) the medium length. In contrast to absorbing resonances, the width of the spectral window in which an EIT medium appears transparent decreases with the product of normalized density and medium length. Substituting the susceptibility of an ideal EIT medium with resonant control field into (26), one finds that the transmittivity near
resonance \( \delta = 0 \) is a Gaussian with width
\[
\Delta \omega_{\text{trans}} = \frac{\Omega_0^2}{\sqrt{\gamma_3 \gamma_{31}}} \frac{1}{\sqrt{\sigma z}} \quad (\sigma z \gg 1).
\] (27)

Here \( \sigma = 3 \lambda^2 / 2\pi \) is the absorption cross section of an atom and \( \rho \) the atom number density. One recognizes a power broadening of the resonance with increasing Rabi-frequency \( \Omega_0 \) or intensity of the coupling field which can however be partially compensated by increasing the density length product \( \rho \sigma z \). This important property is illustrated in Fig. 11, where a finite decay rate \( \gamma_{31} \) of the coherence between the two lower levels was taken into account as well.

The perfect linear dispersion near the EIT resonance, expressed by the linear dependence of the real part of \( \chi \) on \( \delta \) can be used to detect shifts of the EIT resonance by phase sensitive detection. Defining the frequency variation which leads to an accumulated phase shift of \( 2\pi \) after the medium as dispersive width \( \Delta \omega_{\text{disp}} \), one finds an even stronger reduction with the density-length product
\[
\Delta \omega_{\text{disp}} = \frac{2\pi \Omega_0^2}{\Gamma_{31}} \frac{1}{\rho \sigma z} \quad (\rho \sigma z \gg 1)
\] (28)

This effect which is also illustrated in Fig. 11 plays an important role for applications of EIT in optical detection of magnetic fields (Fleischhauer and Scully, 1994; Scully and Fleischhauer, 1992).

The narrowing of the transparency and dispersive width of an EIT medium with increasing density was experimentally observed by Lukin et al. (1997) in a forward four-wave mixing experiment.

### D. Dark-state preparation and Raman adiabatic passage

In the preceding sections we have discussed EIT under steady-state conditions. We have seen that atoms in a particular superposition state, the dark state \( |a^{(0)}\rangle \), can be decoupled from the interaction. If there is two-photon resonance between atoms and fields, this state is stationary. Properly prepared atoms remain in this state and render the medium transparent. An important characteristics of EIT, which distinguishes it from coherent population trapping is the preparation which in EIT happens automatically through Raman adiabatic passage. This process naturally becomes important if an optically thick medium is considered.

The formation of a dark state can either happen via optical pumping, i.e. by an incoherent process, or via a coherent preparation scheme. The first mechanism is obviously required if the atomic ensemble is initially in a mixed state. To achieve electromagnetically induced transparency in optically thick media, optical pumping may however be not well suited. Here radiation trapping can lead to a dramatic slow-down of the pumping process. If the dark state has a finite lifetime, the resulting effective pump rate may become even too slow to achieve transparency at all (Fleischhauer, 1999). Furthermore the preparation by optical pumping is always associated with a non-recoverable loss of photons from the probe field, which could be fatal if the probe field is a few-photon pulse.

The problems associated with radiation trapping and preparation losses can be avoided if the atoms are initially in a pure state, e.g. state \( |1\rangle \). If \( |1\rangle \) is a nondegenerate ground state with a sufficient energy gap to state \( |2\rangle \) this is in general fulfilled. Under certain conditions the dark state is in this case prepared by a coherent mechanism which does not involve spontaneous emission at all: the process which achieves this is called Stimulated Raman Adiabatic Passage (STIRAP) and was discovered and developed by Bergmann et al. (Gaubatz et al., 1990; Kuklinski et al., 1989) after some earlier related but independent discoveries of Eberly and coworkers (Oreg et al., 1984). In the STIRAP process relevant here, the atoms are returned to the initial state after the interaction with the pulsed probe field, thus conserving the number of photons in the pulse. The STIRAP technique is by now a widely used method in atomic and molecular physics for the preparation of specific quantum states and controlling chemical reactions. It has also a large range of applications in quantum optics and matter-wave interferometry. For recent reviews on this subject see Bergmann et al. (1998) and Vitanov et al. (2001).

In STIRAP, the two coherent fields coupling the states of the three-level Lambda system are considered to be time dependent and in two-photon resonance. In the basis of the bare atomic states \( \{ |1\rangle, |2\rangle, |3\rangle \} \) the corresponding time-dependent interaction Hamiltonian in rotating

![Image](image.png)

**FIG. 11** top: Transmission spectrum \( T(\delta) \) of an EIT medium for different density length products \( \rho \sigma z \). bottom: Illustration of dispersive width.

\[ T = \begin{array}{c}
0.8 \quad 0.6 \quad 0.4 \quad 0.2 \quad 0 \quad -0.2 \quad -0.4 \\
5 \quad 10 \quad 50 \end{array} 
\]
wave approximation and in a proper rotating frame is
\[ H(t) = -\frac{\hbar}{2} \begin{bmatrix} 0 & 0 & 0 & \Omega_p(t) \\ 0 & 0 & 0 & \Omega_c(t) \\ \Omega_p(t) & \Omega_c(t) & -2\Delta \end{bmatrix}. \] (29)

We assume that the relative phase between the two fields is arbitrary but constant and thus both \( \Omega_p \) and \( \Omega_c \) can be taken real. This also implies that the two fields are assumed to be transform limited pulses, or more precisely that the beat-note between the two is transform limited. The essence of STIRAP is that a proper adiabatic change of \( \Omega_p(t) \) and \( \Omega_c(t) \) allows a complete transfer of population from the initial state \( |1\rangle \) to the target state \( |3\rangle \) or vice versa without populating the intermediate excited state \( |2\rangle \) and thus without spontaneous emission.

The underlying mechanism can most easily be understood if the time-dependent interaction Hamiltonian in the rotating wave approximation is written in the basis of its instantaneous eigenstates, given in Eqs. (4-6) with the corresponding eigenvalues, given in Eq. (7). The instantaneous eigenstate
\[ |a^0(t)\rangle = \cos \theta(t)|1\rangle - \sin \theta(t)|2\rangle, \quad \tan \theta = \frac{\Omega_p(t)}{\Omega_c(t)} \] (30)
is the adiabatic dark-state, which has no overlap with the excited state \( |3\rangle \) and thus does not lead to spontaneous emission.

The important property of the adiabatic dark state is that depending on the value of the mixing angle \( \theta(t) \) it can coincide with either one of the bare states \( |1\rangle \) and \( |2\rangle \) as well as any intermediate superposition with fixed relative phase. If \( \theta = 0 \), which corresponds to a coupling field much stronger than the pump field, \( |a^0\rangle = |1\rangle \). Likewise, if \( \theta = \pi/2 \) we have \( |a^0\rangle = -|2\rangle \). Thus if all atoms are initially (i.e. for \( t = -\infty \)) prepared in state \( |1\rangle \) and \( |\Omega_c(-\infty)| \gg |\Omega_p(-\infty)| \), i.e. if the coupling pulse is applied first, the system is in the dark-state at \( t = -\infty \). If the mixing angle \( \theta(t) \) is now rotated from \( 0 \) to \( \pi/2 \) the dark-state changes from \( |1\rangle \) to \( -|2\rangle \). If this rotation is sufficiently slow, the adiabatic theorem guarantees that the state vector of the system follows this evolution. In this way population can be transferred with 100% efficiency and without spontaneous emission losses. The only requirement besides the correct initial conditions are that the time rate of change of the fields is sufficiently slow, i.e. adiabatic, and that the relative phase between the fields is at least approximately constant.

To see how slow the change of the fields has to be to stay within the adiabatic approximation we transform the Hamiltonian matrix (29) for the state amplitudes in the bare basis into the basis of instantaneous eigenstates. Since the transformation matrix \( R \) is explicitly time dependent for the case of time-dependent fields, we find \( \tilde{H} = R^{-1}HR - R^{-1}\dot{R} \), or explicitly,
\[ \tilde{H} = -\hbar \begin{bmatrix} \omega^+(t) & \frac{i}{2} \dot{\theta} & 0 \\ -\frac{i}{2} \dot{\theta} & 0 & -\frac{i}{2} \dot{\theta} \\ 0 & \frac{i}{2} \dot{\theta} & \omega^-(t) \end{bmatrix}, \] (31)

where an overdot means a time derivative. Adiabatic evolution occurs if the off-diagonal coupling, proportional to \( \dot{\theta} \) is sufficiently small compared to the eigenvalues \( |\omega^\pm| \)
\[ \left| \frac{\partial}{\partial t} q_0(t) |a_{\pm}(t)\rangle \right| \ll |\omega^\pm(t)| \] (32)
which for the case of single-photon resonance \( \Delta = 0 \) simplifies to
\[ \Omega(t) \equiv \sqrt{\Omega_p^2(t) + \Omega_c^2(t)} \gg |\dot{\theta}(t)|. \] (33)

In lowest order of the non-adiabatic coupling \( \dot{\theta} \) and in the limit \( |\dot{\theta}| \ll \Gamma_3 \), characteristic for long pulses, the excited state population is given by \( |\dot{\theta}|^2/\Omega^2(t) \), such that spontaneous emission leads to an instantaneous loss rate (Fleischhauer and Manka, 1996; Vitanov and Stenholm, 1997)
\[ \Gamma_{\text{eff}} = \Gamma_3 \frac{|\dot{\theta}(t)|^2}{\Omega^2(t)} \] (34)

If the pump and coupling pulses are not transform limited in such a way that the beat-note phase changes in time (Dalton and Knight, 1982), there will be additional non-adiabatic losses due to this effect.

If all atoms are initially in state \( |1\rangle \), the coupling field \( \Omega_c \) is applied before the probe field \( \Omega_p \), and if the characteristic time change is slow on a scale set by the rms Rabi-frequency, the fields prepare the dark-state of EIT by stimulated Raman adiabatic passage. If the two fields are constant the dark state is also self-prepared by the lasers but this takes a finite amount of preparation energy from the probe field and transfers it into the medium and the coupling field. This preparation energy was first determined by Harris and Luo (1995). It is given by the number of atoms in the probe path \( N \) and the stationary values of the Rabi-frequencies
\[ E_{\text{prep}} = \hbar \omega_p N \frac{\Omega_p^2}{\Omega_c^2}. \] (35)

After the preparation energy is transferred to the atoms and the coupling field, the medium remains transparent. On the other hand if the probe field is also switched off before the coupling field, the dark state eventually returns to the initial state \( |1\rangle \) and the preparation energy is transferred back from the medium and the coupling field to the probe field. In this case there is no net loss of energy from the probe field but only a temporary transfer. As we will see later on in Section IV this temporary transfer of energy has some interesting consequences for the propagation of the fields.

E. EIT inside optical resonators

The large linear dispersion associated with EIT in an optically thick medium can substantially affect the
properties of a resonator system, in particular since the medium dispersion can easily exceed that of an empty cavity. It is therefore natural to investigate the possibility of having an EIT medium inside a cavity, such that one of the high-finesse modes replaces the probe field and couples the ground-state $|1\rangle$ to state $|3\rangle$.

If an empty-cavity resonance $\omega_{c0}$ is sufficiently close to the frequency $\omega_0$ of electromagnetically induced transparency, the presence of the medium will lead to a very strong pulling of the resonance frequency $\omega_c$ of the combined atom–cavity system towards this value (Lukin et al., 1998a). At the same time the resonance width is substantially reduced (Lukin et al., 1998a). To see this let us consider the response function of a resonator containing an EIT medium, i.e. the ratio of circulating to input intensity at frequency $\omega$. For simplicity we assume a uni-directional ring resonator of round-trip length $L$ with a single in-out coupling mirror. Then

$$S(\omega) \equiv \frac{I_{\text{circ}}(\omega)}{I_{\text{in}}(\omega)} = \frac{t^2}{1 + r^2 \kappa^2 - 2r\kappa \cos [\Phi(\omega)]}, \quad (36)$$

where $t$ and $r$ are the amplitude transmittivity and reflectivity of the in-out coupling mirror. In the case of a lossless mirror $r^2 + t^2 = 1$. $\Phi(\omega) = \omega/c(L + |\text{Re}[\chi^{(1)}]|)$ is the phase shift and $\kappa = \exp(-\omega |\text{Im}[\chi^{(1)}]|/c)$ the medium absorption per round trip $L$. $l$ is the length of the EIT medium. Close to two-photon resonance $\text{Re}[\chi^{(1)}] \approx \frac{dn}{d\omega}(\omega - \omega_0)$. This yields for the resonance of the cavity + medium system

$$\omega_c = \frac{1}{1 + \xi} \omega_{c0} + \frac{\xi}{1 + \xi} \omega_0, \quad \text{where} \quad \xi = \omega_0 \frac{dn}{d\omega} \frac{l}{L}. \quad (37)$$

One recognizes a strong frequency pulling towards the atomic EIT resonance since the dispersion $\omega_0 dn/d\omega$ can be rather large. Likewise one finds from (36) that the width of the cavity resonance is changed according to

$$\frac{\Delta \omega_c}{\Delta \omega_{c0}} = \frac{1 - r\kappa}{\sqrt{\kappa(1-r)}} \frac{1}{1 + \xi}. \quad (38)$$

The first term accounts for a small enhancement of the cavity width owing to the additional losses induced by the medium. Much more important is however the second term, which describes a substantial reduction of the cavity linewidth due to the linear dispersion of the medium.

Since the EIT resonance depends on the atomic system as well as on the coupling laser, frequency pulling and line-narrowing are relative to the coupling laser. These effects have been observed experimentally in (Müller et al., 1997). Figure 12 shows the transmission spectrum of an empty cavity and a cavity with an EIT medium. Applications for difference-frequency locking and stabilization have been suggested and the reduction of the beat-note linewidth in a laser system below the Shawlow-Townes limit predicted (Lukin et al., 1998a).

![FIG. 12 Experimental demonstration of linewidth narrowing from (Müller et al., 1997): (a) Transmission spectrum of empty cavity and with EIT medium, (b) magnification of Transmission spectrum with EIT.](image_url)

F. Enhancement of refractive index, magnetometry and lasing without inversion

We now undertake a brief survey of some applications of EIT and related phenomena which will not be discussed in detail in the following sections. The use of laser fields to control the refractive index has created much attention. A suggestion for controlling phase-matching in non-linear mixing through the action of an additional field was made by Tewari and Agarwal (1986), but it is since the advent of EIT that these ideas have been extensively studied. Harris treated the refractive properties of an EIT medium theoretically (Harris et al., 1992). In pulsed laser experiments the vanishing of the susceptibility at resonance leads to the elimination of distortion for pulses propagating through a very optically thick medium at resonance (Jain et al., 1995). Related suggestions by Harris for the use of strong off-resonant fields to control the refractive index (Harris, 1994b) have been important in the implementation of maximal coherence frequency mixing schemes with off-resonant lasers.

Extensive theoretical work was carried out by Scully and co-workers (Fleischhauer et al., 1992a,b; Rathe et al., 1993; Scully, 1991; Scully and Zhu, 1992) and others (Wilson-Gordon and Friedman, 1992) on the use of resonant cw fields for modification of the refractive index. The experiments of several workers have extensively studied refractive index modifications in this cw limit (Mose-
ley et al., 1995; Xiao et al., 1995).

Working in Doppler free conditions with the narrow bandwidths attainable through use of cw lasers it is possible to observe very steep EIT induced dispersion profiles. This led to the suggestion of implementation of very sensitive interferometers and magnetometers since tiny variations in frequency couple to large changes in refractive index (Affolderbach et al., 2002; Budker et al., 1999, 1998; Fleischhauer and Scully, 1994; Nagel et al., 1998; Sautenkov et al., 2000; Scully and Fleischhauer, 1992; Stahler et al., 2001). In the magnetometer schemes this greatly increases the sensitivity of the measurement of the Zeeman shifts induced by a magnetic field.

As mentioned earlier, laser without inversion is closely related to the interference important in EIT. There have by now been a number of experimental verifications of this concept with the actual realization of laser action without population inversion in the visible spectral range including several reports of inversionless amplification (Fry et al., 1993; Gao et al., 1992) and (Nottleman et al., 1993) as well as actual demonstrations of laser oscillation without population inversion (Zibrov et al., 1995) and (Padmanabandu et al., 1996). An application of LWI to short wave-length lasing is yet to be shown.

IV. EIT AND PULSE PROPAGATION

So far we have considered EIT only from the point of view of the atomic system and its linear response to stationary monochromatic fields but have not paid attention to propagation effects of pulses. Although the susceptibility discussed in the last section contains already all necessary information, it is worthwhile to devote to this issue a separate section. The more so as the almost perfect transparency at certain frequencies, characteristic for EIT, allows pulse propagation in otherwise optically thick media. Here the action of the medium on the light pulses is – apart from the single frequency for which the medium is transparent – quite substantial, and a number of interesting propagation phenomena are encountered. These effects have often a simple physical origin, but their size and their special properties make them very important for a variety of applications.

A. Linear response slow and ultra-slow light

Let us first consider the properties associated with the linear response of an EIT medium to the probe field $E_p$. We have seen in Sec.III, Eq.(13) that the most characteristic feature of the real part of the susceptibility spectrum is a linear dependence on the frequency close to the two-photon resonance $\delta = 0$. For a negligible decay of the $|1\rangle - |2\rangle$ coherence one finds

$$\text{Re}[\chi^{(1)}] = \eta \frac{2 \Gamma_{31}}{\Omega_c^2} \delta + \mathcal{O}(\delta^2),$$  

(39)

where $\eta = \frac{3}{4\pi} \varphi \lambda^2$ is the normalized density, and $\lambda$ the transition wavelength in vacuum. Since the linear dispersion $dn/d\omega_p$ of the refractive index $n = \sqrt{1 + \text{Re}[\chi]}$ is positive, EIT is associated with a reduction of the group velocity according to

$$v_{gr} \equiv \frac{dn}{d\omega_p} \bigg|_{\delta = 0} = \frac{c}{n + \omega_p \frac{dn}{d\omega_p}},$$  

(40)

which has first been pointed out by Harris et al. (1992). At the same time the index of refraction of the ideal three-level medium is unity and thus the phase velocity of the probe field is just the vacuum speed of light

$$v_{ph} \equiv \frac{\omega_p}{k_p} \bigg|_{\delta = 0} = \frac{c}{n} = c.$$  

(41)

An important property of the EIT system is that the second order term in (39) vanishes exactly if there is also single-photon resonance $\Delta = 0$ of the probe field. As a consequence there is no group velocity dispersion, i.e. no wave-packet spreading. Using (39) yields at two-photon resonance

$$v_{gr} = \frac{c}{1 + n_{gr}} \quad \text{with} \quad n_{gr} = \frac{\alpha \sigma c \Gamma_{31}}{\Omega_c^2},$$  

(42)

where $\eta k = \sigma g$ was used. The reduced group velocity gives rise to a group delay in a medium of length $L$

$$\tau = L \left( \frac{1}{v_{gr}} - \frac{1}{c} \right) = \frac{L n_{gr}}{c} = \sigma L \frac{\Gamma_{31}}{\Omega_c^2}.$$  

(43)

It should be noted that under non-ideal conditions, i.e. if the dark-resonance is not perfectly stable, e.g. due to collisional dephasing of the $|1\rangle - |2\rangle$ coherence or fast phase fluctuations in the beat-note between coupling and probe fields, the denominator in the expression for $n_{gr}$ needs to be replaced by $\Omega_c^2 + \gamma_{31} \gamma_{21}$, where $\gamma_{31}$ and $\gamma_{21}$ are, as defined in Sec.III, the transversal decay rates of the probe transition and the $|1\rangle - |2\rangle$ coherence. In this case there is a lower limit to $v_{gr}$ for a fixed density $g$.

Due to the vanishing imaginary part of the susceptibility, i.e. perfect transparency, at $\delta = 0$, relatively high atom densities $g$ and low intensities of the coupling field $I_c \sim \Omega_c^2$ can be used. Thus the group index $n_{gr}$ can be rather large compared to unity and extremely small group velocities are possible. In the first experiments by Harris and coworkers in lead vapor a group velocity of $v_{gr}/c \approx 165$ was observed (Kasapi et al., 1995). In later experiments by Meschede et al. a dispersive slope corresponding to a value of $v_{gr}/c \approx 2000$ was found (Schmidt et al., 1996). Other earlier experiments in which directly or indirectly slow group velocities were measured have been performed by Xiao et al. (1995) and Lukin et al. (1997). The slow-down of light by EIT attracted a lot of attention when Hau and collaborators observed the spectacular reduction of the group velocity to 17 m/s in a Bose condensate of Na atoms, corresponding to a pulse slow down of seven orders of magnitude (Hau et al.,...
1999). Similarly small values where later obtained in a buffer-gas cell of hot Rb atoms by Kash et al. (1999) and by Budker et al. (1999). More recently a substantial slow-down of the group velocity was observed also in the solid-state by Turukhin et al. (2002).

The loss-less slow-down of a light pulse in a medium is associated with a number of important effects: When a pulse enters such a medium, it becomes spatially compressed in the propagation direction by the ratio of group velocity to the speed of light outside the medium (Harris and Hau, 1999). This compression emerges since when the pulse enters the sample its front end propagates much slower than its back end. At the same time however, the electrical field strength remains the same. The reverse happens when the pulse leaves the sample. In the case of the experiment of Hau et al. (1999) the spatial compression was from a kilometer to a sub-millimeter scale! Since the refractive index is unity at two-photon resonance, reflection from the medium boundary is usually negligible as long as the pulse spectrum is not too large (Kozlov et al., 2002).

Although in the absence of losses the time-integrated photon flux through any plane inside the medium is constant, the total number of probe photons inside the medium is reduced by the factor $v_{gr}/c$ due to spatial compression. Thus photons or electromagnetic energy must be temporarily stored in the combined system of atoms and coupling field. It should be noted that the notion of a group velocity of light is still used even for $v_{gr} \ll c$ where only a tiny fraction of the original pulse energy remains electromagnetic.

It is instructive to consider slow-light propagation from the point of view of the atoms. From this perspective the physical mechanism for the temporary transfer of excitations to and from the medium can be understood as stimulated Raman adiabatic return. Before the probe pulse interacts with the three-level atoms a cw coupling field puts all atoms into state $|1\rangle$ by optical pumping. In this limit state $|1\rangle$ is identical to the dark-state. When the front end of the probe pulse arrives at an atom, the dark-state makes a small rotation from state $|1\rangle$ to a superposition between $|1\rangle$ and $|2\rangle$. In this process energy is taken out of the probe pulse and transferred into the atoms and the coupling field. When the probe pulse reaches its maximum, the rotation of the dark-state stops and is reversed. Thus energy is returned to the probe pulse at its back end. The excursion of the dark-state away from state $|1\rangle$ and hence the characteristic time of the adiabatic return process depends on the strength of the coupling field. The weaker the coupling field the larger the excursion and thus the larger the pulse delay. To put the excursion and thus the larger the pulse delay. To put a quasi-particle picture first introduced by Mazets and Matisov (1996) and independently by Fleischhauer and Lukin (2000) which first applied this concept to pulse propagation. One defines dark ($\Psi$) and bright polariton ($\Phi$) fields according to

$$\Psi(z, t) = \cos \vartheta \mathcal{E}_p(z, t) - \sin \vartheta \sqrt{\rho} \rho_{21}(z, t) e^{i\Delta k z},$$

$$\Phi(z, t) = \sin \vartheta \mathcal{E}_p(z, t) + \cos \vartheta \sqrt{\rho} \rho_{21}(z, t) e^{i\Delta k z},$$

with the mixing angle determined by the group index

$$\tan^2 \vartheta = \frac{\sigma \rho_{31} \Gamma_1}{\Omega_c^2} = n_{gr}. \quad (46)$$

$\mathcal{E}_p$ is the normalized, slowly-varying probe field strength, $\mathcal{E}_p = \mathcal{E}_p / (\omega_0 / 2\pi)$, with $\omega$ being the corresponding carrier frequency. $\Delta k = k^{\|} - k_p$, where $k_p$ is the wavelength of the probe field and $k^{\|}$ the projection of the coupling field wave-vector to the axis $z$. $\rho_{21}$ is the single-atom off-diagonal density matrix element between the two lower states.

In the limit of linear response, i.e. in first order of perturbation in $\mathcal{E}_p$, and under conditions of EIT, i.e. for negligible absorption, the set of one-dimensional Maxwell-Bloch equations can be solved (Fleischhauer and Lukin, 2000, 2002). One finds that only the dark-polariton field $\Psi$ is excited, i.e. $\Phi \equiv 0$ and thus

$$\mathcal{E}_p(z, t) = \cos \vartheta \Psi(z, t), \quad (47)$$

$$\rho_{21}(z, t) = -\sin \vartheta \Psi(z, t) e^{-i\Delta k z} \sqrt{\rho}. \quad (48)$$

Furthermore under conditions of single-photon resonance $\Psi$ obeys the simple shortened wave equation

$$\frac{\partial}{\partial t} + c\cos^2 \vartheta \frac{\partial}{\partial z} \Psi(z, t) = 0, \quad (49)$$

which describes a form-stable propagation with velocity

$$v_{gr} = c \cos^2 \vartheta. \quad (50)$$

The slow-down of the group velocity of light in an EIT medium can now be given a very simple interpretation: EIT corresponds to the loss-less and form-stable propagation of dark-state polaritons (DSPs). These quasi-particles are a coherent mixture between electromagnetic and atomic spin excitations, the latter referring to an excitation of the $|1\rangle \rightarrow |2\rangle$ coherence. The admixture of the states described by the mixing angle $\vartheta$ depends on the strength of the coupling field as well as the density of atoms and determines the propagation velocity. In the limit $\vartheta \rightarrow 0$, corresponding to a strong coupling field, the DSP is almost entirely of electromagnetic nature and the propagation velocity is close to the vacuum speed of light $c$. In the opposite limit, $\vartheta \rightarrow \pi/2$, the DSP has the character of a spin excitation and its propagation velocity is close to zero. The concept of dark- and bright-state polaritons can easily be extended to a quantized description of the probe field (Fleischhauer and Lukin, 2000, 2002) as well as quantized matter fields (Juzeliunas and Carmichael, 2002) in which case the polaritons obey approximately Bose commutation relations. This extension is of relevance for applications in quantum information processing and nonlinear quantum optics discussed later on.
So far the atoms have been assumed to be at rest. However, EIT in moving media shows a couple of other interesting features. First of all, if all atoms move with the same velocity \( v \) (\( |v| < c \)) relative to the propagation direction of the light pulse, Galilean transformation rules predict that the group velocity (50) will be modified according to

\[
v_{gr} = c \cos^2 \vartheta + v \sin^2 \vartheta. \tag{51}
\]

This expression can also be obtained from (40) if one takes into account that due to the Doppler effect the susceptibility or the index of refraction becomes \( k \)-dependent (spatial dispersion). It is interesting to note that \( v_{gr} \) can now be zero or even negative for \( \vartheta \neq \pi/2 \) if the atoms move against the propagation of light. In fact since extremely large values of the group index can be achieved in vapor cells, already the thermal motion of atoms at room temperatures can have a significant effect on the effective group velocity. Making use of this effect Kochanovskaya et al. suggested to “freeze” a light pulse in a Doppler-broadened medium, where a specific velocity class is selectively excited (Kochanovskaya et al., 2001).

When the group velocity is decreased the spectral bandwidth of linear dispersion and transparency decreases. In the limit of zero group velocity the dispersion is infinite, which can clearly be supported only over a vanishingly narrow frequency band. Thus in practice absorption and reflection will prevent the full stop of a light pulse with this method, unless a time-dependent coupling field is used which leads to a dynamical narrowing of the pulse spectrum (see below) or the atoms are accelerated. If the flow of atoms is not parallel to the propagation of light, excitation carried by the atoms can be transported in a perpendicular direction (Juzeliunas et al., 2003), which can be observed also in vapor cells where the atoms undergo a diffusive motion (Zibrov et al., 2002).

The slowing down of light has a number of important applications. A reduction of the group velocity of photons leads to an enhanced interaction time in a nonlinear medium, which is important in enhancing the efficiency of such processes (Harris and Hau, 1999; Lukin and Imamoglu, 2001). When the group velocity is decreased the spectral bandwidth of linear dispersion and transparency decreases. In the limit of zero group velocity the dispersion is infinite, which can clearly be supported only over a vanishingly narrow frequency band. Thus in practice absorption and reflection will prevent the full stop of a light pulse with this method, unless a time-dependent coupling field is used which leads to a dynamical narrowing of the pulse spectrum (see below) or the atoms are accelerated. If the flow of atoms is not parallel to the propagation of light, excitation carried by the atoms can be transported in a perpendicular direction (Juzeliunas et al., 2003), which can be observed also in vapor cells where the atoms undergo a diffusive motion (Zibrov et al., 2002).

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As noticed by Harris, the pulse compression is associated with a substantial enhancement of the spatial field gradient in the propagation direction (Harris, 2000) and thus pondermotive dipole forces on atoms, resulting from off-resonant couplings to the light pulse, are substantially amplified. If \( I_0(z) \) denotes the probe intensity at the input of the medium one finds an enhanced force by the ratio \( c/v_{gr} \)

\[
F_{dip} \sim -\frac{d}{dz} I(z) = -\frac{c}{v_{gr}} \frac{d}{dz} I_0(z) \tag{52}
\]

This has important potential applications in atom and molecular optics as well as laser cooling.

The light-matter coupling associated with EIT can also be used for the preparation and detection of coherent matter wave phenomena in ultra-cold quantum gases. E.g. EIT has been suggested as a probe for the diffusion of the relative phase in a two-component Bose Einstein condensate (Ruostekoski and Walls, 1999a,b). Stopping of a light pulse at a spatial discontinuity of the control field was used to create strongly localized shock-waves in a BEC and study their dynamics by Dutton et al. (2001).

Finally interesting effects have been predicted by Leonhardt and coworkers when the propagation of ultra-slow light is considered in non-uniformly moving media. Using an effective classical field theory for slow light where losses are neglected they predicted an Aharonov-Bohm type phase shift in a rotating EIT medium with a vortex flow (Leonhardt and Piwnicki, 1999). Leonhardt et al. also pointed out an interesting analogy between slow-light in a medium with a vortex flow and general relativistic equations for a black hole (Leonhardt and Piwnicki, 2000). This may allow for laboratory studies of some general relativistic effects when adding an inward flow to the vortex (Leonhardt and Piwnicki, 2000; Visser, 2000). The practical observation of an optical analogy of the event horizon of a black hole will however most likely be prevented by absorption and reflection effects.

To assess the potentials of slow light it is important to consider its limitations. A convenient figure of merit for this is not the achievable group velocity itself, but the ratio of achievable delay time \( \tau_d \) of a pulse in an EIT medium to its pulse length \( \tau_p \). One upper limit for the delay time is given by probe absorption due to the finite lifetime of the dark resonance. Furthermore for a pulsed probe field, i.e. for a probe field with a finite spectral width the absorption of the non-resonant frequency components is nonzero even under ideal conditions of an infinitely long-lived dark state. Thus in order for the absorption to be negligible, firstly the decay time of the \( |1 \rangle - |2 \rangle \) transition must be very small and secondly the spectral width of the pulse \( \Delta \omega_p \) has to be much smaller than the transparency width \( \Delta \omega_{trans} \):

\[
\Delta \omega_p \ll \Delta \omega_{trans} = \frac{\frac{\Omega_c^2}{\sqrt{\gamma_{31}^2 \gamma_{31}}} \cdot 1}{\sqrt{\frac{\Gamma_{31}^2}{\gamma_{31}^2}}} \sqrt[3]{\frac{L}{c}}. \tag{53}
\]

It is instructive to express \( \Delta \omega_{trans} \) in this condition in terms of the group delay \( \tau_d = n_{gr} L/c \)

\[
\Delta \omega_{trans} = \sqrt[3]{\frac{\gamma_{31}}{\tau_d}} \frac{L}{c} \sqrt[3]{\frac{\Gamma_{31}}{\gamma_{31}}}. \tag{54}
\]
This discussion shows that it is not possible to bring a pulse to a complete stop by using EIT with a stationary coupling field, since in this case the transparency width would vanish leading to a complete absorption of the pulse. The same argument holds if one tries to stop an ultra-slow pulse by means of atomic motion against the pulse. The same argument holds if one tries to stop the pulse to a complete stop by using EIT with a stationary coupling field, since in this case the transparency width would vanish leading to a complete absorption of [Harris and Hau, 1999].

The upper limit for the ratio of group delay to pulse length is given by:

\[
\frac{\tau_d}{\tau_p} \ll \frac{1}{\sqrt{\rho L}}.
\]  

The product \(\rho L\) is the opacity of the medium in the absence of EIT, i.e. \(\exp(-\rho L)\) is the transmission coefficient at bare atomic resonance. Thus a noticeable time delay of a pulse by EIT requires an optically thick medium with \(\rho L > 1\).

### B. “Stopping of light” and quantum memories for photons

As discussed in the preceding subsection it is not possible to bring a light pulse to a complete stop with stationary EIT. To achieve this goal the group velocity has to be changed in time as was shown by Fleischhauer and Lukin (2000). It should be mentioned that the possibility to transfer spatial excitation distributions of atomic ensembles to light pulses has been pointed out before by Cszeznegi and Grobe (1997). In the following the “stopping” and “re-acceleration” of a light pulse and its potential applications will be discussed in more detail. At this point a word of caution is needed however: The notion “stopping of light” should not be taken literally. As mentioned before, the reduction of the propagation velocity of light in a loss-less, passive medium is always associated with a temporary transfer of its energy to the medium. In the extreme limit of zero velocity relative to the stationary medium no electromagnetic excitation is left at all. Nevertheless the notion of a vanishing group velocity of light has here the same justification as the notion of a group velocity in the case of ultra-slow pulse propagation where also only a tiny fraction of the original excitation remains in the form of photons.

A key conceptual advance with respect to the ability to bring a light pulse to a complete stop occurred when it was realized in (Fleischhauer and Lukin, 2000) that the bandwidth limitation (53) of EIT can be overcome, if the group velocity is adiabatically reduced to zero in time. This can be achieved for example by reducing the Rabi-frequency of the drive field. In this case the spectrum of the probe pulse narrows proportional to the group velocity

\[
\Delta \omega_p(t) = \Delta \omega_p(0) \frac{\nu_{rz}(t)}{\nu_{rz}(0)}
\]  

and the spectrum of the probe pulse stays within the transparency window at all times, provided it fulfills this condition initially. The propagation equation for the dark-state polariton is then still given by eq. (49), with \(\nu \rightarrow \nu(t)\). Adiabatically rotating \(\nu(t)\) from 0 to \(\pi/2\) leads to a deceleration of the polariton to a full stop. At the same time its character changes from an electromagnetic field to a pure spin excitation. Most importantly, provided the rotation is adiabatic, all properties of the original light pulse are coherently transferred to the atomic spin system in this process modulo an overall phase determined by that of the coupling field.

![FIG. 13 Numerical simulation of dark-state polariton propagation with envelope \(\exp(-(z/10)^2)\). The mixing angle is rotated from 0 to \(\pi/2\) and back according to \(\cot \nu(t) = 100(1 - 0.5 \tanh[0.1(t - 15)]) + 0.5 \tanh[0.1(t - 125)])\). Top: polariton amplitude, bottom-left: electric field amplitude , bottom-right: atomic spin coherence; all in arbitrary units. Axes are in arbitrary units with \(c = 1\).](image)

Conditions of adiabaticity have been analyzed by Matsko et al. (2001c) and Fleischhauer and Lukin (2002). The resulting limitations on the rate of change of the coupling field are rather weak. Furthermore if the coupling field and thus the group velocity is already very small even an instantaneous switch-off would only lead to a loss of the very small electromagnetic component of the polariton (Fleischhauer and Mewes, 2002; Liu et al., 2001; Matsko et al., 2001c). Reversing the adiabatic rotation of \(\nu\) by increasing the strength of the coupling field leads to a re-acceleration of the dark-state polariton associated with a change of character from spin-like to electromagnetic. In Fig. 13 a numerical simulation of the stopping of a dark-state polariton and its successive re-acceleration is shown. The transfer of excitation from the probe pulse to the Raman spin excitation and back is apparent. In this way the excitation and all information contained in the original pulse can be reversibly transferred and stored in long-lived spin coherences. It should be noted that the spin excitation does in general not store the photon energy as most of it is transferred to the coupling field by stimulated Stokes emission.
Doppler shifts degenerate Zeeman sublevels were used for states $|1\rangle$ and $|2\rangle$. Moreover to reduce effects of atomic motion a He buffer gas was employed. Here storage times of up to 0.5 ms could be obtained for pulse length of again several $\mu$s. The first successful demonstration of light “storage” in a solid was achieved by Turukhin and co-workers in Pr doped Y$_2$SiO$_5$ after proper preparation of the inhomogeneously broadened material by a special optical hole- and anti-hole burning technique (Turukhin et al., 2002). A direct proof of the coherence of the storage mechanism was provided by Mair et al. (2002), where the phase of the stored light pulse was coherently modified during the storage time by a magnetic field.  

Experimental studies of light storage under conditions of large single-photon detuning were performed moreover by Kozuma et al. (2002). All of these experiments have to be considered as a proof of principle since the transfer efficiency or fidelity of the storage is still rather small due to a variety of limiting effects such as dark-state dephasing, atomic motion, and mode mismatch. An application which does not suffer from these effects is the use of the (partially dissipative) “stopping” of a light pulse at a spatial discontinuity of the drive field to create well defined shock waves in an atomic Bose-Einstein condensate as demonstrated by Dutton et al. (2001).

The most important potential application of light “stopping” is certainly in the field of quantum information. The dark-state polariton picture of light “stopping” also holds for quantized radiation. Here quantum states of photons are transferred to collective excitations of the medium. For example a single-photon state of a single radiation mode $|1\rangle_{\text{ph}}$ is mapped to an ensemble of $N$ three-level atoms according to:

$$|1\rangle_{\text{ph}} \otimes |1,1,\ldots 1\rangle$$

$$\downarrow$$

$$|0\rangle_{\text{ph}} \otimes \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |1,1,\ldots 1,2,\ldots 1\rangle$$

In this way the EIT medium can act as a quantum memory for photons. As compared to other proposals for quantum memories, such as single-atom cavity systems (Cirac et al., 1997), continuous Raman scattering (Schori et al., 2002), or storage schemes based on echo techniques, the EIT based system is capable of storing individual photon wave-packets with high fidelity and without the need for a strongly coupling resonator. It should be mentioned that a time symmetric photon-echo technique has been proposed by Moiseev and Kroll (2001), which is also capable of storing single photons but has not been experimentally implemented. Several limitations of the EIT storage technique have been studied including finite two-photon detuning (Mewes and Fleischhauer, 2002), decoherence (Fleischhauer and Mewes, 2002; Mewes and Fleischhauer, 2004), and fluctuations of coupling parameters (Sun et al., 2003). An extension of the model to three spatial dimensions was given in (Duan et al., 2002).

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**FIG. 14** top: Experimental set-up for observing light storage and retrieval in ultra-cold Na atoms from (Liu et al., 2001). bottom: Measurements of delayed (a) and revived probe pulse after storage (b). Dashed line shows intensity of coupling fields. For details see (Liu et al., 2001).

The classical aspects of “stopping” and “re-accelerating” of light pulses have been experimentally demonstrated by Liu and coworkers in ultra-cold Na (Liu et al., 2001), by Phillips et al. in hot Rb vapor (Phillips et al., 2001), and in solids by Turukhin and collaborators (Turukhin et al., 2002). The experiment of Liu et al. used a similar setup as used for the demonstration of the group velocity reduction to 17 m/s (Hau et al., 1999). Here a light pulse has been slowed and spatially compressed in an ultra-cold and dense vapor of Na atoms at a temperature just above the point of Bose condensation and then “stopped” by turning off the coupling laser. Storage times of up to 1.5 ms for pulse length of a few $\mu$s were achieved. Fig. 14 shows the experimental set-up as well as the storage and retrieval of a light pulse. The use of ultra-cold gases has advantages like the reduction of two-photon Doppler shifts and high densities. They are however not necessary to achieve light “stopping”. In the experiment of Phillips et al. a Rb vapor gas cell at temperatures of about 70-90°C was used. To eliminate two-photon
An interesting extension of the light “stopping” scheme was very recently experimentally demonstrated by Bajcsy et al. (2003). After the coherent transfer of a light pulse to an ensemble of Rb atoms in a vapor cell, two counter-propagating coupling laser were applied rather than one, forming a standing-wave pattern. At the nodes of this pattern, a periodic structure of spatially narrow absorption zones emerged. The two counter-propagating components of the coupling laser regenerated two Stokes pulses with opposite wave-vectors. Thus also the regenerated light pulses formed a standing-wave pattern, which matched that of the counterpropagating drive fields. In this way a stationary pulse of light was created and stored for several μsec. Although the regenerated pulse contained only a small fraction of the original photon number, the electromagnetic field was nonzero during the storage period in contrast to the above mentioned experiments. This generation of stationary light pulses may be an important tool for nonlinear optical processes with few photons.

“Stopping” of light can in principle also be used to prepare atomic ensembles in specific nonclassical or entangled many-particle states (Lukin et al., 2000b). An alternative and experimentally often simpler way to achieve the same is to use absorptive or dispersive interaction of continuous-wave light fields with atomic ensembles and detection. Kuzmich et al. proposed the generation of spin-squeezed ensembles by absorption of squeezed light (Kuzmich et al., 1997), which was experimentally demonstrated later by Hald et al. (1999). Employing similar ideas Polzik suggested to create two Einstein-Podolsky-Rosen entangled atomic ensembles by absorption of quantum-correlated light fields (Polzik, 1999) and Kozhekin et al. proposed a memory for quantum states of cw light fields along these lines (Kozhekin et al., 2000). Recently experimental progress toward the implementation of the cw quantum memory has been reported (Schori et al., 2002). A very intriguing technique to create nonclassical or entangled atomic ensembles is the measurement of the collective spin using off-resonant dispersive interactions. Kuzmich et al. performed in this way quantum non-demolition measurements to prepare quantum states with sub-shot noise spin fluctuations (Kuzmich et al., 2000, 1999). Duan et al. (2000) suggested a detection scheme where coherent light probes simultaneously two atomic ensembles to prepare an Einstein-Podolsky-Rosen correlation between them with potential applications in continuous teleportation of collective spin states. Recently, Julsgaard and coworkers have demonstrated this technique in a remarkable experiment (Julsgaard et al., 2001).

Finally Imamoglu (2002) and James and Kwiat (2002) suggested to use the transfer of photons to atomic ensembles to build single-photon detectors with high efficiency, which are able to distinguish between single and multiphotons. Such a device has particular importance for quantum information processing with linear optical elements (Knill et al., 2001).

C. Nonlinear response: adiabatic pulse propagation and adiabatons

So far we have discussed the interaction of light pulses with ensembles of three-level atoms only in the linear response limit, i.e. assuming a weak probe field. On the other hand the nonperturbative situation of probe and coupling pulses with comparable strength shows a number of interesting additional features. New effects arise in particular because in this case the medium has an effect on the propagation of both pulses.

In almost all theoretical treatments a quasi one-dimensional situation is considered where all fields propagate parallel to the z direction, a homogeneous medium is assumed and atomic motion is disregarded. The properties of the atoms are described by a state vector by symmetric superposition of atomic coherence amplitudes \( C_n(z,t) \), \( n \in \{1,2,3\} \). For simplicity let us assume that the carrier frequencies of coupling and probe pulses are on resonance with the corresponding atomic transitions, which allows us to treat the slowly-varying field amplitudes as real. The evolution of the atomic state is described by a Schrödinger equation for the \( C_n(z,t) \) with the three-level Hamiltonian given in Eq. (29). Because decay of the polarization plays an important role for the propagation of the fields, a decay out of the excited state \( |3\rangle \) with rate \( \Gamma_3 \) is included by adding an imaginary term \( i\hbar \Gamma_3/2 \) to the energy of the excited state. The propagation of the field amplitude is most easily written in terms of the corresponding Rabi-frequencies. In slowly-varying amplitude and phase approximation one finds the shortened wave equations for the probe (\( \Omega_p \)) and coupling pulse (\( \Omega_c \))

\[
\left( \frac{\partial}{\partial t} + c \frac{\partial}{\partial z} \right) \Omega_p(z,t) = \frac{i \alpha_p}{2} C_2(z,t) C_1^*(z,t), \tag{58}
\]

\[
\left( \frac{\partial}{\partial t} + c \frac{\partial}{\partial z} \right) \Omega_c(z,t) = \frac{i \alpha_c}{2} C_2(z,t) C_3^*(z,t), \tag{59}
\]

where the effect of the atoms on the fields is parameterized by the resonant absorption coefficients

\[
\alpha_p = \frac{\omega_p \theta |\mu_{12}|^2}{2\epsilon_0 \hbar}, \quad \alpha_c = \frac{\omega_c \theta |\mu_{13}|^2}{2\epsilon_0 \hbar}, \tag{60}
\]

with \( \mu_{ij} \) being the dipole moments of the corresponding transitions.

If the amplitudes of the fields vary sufficiently slowly such that the adiabatic approximation holds and if the appropriate initial conditions are fulfilled, the atoms stay at all times in the instantaneous dark state, \( |d(t)\rangle = \cos \theta(t) |1\rangle - \sin \theta(t) |2\rangle \), Eq. (17), with \( \tan \theta(t) = \Omega_p(t)/\Omega_c(t) \). As mentioned before an important property of this state is the absence of a component in state \( |3\rangle \). As a consequence of this there is also no dipole moment on either of the two transitions coupled by the probe and coupling pulses. Atoms in the instantaneous dark state have therefore no effect at all. Light and matter are exactly decoupled.
As first noted by Harris (1993) there is another case in which bi-chromatic pulses are exactly decoupled from the atoms even if they do not vary slowly. The dark state corresponding to a pair of pulses with identical envelope, so-called matched pulses, i.e. $\Omega_p(z,t) = \Omega_p f(z,t)$ and $\Omega_c(z,t) = \Omega_c g(z,t)$, is time independent and thus a true eigenstate of the system. After an appropriate preparation of the medium, matched pulses will thus remain exactly decoupled from the interaction for all times and propagate with the vacuum speed of light.

As shown by Harris and Luo a pair of matched pulses applied to an atomic ensemble in the ground state, which in this case does not correspond to the dark state, will prepare the atoms by stimulated Raman adiabatic passage (STIRAP) (Harris and Luo, 1995). During the first few single-photon absorption lengths, the front end of the probe pulse experiences a small loss. In this way a counter-intuitive pulse ordering is established. This provides asymptotic connectivity of the dark state to the initial state of the atoms. The leading end of this slightly deformed pair of pulses will prepare all atoms in the pathway via STIRAP and the pulses can propagate unaffected through the rest of the medium (Eberly et al., 1994; Harris, 1994a). This is illustrated in Fig. 15 where numerical calculations are used to show the propagation of two matched pulses in an unprepared medium.

As noted above an EIT medium does not couple to the fields at all in the adiabatic limit. In order to cover effects like the group-velocity reduction, it is necessary to include first-order non-adiabatic corrections in the dynamics of the atoms, which lead to small contributions to the excited state amplitude. Taking them into account one finds for the state amplitudes

$$C_1 = \cos \theta, \quad C_2 = \frac{2 i}{\Omega} \hat{\theta}, \quad C_3 = -\sin \theta,$$

where $\Omega = \sqrt{\Omega_p^2 + \Omega_c^2}$. It was shown by Grobe et al. (1994) that the nonlinear equations of propagation (58,59), together with (64) are adiabatically integrable even for fields of comparable strength and with arbitrary shape. To see this, we adopt the method of (Fleischhauer and Manka, 1996) and transform the field equations for $\Omega_p$ and $\Omega_c$ in propagation equations for the rms Rabi-frequency $\Omega$ and the non-adiabatic coupling $\hat{\theta}$. Under nearly adiabatic conditions $|\hat{\theta}| \ll \Omega$, a weak-coupling approximation is justified. In this limit, assuming furthermore equal coupling strength $\alpha_p = \alpha_c = \alpha$, one finds that the total Rabi-frequency fulfills the free-space propagation equation

$$\left( \frac{\partial}{\partial t} + \frac{\partial^2}{\partial z^2} \right) \Omega(z,t) \approx 0.$$

No photons are lost by absorption and there is only a coherent transfer from one field to the other. At the same time $\hat{\theta}$ obeys the equation

$$\left( \frac{\partial}{\partial t} + \frac{\partial^2}{\partial z^2} \right) \hat{\theta}(z,t) = -\alpha \frac{\partial}{\partial t} \left( \hat{\theta} \Omega^2 \right),$$

which is exactly integrable. The corresponding solutions, called adiabatons (Grohe et al., 1994), are particularly simple if $\Omega$ is approximately constant over the time interval of interest. In that case the probe and coupling pulses have complementary envelopes and $\hat{\theta}$ propagates without changing form with velocity $v_{\text{gr}} = c/(1 + \alpha/\Omega^2)$. The quasi form-invariant propagation of an adiabaton is tend to adjust their amplitude modulations $f(z,t)$ and $g(z,t)$ in the course of propagation, i.e.

$$\left. \frac{f(z,t)}{g(z,t)} \right|_{z \to \infty} = 1.$$
shown in Fig. 16. If the Rabi-frequency of the coupling field is much larger than that of the probe field, i.e. in the perturbative limit, the adiabaton shows no noticeable dip in the coupling-field strength. This case thus resembles the propagation of a weak pulse in EIT with reduced group velocity.

First experimental evidence of adiabaton propagation in Pb vapor was reported by Kasapi et al. (1996). Adiabatons in more complicated configurations, such as double-Lambda systems and double pairs of pulses, were studied by Cerboneschi and Arimondo (1995) as well as Hioe and Grobe (1994). The effect of non equal coupling strength on the adiabaton propagation was studied by Grigoryan and Pashayan (2001). The adiabaton solutions are approximately stable over many single-photon absorption length. However, as shown by Fleischhauer and Manka (1996), they eventually decay to matched pulses after sufficiently long propagation distances.

For time scales short enough such that decays can be ignored, and equal coupling strength, exact solutions of the nonlinear propagation problem in V and A-type systems exist even beyond the adiabatic approximation. Konopnicki and Eberly (1981) and Konopnicki et al. (1981) have found soliton solutions with identical pulse shape, called simultons. An interpretation of simultons in terms of solitons corresponding to an effective two-level shape, called Konopnicki and Eberly (1981) and Konopnicki (1981) have found soliton solutions with identical pulse shape, called

FIG. 16 Propagation of adiabatons: Shown are amplitudes of probe and coupling fields as function of time in a co-moving frame for $n_g z/c = 0$ (a), 25 (b), 50 (c), and 100 (d) from numerical solution of propagation equations (Fleischhauer and Manka, 1996). Arbitrary space and time units with $c = 1$.

V. ENHANCED FREQUENCY CONVERSION

A. Overview of Atomic Coherence Enhanced Non-Linear Optics

Harris and co-workers identified the enhancement of non-linear optical frequency conversion as a major benefit of electromagnetically induced transparency in their paper of 1990 (Harris et al., 1990). Hemmer and co-workers (Hemmer et al., 1995) introduced the double Lambda scheme as an important tool to EIT-based resonant four-wave mixing. Coherent preparation of a maximal coherence was likewise found by a number of researchers, including the groups of Harris (Jain et al., 1996) in the USA and Hakuta in Japan (Hakuta et al., 1997), to significantly improve the conversion efficiency in four-wave mixing. Whilst destructive interference reduces the linear susceptibility of the laser dressed system this is not so for the non-linear susceptibility in four-wave mixing which in fact undergoes constructive interference. To see how this leads to improved frequency mixing efficiency we need to consider the factors which determine how a generated field can grow effectively in a four-wave mixing process.

Laser fields applied close to resonance in an atomic medium drive various frequency components of the polarization in that medium, both at the frequency of the driving fields and at new frequencies given by combinations of the applied ones. Figure 17 illustrates four schemes in which we will be interested where 3- or 4-level atoms are driven, at single or two-photon resonance, by laser fields. The polarization component at the new frequencies will act as a source of new electromagnetic fields. Thus in the schemes illustrated in Fig. 17 the material polarization $P(\omega_4)$ is driven at frequency $\omega_4$ by fields applied at $\omega_1$, $\omega_2$, and $\omega_3$, and will in its turn drive a new electromagnetic field at $\omega_4$. The growth of the new field depends upon the magnitude of the non-linear source term $P^{nl}(\omega_4)$ and upon the linear polarization $P^l(\omega_4)$ of the medium at this frequency which will determine the absorption and dispersion of the generated field. The equation that describes the growth of this new field $E_4$ is derived from Maxwell’s equations within the slowly-varying envelope approximation (see for example Reintjes (1984)) and can be written in terms of the positive frequency component of the field as:

$$\frac{\partial E_4}{\partial z} + \frac{1}{c} \frac{\partial E_4}{\partial t} = \frac{i}{2 c^2} \left( P^{nl} + P^l \right).$$

Here the vector character of field an polarization was suppressed for simplicity. The term on the right-hand side contains the effect of the linear polarization upon the propagation of the generated field and the nonlinear polarization which is the source of the new field.

The response of the medium to an electric field is governed by its polarisation. In the framework of the density matrices for a three-level atom we can write the positive
and the non-linear response as:

Thus we can express the linear response of the atom to detuning terms in the denominator whilst the linear susceptibility will be characterized by the appearance of three susceptibilities using the familiar polarization expansion.

We can express the total polarization in terms of these dressed and Doppler integrated susceptibilities that can be derived by a full quantum treatment in a calculation.

The form of the equation that describes the (stationary) growth of the (slowly-varying) field $E_4$ can now be recast in terms of these dressed and Doppler integrated susceptibilities:

$$\frac{dE_4}{dz} = i \frac{\omega_4^2}{4c} \chi^{(3)} E_1 E_2 E_3 e^{i \Delta k z} - \frac{\omega_4^2}{2c} |\chi^{(1)}| E_4 + i \frac{\omega_4^2}{2c} \text{Re}[\chi^{(1)}] E_4,$$

where $\Delta k = k_4 - (k_1 + k_2 + k_3)$ is the wave-vector mismatch due to the refractive index effect of all other resonances in the atom and can also include any other phase-matching effects such as beam geometry and plasma dispersion. It is easy to see from Eq. (67) that if the non-linear polarization reaches a large value then the source of the new field will be strong, thus the production of the new field will be enhanced. The absorption and refraction of the generated field are determined by the linear polarization of the medium at the generated frequency. The refraction has a very important effect upon the growth of the field by determining whether or not the generated field remains in phase with the polarization which drives it. This phase-matching condition is essential and dictates (at resonance where $\text{Re}[\chi^{(1)}] = 0$) that we also require $\Delta k = 0$ for efficient growth of the field. For a generated field on resonance both absorption and dispersion can be problematic. In general $\Delta k$ will be finite resulting in a limited length over which the field grows before slipping out of phase with the driving polarization $l_{coh} = 1/\Delta k$. But in the presence of EIT, and with the elimination of the linear susceptibility to vacuum values, the result is no absorption and no refraction due to the resonant level and only non-resonant transitions to other levels contribute to the wave-vector mismatch.

We examine various four-wave-mixing schemes in which EIT or related effects have been found to be advantageous; these are shown in Fig. 17. There are several interconnected mechanisms through which atomic coherence prepared by the applied laser fields can increase the conversion efficiency of these wave-mixing processes:

(i) We have here a four-wave mixing scheme where a two-photon and a single photon resonance are used for the applied fields generating a field in resonance with an atomic transition (figure 17(i)). The generated field and the single photon resonant applied field act as probe and coupling fields respectively in a Lambda EIT scheme. EIT then leads to an elimination of absorption and refraction for the generated field. The non-linear susceptibility

frequency component of the polarization as:

$$P = \rho \text{Tr}[\hat{\rho}] =$$

$$= \rho \left( \mu_{12} \rho_{21} + \mu_{13} \rho_{31} + \mu_{23} \rho_{32} \right).$$

We can express the total polarization $P$ in terms of susceptibilities using the familiar polarization expansion. Thus we can express the linear response of the atom to the field at frequency $\omega_4$ as:

$$P^{(1)}(\omega_4) = \epsilon_0 \chi^{(1)} E_4,$$

and the non-linear response as:

$$P^{(3)}(\omega_4) = \frac{3}{2} \epsilon_0 \chi^{(3)} E_1 E_2 E_3,$$

where $E_j$ is the electric field amplitude associated with the frequency component $\omega_j$. The form of the susceptibilities that can be derived by a full quantum treatment of the atom are extensively discussed in the literature on non-linear optics (see for example Reintjes (1984)). In general for non-resonant non-linear mixing the susceptibility will be characterized by the appearance of three detuning terms in the denominator whilst the linear susceptibility has only a single detuning term. As was shown in section III in the case of EIT it is of course necessary to use the dressed susceptibilities that already include the effect of the strong coupling field to all orders. It is the difference of these susceptibilities from the normal form that give the advantages for non-linear mixing. The form of the dressed susceptibilities for EIT are given in section III. These reflect the resonant nature of the fields involved and now contain the important interference character due to the dressing field. Inclusion of collisional de-phasing and integration over the Doppler profile is usually required to yield the effective susceptibilities that can be used in a calculation.

The form of the equation that describes the (stationary) growth of the (slowly-varying) field $E_4$ can now be recast in terms of these dressed and Doppler integrated susceptibilities:

$$\frac{dE_4}{dz} = i \frac{\omega_4^2}{4c} \chi^{(3)} E_1 E_2 E_3 e^{i \Delta k z} - \frac{\omega_4^2}{2c} |\chi^{(1)}| E_4 + i \frac{\omega_4^2}{2c} \text{Re}[\chi^{(1)}] E_4,$$

where $\Delta k = k_4 - (k_1 + k_2 + k_3)$ is the wave-vector mismatch due to the refractive index effect of all other resonances in the atom and can also include any other phase-matching effects such as beam geometry and plasma dispersion. It is easy to see from Eq. (67) that if the non-linear polarization reaches a large value then the source of the new field will be strong, thus the production of the new field will be enhanced. The absorption and refraction of the generated field are determined by the linear polarization of the medium at the generated frequency. The refraction has a very important effect upon the growth of the field by determining whether or not the generated field remains in phase with the polarization which drives it. This phase-matching condition is essential and dictates (at resonance where $\text{Re}[\chi^{(1)}] = 0$) that we also require $\Delta k = 0$ for efficient growth of the field. For a generated field on resonance both absorption and dispersion can be problematic. In general $\Delta k$ will be finite resulting in a limited length over which the field grows before slipping out of phase with the driving polarization $l_{coh} = 1/\Delta k$. But in the presence of EIT, and with the elimination of the linear susceptibility to vacuum values, the result is no absorption and no refraction due to the resonant level and only non-resonant transitions to other levels contribute to the wave-vector mismatch.

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![Figure 17](image-url)
is enhanced by proximity to resonance since this term is subject to constructive interference between the pair of dressed states. This mechanism is relevant to both CW and pulsed lasers fields (Harris et al., 1990; Zhang et al., 1993).

(ii) Here a pair of strong fields are applied resonantly in a Lambda configured three-level system near to both Raman and single photon resonance (figure 17(ii)). The presence of EIT on both fields eliminates absorption and dispersion in their propagation. They can then propagate without loss or distortion in what would otherwise be an extremely optically dense gas. This enables a dark-state with maximum amplitude and equally phased atomic coherence to be formed along the entire propagation length. Four-wave mixing from the maximal coherence leads to large up-conversion efficiency. This requires the use of fields strong enough to ensure adiabatic evolution of the dark state so is restricted to high power pulsed lasers or to Doppler free CW laser experiments (Harris and Jain, 1997; Merriam et al., 2000, 1999).

(iii) If two very strong fields are applied close to Raman resonance they will still lead to the formation of a dark-state maximal coherence even if they are detuned far from single photon resonance (detuning $\Delta$) (figure 17(iii)) providing that they are strong enough so that the condition $\Omega_1\Omega_2 > \Delta^2\gamma_1$ is satisfied (where $\gamma_1$ is the dephasing rate of the $|1\rangle - |2\rangle$ coherence). In this case EIT plays no role in so far as absorption is concerned as the fields are far from resonance. Nevertheless the choice of the correct magnitude of Raman detuning can lead to cancellation of the material refraction for the pair of applied fields (Harris et al., 1997). The maximal coherence, however, greatly boosts the conversion efficiency in four-wave mixing (Hakuta et al., 1997) as for (b). If the maximal coherence is excited between molecular vibrational or rotational states the medium is very suited to high-order Raman side-band generation (Sokolov et al., 2000). The requirement of high power to achieve adiabatic evolution restricts this to pulsed lasers.

(iv) The third field may also be close to resonant with another transition in this case the generated field completes a double-$\Lambda$ scheme which can enhance the mixing further. An important variant of the double-$\Lambda$ scheme with 3 applied fields is that explored by Hemmer et al. (1995) and by Zibrov and co-workers where only two fields are injected (Zibrov et al., 1999) (figure 17(iv)). These fields are applied close to single photon resonance with transitions in each of the two $\Lambda$ systems formed in alkali metal atoms. This scheme with CW laser fields has remarkable properties including, if the applied fields are co-propagating, the growth of both pairs of fields, and if the applied fields are counter-propagating mirrorless oscillation as experimentally demonstrated by Zibrov et al. (1999).

The advantages of all of these schemes lies in the reduction of unwanted absorption and dispersion for the generated and the driving fields whilst the source term for new fields is boosted. As a consequence of the elimination of drive field dispersion and absorption large values of coherence can be driven in the case of (ii), (iii) and (iv), in fact to the maximal value (i.e. $\rho_{12} = 0.5$), in all the atoms (molecules) within the interaction region. The frequency mixing process then proceeds with the atomic coherence acting as the local oscillator with which the final drive field beats to produce a generated field with a high overall conversion efficiency.

In addition to the different four-wave mixing schemes discussed in the remainder of this section other exciting applications of EIT to nonlinear optics have been proposed and experimentally investigated. One recent development is the demonstration of high efficiency multi-order Raman side-band generation through the modulation of optical fields in a molecular medium prepared in a maximally coherent state of the $v=0$ and $v=1$ vibrational states (Hakuta et al., 1997; Sokolov et al., 2000). The possibility of using the broad-bandwidth spectrum of phase coherent side-bands to synthesize ultra-short pulses was first predicted (Harris and Sokolov, 1997) and then demonstrated by the Stanford group (Sokolov et al., 2001). Another interesting effect happens when in an EIT medium the light velocity matches the speed of sound. As shown by Matsko and collaborators a new type of stimulated Brillouin scattering should occur in this case which in contrast to the usual situation allows efficient forward scattering (Matsko et al., 2001b).

B. Non-linear Mixing and Frequency Up-Conversion with Electromagnetically Induced Transparency

The enhancement of four-wave mixing in a two-photon resonant scheme of the type illustrated in Fig. 17i was first treated by Harris et al. (1990). The idea of improving four-wave mixing using an additional control field primarily to adjust the refractive index of the medium was treated by Tewari and Agarwal (1986). In their scheme, however, the control field is not incorporated directly into the mixing fields and there is no constructive interference of the non-linear susceptibility.

Taking the linear and non-linear susceptibilities derived in section III we can proceed with an analysis of the case of four-wave mixing in a Lambda EIT scheme. Proceeding from Eq. (71) and integrating with appropriate boundary conditions (as is usual in all four-wave mixing processes) leads to an expression for the generated intensity of the form:

$$I_4 = \frac{3n\omega^2}{8Z_o c^2} |\chi^{(3)}|^2 |E_1|^2 |E_2|^2 |E_3|^2 F(\Delta k, z, R, I),$$

(72)

where $z$ is the propagation distance, $Z_o = 376.7\Omega$ is the impedance of free space, $n$ is the refractive index at the generated frequency (typically close to unity), $R$ and $I$ are shorthand for $\text{Re}\chi^{(1)}$ and $\text{Im}\chi^{(1)}$ respectively and $c$ is the speed of light in vacuum. Here we have introduced the phase-matching factor $F(\Delta k, z, R, I)$ that depends
upon the details of the mixing scheme and the focal geometry. This general result indicates the importance of the phase-matching (and hence $\Delta k$) upon the generated field intensity.

To obtain clear insight into the effects of EIT we will follow the solution provided by Petch et al. (1996). For the case of plane-waves propagating in a homogeneous medium this treatment yields:

\[
I_4 = \frac{3n\omega^2}{8Z_o c^2} \left| \chi^{(3)} \right|^2 \left| E_1 \right|^2 \left| E_4 \right|^2 \left| E_3 \right|^2 \times \frac{1 + e^{-\frac{\Delta k}{2\alpha}} - 2e^{-\frac{\Delta k}{2\alpha}} \cos[(\Delta k + \frac{\omega R}{2c})z]}{\left[ \frac{\alpha z}{2} \right]^2 + \left[ \Delta k + \frac{\omega R}{2c} \right]^2}. \quad (73)
\]

It is implicit that $\chi^{(3)}$, Re[$\chi^{(1)}$] and Im[$\chi^{(1)}$] are the laser dressed expressions introduced in section III with any required inclusion of collisions and Doppler integration having been carried out. In the limit of a large propagation length $z$ the term in the numerator will go to unity and if also $\Delta k = 0$ the intensity is simply related to the susceptibilities by:

\[
I_4 \sim \left| \frac{\chi^{(3)}}{\chi^{(1)}} \right|^2 \quad (74)
\]

as was pointed out first by Harris et al. (1990) and subsequently (in the form used here) by Petch et al. (1996). It is very clear that EIT increases this ratio by both increasing the value of the numerator and reducing the denominator. It should be noted that a dramatic change in conversion efficiency as a function of coupling strength is only apparent in an inhomogeneously broadened medium. In a homogeneously broadened medium although EIT increases the efficiency, the ratio of the susceptibilities remains independent of the coupling strength.

In the case of four-wave mixing with EIT we usually assume a weak probe field so that excited state populations and coherences remain small (see figure 17(i)). The two-photon transition need not be strongly driven (i.e. a small two-photon Rabi frequency can be used) but a "strong" coupling laser is required. The coupling laser amplitude must be sufficient that the Rabi frequency is comparable to or exceeds the inhomogeneous spectral widths in the system (e.g. Doppler width). For example a laser intensity of above $1MWcm^{-2}$ is required for a typical Doppler broadened visible transition. This is trivially achieved even for unfocused pulsed lasers, but does present a substantial barrier for the use of continuous wave (CW) lasers unless a specific Doppler free configuration is employed. The latter is not normally suitable for a frequency up-conversion scheme if a large frequency up-conversion factor is to be achieved e.g. to the vacuum ultra-violet (VUV). This is in large part because atomic species with high ionisation potentials (> 10eV) that are suitable for frequency up-conversion to the VUV are not easily trapped and cooled at high density. Recent experiments, by for instance Hemmer, report significant progress in CW non-linear optical processes using EIT (Babin et al., 1996; Hemmer et al., 1995). A number of other possibilities e.g. laser cooled atoms and Doppler free geometries have also been explored.

In the case of frequency up-conversion to short wavelengths, Doppler shifts arising from the Maxwellian velocity distribution of the atoms or molecules in a gas at finite temperature lead to a corresponding distribution in the detunings for the atomic ensemble. The calculation of the response of the medium, as characterized by the susceptibilities, must therefore include the Doppler effect by performing a weighted sum over possible detunings. From this operation the effective value of the susceptibilities at a given frequency are obtained, and these quantities can be used to calculate the generated field. The interference effects persist in the dressed profiles provided the coupling laser Rabi frequency is comparable to or larger than the inhomogeneous width. This is because the Doppler profile follows a Gaussian distribution which falls off much faster in the wings of the profile than the Lorentzian profile arising from lifetime broadening. Indeed in the case of Doppler broadening the effects of increasing $\Omega_c$ manifest themselves most clearly. As discussed by Harris et al. (1990) there is expected to be a step-like increase in the conversion efficiency when the magnitude of $\Omega_c$ approaches the Doppler width of the transition. For coupling strengths below this value the atomic sample remains opaque as it would under ordinary resonant conditions; in contrast for a coupling strength exceeding the Doppler width the full benefit of the quantum interference is obtained.

In the Doppler broadened case therefore the coupling field Rabi frequency must remain greater than the Doppler width for a significant fraction of the interaction time. Pulsed lasers, because of the high peak powers that arise, are required. Moreover a transform limited single mode laser pulse is essential for the coupling laser fields which drive EIT since a multi-mode field will cause an additional dephasing effect on the coherence resulting in a deterioration of the quality of the interference, as discussed in Sec. III.

The work of the Stanford group has highlighted that when pulsed laser fields are used additional considerations must be made. Kasapi and co-workers showed (Kasapi et al., 1995) that the group velocity of a 20ns pulse can be modified for pulses propagating in the EIT; large reductions, e.g. by factors down to $< c/100$, in the group velocity have been observed. Another consideration beyond that found in the simple steady-state case is that the medium can only become transparent if the pulse contains enough energy to dress all of the atoms in the interaction volume. This preparation energy condition was discussed in section IV. This puts additional constraints on the laser pulse parameters.

Up-conversion to the UV and vacuum UV has been enhanced by EIT in a number of experiments. The first experiment to show EIT in a resonant scheme, where both the EIT effect on opacity and phase-matching were
important, was reported by Zhang et al. (1993). They employed a four-wave mixing scheme in Hydrogen (equivalent to figure 17(ii)), where the EIT was created on the 3p-1s transition at 103nm in a Lambda scheme by the application of a field at 656nm on the 2s-3p transition (figure 18). A field at 103nm is generated by the four-wave mixing process that was enhanced by this EIT. In these experiments the opening of photoionization channels from both of the involved excited atomic states leads to loss of the conditions for perfect transparency. Nevertheless because the photoionization cross-section for the 2s metastable level is about one order of magnitude less than that from the 3p state, the effect of EIT is only partially reduced. It is in general important to keep the photoionization rate sufficiently small so as to not to quench the coherence through electron impact broadening (see Buffa et al. (2003)). In this experiment, and subsequent work by the same authors, conversion efficiencies up to 2 \times 10^{-4} were reported (Zhang et al., 1995).

![Ionization Region](image)

**FIG. 18** Experimental demonstration of sum-frequency generation in H from (Zhang et al., 1993). *left*: Level and coupling scheme. *right*: Calculated (dashed) and measured (full line) nonlinear susceptibility as function of detuning from 3p - 1s transition for different density length products of the medium \((nL)\), only under the least dense condition is the Autler-Townes split structure apparent in the generated signal, at higher density strong resonant enhancement is seen (Zhang et al., 1993).

A limit to the conversion in an EIT enhanced four-wave mixing scheme is set by the Doppler width. A large Doppler width dictates that a comparable Rabi frequency is required to create good transparency, this in turn leads to a reduction in the non-linear susceptibility. This is one of the most important factors limiting the conversion efficiency in the hydrogen scheme (Hakuta, unpublished). The requirements on a minimum value of \(\Omega_c > \Delta_{Doppler}^{\text{constrains}}\) the conversion efficiency that can be achieved because of a scaling factor by \(1/\Omega_c^2\) that ultimately leads to diminished values of \(\chi^{(3)}\). The use of gases of higher atomic weight at low temperatures is therefore highly desirable in any experiment utilising EIT for enhancement of four-wave mixing to the VUV. A combination of the low mass of the H atom and the elevated temperatures required in the dissociation process leads to especially large Doppler widths (> 10 GHz). To overcome such a severe limit investigations were undertaken by one of the authors in krypton gas where the relevant Doppler width is only 1 GHz. Conversion efficiencies approaching \(10^{-2}\) were obtained (Dorman et al., 2000b). In this case, the conversion efficiency appears to be limited by the finite value of \(\Delta k\) due to the other atomic levels rather than absorption in the medium (Dorman et al., 2000a).

In a Doppler free medium the effect of the constructive interference for the non-linear term is most apparent. In a system without inhomogeneous broadening perfect transparency can be induced with \(\Omega_c \ll \gamma_{31}\). As \(\Omega_c\) is small relative to \(\gamma_{31}\) the non-linear susceptibility will, because of the constructive interference, have a value essentially identical to that of the unmodified resonant atomic system. The transparency dip will be very narrow (see Figure 7). The spectral widths of these features are typically sub-natural and are therefore accompanied by very steep normal dispersion, which corresponds to a much reduced group velocity. As was pointed out by several authors including Schmidt and Imamoglu (1996), and Harris and Hau (1999), the non-linear susceptibilities in this case can be extremely large as there is a constructive interference. Non-linear optics at very low light levels, i.e. at the few-photon limit, is possible in this regime. Associated with their first dramatic measurements of ultra-slow light Hau et al. (1999) reported a non-linear refractive index in an ultra-cold Na vapour that was 0.18\(cm^2/W^{-1}\), this is already 10^6 times larger than that measured for cold Cs atoms in a non-EIT system, which itself was much greater than the non-linearity in a typical solid state system. The discussion of this process in the few photon limit is given in section VI.

### C. Non-linear Optics with Maximal Coherence

Harris demonstrated an important extension of the EIT concept that occurs if the atomic medium is strongly driven by a pair of fields in Raman resonance in a 3-level system. Due to the elimination of absorption and refraction, both fields can propagate into the medium and large values of the material coherence are created on the non-driven transition. Considering the system illustrated in figure 17 (ii) we can imagine that both applied fields are now strong. Under appropriate adiabatic conditions (see section III) the system evolves in the dark-state to produce the maximum possible value for the coherence \(\rho_{12} = 0.5\). Adiabatic evolution into the maximally coherent state is achieved by adjusting either the Raman detuning or the pulse sequence (i.e. to "counter-intuitive" order). The pair of fields may also be in single photon resonance with a third level, in which case the EIT-like elimination of absorption will be important. This situation is equivalent to the formation of a dark-state, since neither of the two strong fields is absorbed by the medium. For
sufficiently strong fields the single-photon resonance condition need not be satisfied and a maximum coherence can still be achieved (see figure 17(iii)) provided that the condition $\Omega_1\Omega_2 > |\Delta|^2$ is met, where $\Delta$ is the detuning of the strong fields from state $|3\rangle$.

Under the conditions of STIRAP, or for matched pulse propagation, large populations of coherent population trapped states are created. To achieve this situation the laser electric field strength must be large enough to create couplings that will ensure adiabatic atomic evolution, and laser pulses that are sufficiently energetic to prepare all of the atoms in the beam path. The $\rho_{12}$ coherence thus created will have a magnitude $|\rho_{12}| = 0.5$ (i.e. all the atoms are in a coherent state) and negative sign (i.e. all the atoms are in the population trapped dark-state). The complex coherence varies in space and time and can be written:

$$
\rho_{12} = -\frac{\Omega_1\Omega_2^*}{\sqrt{\Omega_1^2 + \Omega_2^2}} \exp\left\{i(\omega_1 - \omega_2)t - (k_1 - k_2)z\right\}
= |\rho_{12}| e^{-i(k_1 - k_2)z} e^{i(\omega_1 - \omega_2)t}.
$$

(75)

Under these circumstances mixing of additional fields with the atom can become extremely efficient (Harris et al., 1997).

The preparation energy condition for the creation of a maximal coherence requires that the pulse energy must exceed the following value:

$$
E_{\text{prep}} = \frac{f_{11}}{f_{23}} \rho_{21} A L \hbar \omega,
$$

(76)

where $f_{ij}$ are the oscillators strengths of the transitions and $\rho_{21}$ the product of the density and the length (Harris and Luo, 1995). Essentially the number of photons in the pulse must exceed the number of atoms in the interaction volume to ensure all atoms are in the appropriate dressed state. It should be noted that the preparation energy is much smaller for non-linear conversion processes not using maximal coherence. Furthermore this preparation energy is not lost by dissipation if the pulses evolve adiabatically and can be recovered.

An additional field applied to the medium can participate in sum- or difference frequency mixing with the two Raman resonant fields. The importance of the large value of coherence is that it is the source polarization that drives the new fields generated in the frequency mixing process. This is described by the following equation,

$$
\frac{dE_3}{dz} = -i\hbar \omega_4 \frac{\rho_{21}}{4} \left[ (a_4 \rho_{11} + d_4 \rho_{22}) E_1 + b_4 \rho_{12} E_2 \right],
$$

(77)

with some coefficients $a_4, b_4$ and $d_4$ that depend upon atomic factors. The second term on the right hand side ($b_4 \rho_{12} E_3$), the source of the new field, is comparable in magnitude to the first term that describes the dispersion and loss. Complete conversion can occur over a short distance if the factor $\rho_{12}$ is large, this significantly relaxes the constraints usually set by phase-matching in non-linear optics. Recently near unity conversion efficiencies to the far-UV were reported in an atomic lead system where maximum coherence had been created (Harris et al., 1997; Jain et al., 1996; Merriam et al., 2000, 1999).

An example of high conversion efficiency four-wave mixing is shown in figure 19 (after Jain et al. (1996)).

![FIG. 19 Parametric up-conversion with maximum coherence from (Harris et al., 1997): A pair of (near) resonant coupling ($\Omega_i$) and probe pulses ($\Omega_p$) as shown in (a) generate maximum coherence between states $|1\rangle$ and $|2\rangle$. The coherently prepared medium then leads to efficient up-conversion of second coupling pulse $\Omega_c$ into $\Omega_b$ (b). Also shown is the conversion efficiency from 425 nm to 293 nm as function of coupling laser intensity (after Jain et al. (1996)).](image)

An alternative way to look at the origin of the large four-wave mixing efficiency is to consider the susceptibilities. Recalling the density matrix results of a A system, eq.(12), one recognizes that the polarization at $\omega_4$ proportional to $\rho_{21}$ is a sum of two terms, the first being proportional to the linear susceptibility and the second to the non-linear susceptibility. The drive fields $E_1$ and $E_2$ are now implicitly included in $\rho_{12}$ that has taken the maximal value of 0.5. These drive fields are resonant so the structure of $\chi^{(3)}$ simplifies to a single non-resonant term in the denominator. The non-linear susceptibility therefore shares this single non-resonant term in the denominator with the linear susceptibility and so these two quantities are of comparable size. This is a very unusual situation in non-linear optics and the length over which efficient frequency conversion can occur is now reduced to values comparable to the coherence length. A consequence of this is that complete non-linear frequency conversion of the fields can occur in a distance of the order of the coherence length (determined by the real part of the susceptibility). This is equivalent to having near vacuum conditions for the refraction (and absorption) of the medium whilst the non-linearity remains large.

In a molecular medium large coherence between vibrational or rotational levels have also been achieved using adiabatic pulse pairs in gas phase hydrogen and deuterium (Sokolov et al., 2000). Efficient multi-order Raman side-band generation has been observed to occur. In this case a pair of lasers that are slightly detuned from Raman resonance are used to adiabatically establish a superposition of two molecular states. The large energy separation to the next excited electronic state in these molecules (10 eV) means that the situation is similar to...
that illustrated in figure 17(iii). This superposition then mixes with the applied fields to form a broad spectrum of sidebands through multi-order mixing. Molecular coherence in solid hydrogen has recently also been used to eliminate phase-mismatch in a Stokes or anti-Stokes stimulated Raman frequency-conversion scheme (Hakuta et al., 1997). In this scheme, $v=0$ and $v=1$ vibrational states of the hydrogen molecule electronic ground state form the lower states of a Raman scheme. Since the $|1\rangle - |2\rangle$ dephasing rate is very small, in appropriately prepared samples of solid hydrogen, interference that causes the dispersion to become negligible can occur. Because of the removal of the usual phase-mismatch, efficient operation of these frequency conversion schemes over a broad range of frequencies (infra-red to vacuum UV) has been shown.

A recent prediction is of broadband spectral generation associated with strong field refractive index control (Harris and Sokolov, 1997; Kien et al., 1999). The observations of very efficient high order Raman side-band generation lead the way to synthesizing very short duration light pulses since the broad-band Raman side-band spectrum has been proved to be phase-locked (Sokolov et al., 2001). It is anticipated that high power sources of sub-femtosecond duration pulses might be achieved through this technique.

D. Four-Wave Mixing in Double-$\Lambda$ Systems

Four-wave mixing with all the optical fields close to resonance is achieved in atoms with 4-levels in the so-called double-Lambda configuration (Hemmer et al., 1995), which is detailed in Fig. 20. The fully resonant character of the light-matter interaction for all of the fields (both those applied and those generated) involved in four-wave mixing leads potentially to very high conversion efficiencies and to a number of important characteristics. Earlier work on double-Lambda schemes in which all four fields are applied has shown for instance that the formation of dark-states depends upon the relative phase of the fields (Arimondo, 1996; Buckle et al., 1986). Experiments in the CW limit have been carried out in sodium vapour that show the phase-sensitivity of EIT in a double-Lambda scheme (Korsunsky et al., 1999).

First we will consider the case where three resonant fields are applied such that the fourth field is generated in resonance between the highest of the excited states and the ground state. Under appropriate conditions the presence of the three applied fields eliminates not only the resonant absorption/refraction of the generated field but also for the applied fields themselves. The situation of four-wave mixing in a double-lambda has been treated theoretically in detail by Korsunsky and Kosachov (1999), where the relative phases of the four fields were found to evolve with propagation so as to support the lossless propagation in a fashion similar to that predicted by Harris for a pair of fields propagating in resonance in a Lambda medium (Harris, 1993). The phase and the amplitudes of all four fields, the generated field and the applied fields, are predicted to adjust as they propagate so as to support efficient transfer of energy between the fields, under loss-less conditions.

An experimental realization of the up-conversion in a double-Lambda scheme driven by three resonantly tuned pulsed fields with the new field generated in the far-UV at 186nm was achieved by Merriam et al. (2000) in lead vapour. High conversion efficiencies, more than 0.3 from the shortest wavelength applied field at 233nm to the 186nm field, were indeed observed. The experiments confirm that the double-Lambda scheme supports loss-less propagation provided that the pairs of Lambda resonant fields have matched ratios of Rabi frequency. For pulses not initially satisfying the condition of matched Rabi frequencies absorptive loss and non-linear energy transfer occur until the condition is reached after which all fields propagate without further loss and refraction. Limits to the up-converted power density were found, however, to arise from power broadening.

An interesting alternative to the schemes just described for efficient non-linear optical conversion in a Double-Lambda scheme was identified by Zibrov et al. (1999). In essence the scheme is very simple, two fields are applied resonantly one in each of the two Lambda systems. These then efficiently couple to the pair of fields that complete the double-Lambda configuration. The generated fields correspond to Stokes and anti-Stokes components of the two applied fields. In the original experiment in Rb vapour the applied fields counter-propagate and the resulting non-linear gain and efficient intrinsic feedback in this configuration lead to mirror-less self oscillation at extremely low applied field strengths (at the $\mu$W level of CW power). Further analysis of the quantum dynamics in this situation has highlighted the possibility of using this technique to generate narrow-band sources of non-classical radiation (Lukin et al., 1999).
VI. EIT WITH FEW PHOTONS

In the absence of a strongly polarizable medium, photons are essentially non-interacting particles. In communication technology this is a desired property and optics has emerged over the last two decades as the preferred method for communicating information. For the same reason, attempts to use light in information processing tasks such as computation has failed. For the latter, one needs strong, dissipation free light-light interactions. In this section we summarize and discuss some of the potentials and limitations of electromagnetically induced transparency in this respect.

A. Giant Kerr effect

Information processing based on light requires nonlinear interactions that would lead e.g. to a Kerr effect, or equivalently, a cross-phase modulation where the phase of a light field is modified by an amount determined by the intensity of another optical field. If the nonlinear phase shifts arising from such a Kerr effect were on the order of \( \pi \)-radians, it would be possible to implement all-optical switching. Such large Kerr nonlinearities without appreciable absorption can however, only be obtained for intense laser pulses containing roughly \( 10^{10} \) photons.

One of the principal challenges of nonlinear optics is the observation of a mutual phase shift exceeding \( \pi \)-radians using two light fields each containing a single order Kerr susceptibility and small linear susceptibility at two-photon resonance (\( \Delta = 0 \)). This so-called "giant Kerr" effect, proposed by Schmidt and Imamoglu (1996) can be understood using the four-level system depicted in Fig. 21: the principal element here is once again the Lambda subsystem consisting of states \( |1\rangle \), \( |2\rangle \), and \( |3\rangle \). The state \( |4\rangle \) has the same parity as \( |3\rangle \) and has an electric-dipole coupling to \( |2\rangle \); the signal field at frequency \( \omega_s \) is applied on this transition, with a Rabi frequency \( \Omega_s \). The cross-phase modulation nonlinearity

![Figure 21: Level scheme for giant Kerr effect.](image)

To calculate the third order susceptibility corresponding to the cross-phase modulation Kerr nonlinearity, we assume both \( \Omega_p \) and \( \Omega_c \) to be perturbative. As we have discussed in Sec. III, we can use the effective non-Hermitian Hamiltonian

\[
H_{\text{eff}} = \frac{\hbar}{2} \left[ \Omega_p \sigma_{31} + \Omega_c \sigma_{32} + \Omega_s \sigma_{42} + h.c. \right] + \hbar \left( \Delta - \frac{i}{2} \Gamma_3 \right) \sigma_{33} + \hbar \left( \delta - \frac{i}{2} \Gamma_2 \right) \sigma_{22} + \hbar \left( \Delta \omega_{42} - \frac{i}{2} \Gamma_4 \right) \sigma_{44}
\]

(78)

to describe the atomic dynamics in this limit. Here, we have introduced the detuning \( \Delta \omega_{42} = \omega_{42} - \omega_s \) and the spontaneous emission rate \( \Gamma_4 \) out of state \( |4\rangle \). Once again, we can solve the Schrödinger equation with the Hamiltonian of Eq. (78) to obtain the probability amplitude for finding the atom in state \( |3\rangle \) \( (a_3(t)) \) and use it to evolve the linear and nonlinear components of the polarization at \( \omega_p \):

\[
P(t) = N \mu_{13} a_3(t) e^{-i\omega_p t} + h.c.
\]

\[
= \epsilon_0 \chi^{(1)}_{\omega_p} \left( -\omega_p, -\omega_p \right) E_p e^{-i\omega_p t} + \epsilon_0 \chi^{(3)}_{\omega_p} \left( -\omega_p, \omega_s - \omega_p, -\omega_s \right) |E_s|^2 E_p e^{-i\omega_p t} + h.c.
\]

(79)

Similarly, we can obtain the linear and nonlinear susceptibilities at \( \omega_s \). For simplicity, we consider only the resonant case where \( \Delta = \delta = 0 \).

First we emphasize that the linear susceptibility for both probe and signal fields vanish in the limit \( \Gamma_2 = 0 \). This is trivially true for \( \omega_s \) since it is assumed not to couple to state \( |1\rangle \) atoms, and is the case for \( \omega_p \) to the extent that \( \omega_s \) remains perturbative. We also note that self-phase modulation type Kerr nonlinearities \( \chi^{(3)}_{\omega_p} \left( -\omega_p, -\omega_p, -\omega_p \right) \) and \( \chi^{(3)}_{\omega_p} \left( -\omega_p, \omega_s - \omega_p, -\omega_s \right) \) for both fields are identically zero. The cross-phase modulation nonlinearity \( \chi^{(3)}_{\omega_{pm}} = \chi^{(3)}_{\omega_p, \omega_s, -\omega_s} \) =
\[ \chi^{(3)}(\omega_s, \omega_p, -\omega_p) \] on the other hand is given by (Schmidt and Imamoglu, 1996)

\[ \chi^{(3)}_{\text{xpm}} = \frac{|\mu_{13}|^2|\mu_{24}|^2 \varrho}{2\epsilon_0 h^3 \Omega_c^2} \left( \frac{1}{\Delta \omega_{12}} + i \frac{\Gamma_4}{2 \Delta \omega_{12}^2} \right). \] (80)

To evaluate the significance of this result, it is perhaps illustrative to compare it to the Kerr nonlinear susceptibility of a standard three-level atomic system \( \chi^{(3)}_{\text{3-level}} \) depicted in Figure 22a: for this system consisting of the ground state \( |a \rangle \) and an intermediate state of opposite parity \( |b \rangle \) and a final state \( |c \rangle \), we find

\[ \text{Re}[\chi^{(3)}_{\text{3-level}}] = \frac{|\mu_{ab}|^2|\mu_{bc}|^2 \varrho}{2\epsilon_0 h^3 \Delta \omega_{ab} \Delta \omega_{bc}}. \] (81)

where \( \mu_{ij} \) and \( \Delta \omega_{ij} \) denote the dipole matrix element of and the detuning from the transition \( |i \rangle \rightarrow |j \rangle \), respectively \( (i, j = a, b, c) \). When we compare the two expressions, we observe that they have practically identical forms: in the EIT-xpm scheme \( 1/(4\Delta \omega_{ab}^2) \) is replaced by \( 1/\Omega_c^2 \). Even though one needs to have \( \Delta \omega_{ab}^2 \geq \Omega_c^2 \) in a three-level scheme to avoid absorption, \( \Omega_c^2 \ll \Gamma_4^2 \) is possible in the EIT-xpm system. This in turn implies that the magnitude of Kerr nonlinearity can be orders of magnitude larger in the latter case: this is the essence of the giant Kerr effect (Schmidt and Imamoglu, 1996).

Figure 22: Level configuration for cross-phase modulation in ordinary three-level scheme (left) and dressed-state representation of giant-Kerr scheme (right).

We can understand the origin of this enhancement using the dressed-state picture depicted in Figure 22b: when we apply the dressed-state transformation introduced in Sec. III, we find that the initial and final states (which remain unchanged) are coupled via two intermediate states \( |2d \rangle \) and \( |3d \rangle \). The two paths contributing to virtual excitation of state \( |4 \rangle \) experience a constructive interference for \( \omega_p \) tuned in between the two dressed-states, partially leading to the above mentioned enhancement. Perhaps more importantly however, the “effective detuning” between the intermediate dressed-states is given by \( \Omega_c \) and can be much smaller than their width.

Alternatively, we can understand the predicted enhancement of the Kerr nonlinearity by recalling the steep dispersion obtained at transparency for \( \Omega_c^2 \ll \Gamma_4^2 \): in this limit, small changes in two-photon detuning \( \delta \) that are caused for example by a shift in the energy of state \( |2 \rangle \) can give rise to drastic increase in Re\[ \chi^{(1)} \] experienced by the probe field. We can understand the role of the signal field as creating an ac-Stark shift of state \( |2 \rangle \), thereby leading to a large change in the index of refraction at \( \omega_p \) that is proportional to \( \Omega_c^2 \).

Finally, we note that by optical pumping and by choosing the polarization of the lasers appropriately, the energy level diagram depicted in Fig. 21 can be realized practically for all alkali atoms. It has been estimated that the enhancement of \( \chi^{(3)}_{\text{xpm}} \) over a standard three-level atomic medium with identical density and dipole matrix elements could be as much as six orders of magnitude (Schmidt and Imamoglu, 1996). Naturally, the interest in this system arises from the possibility of doing nonlinear optics on the single photon level, without requiring a high-finesse cavity; this possibility was already discussed in the original proposal (Schmidt and Imamoglu, 1996), but in the limit of continuous-wave fields. Recent experiments have already demonstrated the enhancement of Kerr nonlinearities in the limit of \( \Delta \omega_{ab}^2 \ll \Omega_c^2 \) (Braje et al., 2003); this is the purely dissipative limit of \( \chi^{(3)}_{\text{xpm}} \) and was discussed by Harris and Yamamoto in the context of a two-photon absorber (Harris and Yamamoto, 1998).

As we have already discussed above, the steep dispersion curve of the probe field is responsible for the enhancement in the Kerr effect. It is interesting that it is this steep dispersion, or the slow group velocity that arises from it, that at the same time gives rise to a stringent limit on the available single-photon nonlinear phase shifts for pulsed fields. This limitation arises from the fact that the probe and signal pulses travel at vastly different group velocities in the limit \( \Omega_c^2 \ll \Gamma_4^2 \) and their interaction time will be limited by the fact that the slow probe pulse will separate spatially from the fast signal pulse (Harris and Hau, 1999).

To make quantitative predictions on the limitation on nonlinear phase shifts arising from disparate group velocities, we assume a nearly ideal EIT scheme where \( \Gamma_3 \Gamma_2 < \Omega_c^2 < \Gamma_3^2 \), \( \Delta = 0 \), and concentrate on the limit of small \( |\delta| \). For this system, we have a probe field group velocity

\[ v_p = \frac{\Omega_c^2}{\Gamma_3} \frac{1}{\varrho \sigma_{13}}, \] (82)

where \( \sigma_{13} = 3 \lambda_p^2/2\pi \) is the peak absorption cross-section of the \( |1 \rangle \rightarrow |3 \rangle \) transition (\( \lambda_p = 2\pi c/\omega_p \)). Given that \( v_p \ll c \), the time it takes for the probe pulse to traverse the medium

\[ \tau_d = \frac{\Gamma_3}{\Omega_c^2} \varrho \sigma_{13} L, \] (83)
referred to as the group delay time (Harris and Hau, 1999; Harris and Yamamoto, 1998), can be much longer than the traversal time of the signal field $T_s = L/c$ and the pulse-widths of the probe ($\tau_p$) and signal ($\tau_s = \tau_f$) fields. As discussed earlier, the pioneering experiment of Hau et al. (1999) already demonstrated $\tau_D > 1$ msec $\gg \tau_p$.

The generated nonlinear phase shift is proportional to the interaction length of the probe and signal fields. In the EIT giant Kerr scheme, this is determined by $\min[L, L_h]$, where $L_h$ is the probe propagation length that would give rise to a time delay between the two pulses that is equal to their pulse-width $\tau_p$; i.e.

$$L_h = v_p \tau_p = \frac{\Omega_s^2}{\Gamma_4} \frac{1}{\theta \sigma_{13}} \tau_p,$$

which was originally defined as the Hau-length (Harris and Hau, 1999). The nonlinear phase shift can be determined from the slowly varying envelope equation (Harris and Hau, 1999)

$$\frac{\partial E_p}{\partial z} + \left( \frac{1}{v_p} + \frac{1}{c} \right) \frac{\partial E_p}{\partial t} = i \frac{\pi}{\sum p} \chi^{(3)} \left[ E_s \left( t - \frac{z}{c} \right) \right]^2 E_p$$

$$= \left( \frac{i}{4(\Delta \omega_{42} + i \Gamma_{44}/2) v_p} \right) \left| \Omega_s \left( t - \frac{z}{c} \right) \right|^2 E_p,$$

where we have used Eq. (80).

When the group delay is negligible compared to $\tau_p$ (i.e. $L \ll L_h$), and we assume Gaussian input pulses, we find for the peak (complex) phase shift for the maximum of the pulse

$$\phi_{xpm} \rightarrow \frac{\Gamma_4}{\Delta \omega_{42} + i \Gamma_{44}/2} \frac{n_s \sigma_{24}}{A} \frac{\ln 2}{\pi} \frac{\tau_d}{\tau_p},$$

where $n_s$ is the number of photons in the pulse and $A$ its cross section area. Eq. (86) is, as expected, the result one would obtain using Eq. (80) without taking into account the group velocity mismatch between the two pulses. In the opposite limit of $L \gg L_h$, we have

$$\phi_{xpm} \rightarrow \frac{\Gamma_4}{\Delta \omega_{42} + i \Gamma_{44}/2} \frac{n_s \sigma_{24}}{A} \frac{\ln 2}{\pi} \frac{\tau_d}{\tau_p},$$

This result shows that the maximum nonlinear phase shift that can be obtained in the EIT system is independent of the system parameters (such as $\Omega_c, \varrho, L$) (Harris and Hau, 1999), and is on the order of 0.1 radians for single photon-pulses focused into an area $A \sim \sigma_{24}$. This remarkably simple result assumes that we take $\Delta \omega_{42} \sim \Gamma_{44}/2$, in which case we get a two-photon absorption coefficient that is comparable to the nonlinear phase shift.

### B. Cross-phase modulation using single-photon pulses with matched group velocities

From the perspective of applications in quantum information processing, the result obtained in the previous subsection has a somewhat negative message: while EIT allows for weak optical pulses containing $n \sim 10$ photons to induce large nonlinear phase shifts, it cannot provide strong dissipation free interactions between two single-photon pulses. The limitation, as the presented analysis has revealed, is due to the disparate group velocities of the two interacting pulses. The natural question to ask then is whether one can obtain larger nonlinear phase shifts by assuring that both probe and signal pulses travel at the same ultra-slow group velocity.

Before proceeding with the analysis, we emphasize that it is possible to consider an atomic medium with two coherently driven atomic species, each establishing EIT at different frequencies. If the two atomic species are different isotopes of the same element, then the frequencies at which the medium becomes transparent can differ by an amount that is on the order of the hyperfine splitting. We can then envision a scenario where two different light pulses, centered at the two transparency frequencies, travel with the identical ultra-slow group velocity through the medium containing an optically dense mixture of these two isotopes (A and B) of the same element. By adjusting the external magnetic field, it is also possible to ensure that one of these light pulses that sees EIT induced by species B will also be (nearly) resonant with the $|2\rangle \rightarrow |4\rangle$ transition of species A. By choosing this pulse to be the signal pulse, one can realize giant Kerr interaction between two ultra-slow light pulses (Lukin and Imamoglu, 2000). An alternative scheme based on only a single atomic species was recently proposed by Petrosyans and Kurizki (Petrosyan and Kurizki, 2002).

Even though the analysis of nonlinear optical processes using single-photon pulses requires a full quantum field theoretical approach, we first consider the classical limit following up on the analysis previously presented for a fast signal pulse. In the limit of a slow signal pulse with group velocity $v_s$, we obtain

$$\phi_{xpm} = \frac{\Gamma_4}{\Delta \omega_{42} + i \Gamma_{44}/2} \frac{n_s \sigma_{24}}{A} \frac{\ln 2}{\pi} \frac{\tau_d}{\tau_p},$$

which is identical in form to that given in Eq. 86. In contrast to that earlier result however, the expression in Eq. 88 is valid even when $\tau_d \gg \tau_p$ (Lukin and Imamoglu, 2000).

To determine the maximum possible phase shift, we recall that for an optically dense medium the EIT transmission peak has a width given by $\Delta \omega_{trans} = \Omega^2_s/(\theta \sigma_{13} L)$. If we set $\tau_p = \Delta \omega_{trans}^{-1}$, we find

$$\frac{\tau_d}{\tau_p} \leq \sqrt{\theta \sigma_{13} L}.$$

Since the other factors on the right hand side of Eq. 88 can be of order unity, we can obtain $\phi_{xpm} \geq \pi$, provided that we have $\theta \sigma_{13} L \geq 10$: presence of an optically thick medium is essential for large photon-photon interaction. At the same time, we note that this interaction is necessarily slow and has a $\phi_{xpm}$-bandwidth product that is
interaction of two single-photon pulses. To this end, we replace the classical electric fields describing the signal and probe fields with operators \( \hat{E}_s(z,t) = \hat{E}_s^{(+)}(z,t) + \hat{E}_s^{(-)}(z,t) \), where

\[
\hat{E}_s^{(+)}(z,t) = \sum_k \frac{\hbar c}{2\omega_0 V} \hat{a}_{kl}(z,t) e^{ik(z-ct)}.
\]

Here \( i = p, s \) and \( V \) denotes the quantization volume. We suppress the polarization index for convenience. In order to eliminate dissipation and simplify the Heisenberg equations for the quantized fields, we assume \( \Gamma_2 = 0 \) and \( |\Delta\omega_{42}| \gg \Gamma_4 \). To ensure however that we can adiabatically eliminate atomic bandwidths of freedom, we also need to impose a finite bandwidth \( \Delta_q \) to the quantized field: with this assumption, the equal space-time commutation relations explicitly depend on this bandwidth (Lukin and Imamoglu, 2000). Finally, to guarantee negligible pulse spreading, we take \( \tau_p^{-1} < \Delta_q \ll \Delta\omega_{\text{trans}} \) and arrive at a pair of operator equations for \( \hat{E}_p(z,t) \) and \( \hat{E}_s(z,t) \) that have the identical form as the one given in Eq. (85), provided we replace \( c \) by \( v_p \) on the right-hand-side. These equations can then be solved to yield

\[
\hat{E}_s^{(+)}(z,t) = \hat{E}_s^{(+)}(0,t') \\
\times \exp \left[ \frac{\hbar \omega_1}{2} \frac{\tau_4}{\hbar^2} \hat{E}_s^{(-)}(0,t') \hat{E}_s^{(+)}(0,t') \right],
\]

\[
\hat{E}_s^{(+)}(z,t) = \hat{E}_s^{(+)}(0,t') \\
\times \exp \left[ \frac{\hbar \omega_1}{2} \frac{\tau_4}{\hbar^2} \hat{E}_s^{(-)}(0,t') \hat{E}_s^{(+)}(0,t') \right],
\]

where \( t' = t - z/v_p \).

Using the expressions in Eq. (91), we can address the question of nonlinear phase shift obtained during the interaction of two single-photon pulses. To this end, we assume an input state of the form

\[
|1_i\rangle = \sum_k \xi_k \hat{a}_{kl}^\dagger |0\rangle,
\]

for both fields, where we require \( \sum_k |\xi_k|^2 = 1 \) to ensure proper normalization. The spatio-temporal wavefunction of each pulse is given by the single-photon amplitude \( \Psi_i(z,t) = \langle 0 | \hat{E}_i^{(+)}(z,t) |1_i\rangle \), where \( i = s, p \). To evaluate the correlations induced by the interaction, we concentrate on the two-photon amplitude

\[
\Psi_{sp}(z,t;t',t') = \langle 0 | \hat{E}_s^{(+)}(z,t) \hat{E}_p^{(+)}(z',t') |1_s, 1_p\rangle.\]

One finds that the equal-time \( t = t' \) local \( z = z' \) correlations can be written as

\[
\Psi_{sp}(z,t;t,t) = \Psi_s(0,t) \Psi_p(0,t) e^{i\phi},
\]

where \( \phi = \tau_4 \Delta_q \sigma_24 \Gamma_4/(4\pi A\Delta\omega_{42}) \). Since the equal space and time commutation relations are proportional to \( \Delta_q \), there is explicit dependence on this quantity. The nonlinear phase shift \( \phi \) is the counterpart of the classical phase \( \phi_{\text{exp}} \) obtained earlier in Eq. (88); the equivalence is obtained by replacing \( \tau_p^{-1} \) with the quantization bandwidth \( \Delta_q \) (Lukin and Imamoglu, 2000).

A large single-photon nonlinear phase shift \( \phi \) that exceeds \( \pi \) radians can be used to implement two-qubit quantum logic gates (Nielsen and Chuang, 2000). However, we have seen that creating large phase shifts between single photon pulses also changes the mode profile of the signal (probe) pulse, conditioned on the presence of the probe (signal) pulse. Therefore, we expect entanglement between the internal degrees of freedom of the pulses, such as photon number or polarization, and the external degrees of freedom, such as mode profile determined by \( \{\xi_k\} \), invalidating the simplistic assumption of ideal single-photon switching using large Kerr nonlinearity.

### C. Few-photon four-wave mixing

Besides the resonantly enhanced Kerr effect discussed in the previous subsections, nonlinear interactions on the few-photon level have been predicted and analyzed in more detail in the four-wave mixing schemes shown in figure 23. In the first scheme discussed by Harris and Hau (1999) photons from two pulses \( (\omega_1) \) and \( (\omega_3) \) are converted into an electromagnetic field pulse at \( \omega_4 \) in the presence of a strong monochromatic drive field at \( \omega_2 \) (figure 23(a)). In the second scheme (figure 23(b)) discussed in (Johnsson et al., 2002) for co-propagating pump fields and in (Fleischhauer and Lukin, 2000; Lukin et al., 1999) for counter-propagating pump fields, the field \( \omega_2 \) is initially absent and spontaneously generated along with \( \omega_4 \).

\[\begin{array}{c}
\text{(a)}
\end{array}\]

\[\begin{array}{c}
\text{(b)}
\end{array}\]

FIG. 23 Four-wave mixing schemes for the generation of one field of frequency \( \omega_4 \) (a), and for the generation of two field \( \omega_2 \) and \( \omega_4 \) (b).

In the first scheme, see Fig. 23(a), the input pulse \( \omega_1 \) experiences a group delay due to EIT on the \( |1\rangle \rightarrow |3\rangle \) transition, while the second one, \( \omega_3 \), moves almost with the vacuum speed of light. Harris and Hau showed within an undepleted pump approximation, that the conversion efficiency, i.e. the ratio of the number of generated photons at \( \omega_4 \) to the number of input photons at \( \omega_1 \) is given by a dimensionless function \( \Phi(q,\kappa L) \) multiplied by the
number of input photons per atomic cross section.

\[
\frac{n_{\text{out}}}{n_{\text{in}}} = \Phi(\eta, \kappa L) \frac{\eta_{24}^{\text{in}}}{A_3 \sigma_{24}} \tag{95}
\]

Here \( \kappa \) is the (E-field) absorption coefficient at the \( |1\) – \( |4 \) transition and \( L \) the medium length. \( \eta = L/L_b \) is the ratio of the length \( L_b \), introduced in Eq. (84), i.e. the probe propagation length corresponding to a time delay equal to the pulse width, to the medium length \( L \). In the limit of an optically thin medium \( \kappa L \rightarrow 0 \) and of a small group delay within the medium \( \eta \rightarrow 0 \), the usual result of perturbative long-pulse nonlinear optics is obtained, which for Gaussian pulses reads

\[
\Phi(\eta, \kappa L) = \left[ \frac{\ln(2)}{\pi} \right]^{1/2} \frac{T_a}{\sqrt{T_a^2 + T_b^2}} \eta \kappa L. \tag{96}
\]

Here the \( T_a \)'s denote the temporal length of the pulses. In the limit of \( \eta \rightarrow \infty \), i.e. if the group velocity reduction of \( \omega_1 \) is large, such that the \( \omega_2 \) pulse sweeps over it very rapidly, the function is given by

\[
\Phi(\eta, \kappa L) = \left[ \frac{\ln(2)}{\pi} \right]^{1/2} \frac{\kappa L}{\eta} \exp(-2\kappa L) \tag{97}
\]

i.e. falls off with \( \eta^{-1} \). A maximum of \( \Phi \) of about 0.074 is attained when \( \eta = \kappa L = 1 \), i.e. when the pulse delay is just one pulse length and if the medium has an optical thickness of one. In this case about 10 photons per atomic cross section are sufficient to obtain perfect conversion.

In the second scheme, where the field \( \omega_2 \) is generated, see Figure 23(b), a finite detuning from level \( |3 \) is needed to suppress absorption. In order to cancel ac-Stark shifts the frequencies \( \omega_1 \) and \( \omega_2 \) can be tuned midway between two states with appropriate transition matrix elements (Johnsson et al., 2002). Efficient frequency conversion (Babin et al., 1999; Ham et al., 1997b; Hemmer et al., 1994; Hinze et al., 1999), generation of squeezing (Lukin et al., 1999), as well as the possibility of mirrorless oscillations (in counter-propagating geometry) have been predicted (Lukin et al., 1998b) and in part experimentally observed (Zibrov et al., 1999). It has been shown in (Fleischhauer and Lukin, 2000) that an extremely low input photon flux \( \phi_{\text{thr}} \) is sufficient to reach the threshold of mirrorless oscillations

\[
\phi_{\text{thr}} = f N \gamma_{12} \tag{98}
\]

where \( N \) is the number of atoms in the beam path and \( \gamma_{12} \) is the coherence decay rate of the \( |1\) – \( |2 \) transition. \( f \) is a numerical pre-factor of order unity. Johnsson et al. have shown that in the adiabatic limit and for \( \gamma_{12} = 0 \) an effective interaction hamiltonian of the four-wave mixing process can be derived which reads for a one-dimensional model (Johnsson and Fleischhauer, 2002)

\[
H_{\text{int}} = \frac{\hbar g c}{2 \Delta} \int dz \frac{\tilde{\Omega}_1^{\text{in}} \tilde{\Omega}_3^{\text{in}} \tilde{\Omega}_2^{\text{in}} \tilde{\Omega}_4^{\text{in}} + \text{h.c.}}{\tilde{\Omega}_1^{\text{in}} \tilde{\Omega}_3^{\text{in}} + \tilde{\Omega}_2^{\text{in}} \tilde{\Omega}_4^{\text{in}}}, \tag{99}
\]

where \( \tilde{\Omega}_i \) are the operators corresponding to the (complex) Rabi-frequencies of the fields and a common coupling constant \( g = \mu \omega /2\hbar \) of all transitions was assumed. One recognizes that in contrast to the case of usual non-resonant nonlinear optics, the sum of the intensities of the resonant fields appears in the denominator. As a consequence in a co-propagating geometry, the conversion length, i.e. the length after which a pair of photons in the input fields \( \omega_1 \) and \( \omega_3 \) is completely converted into a pair of photons in the fields \( \omega_2 \) and \( \omega_4 \) decreases with decreasing input intensity. Thus weak input fields lead to a faster conversion than stronger ones.

Finally, we note that although there had been a number of theoretical proposals on EIT-based quantum nonlinear optics, it is only since very recently that there have been first experimental demonstrations of the predicted effects. Kuzmich and coworkers (Kuzmich et al., 2003) and independently van der Wal and collaborators (van der Wal et al., 2003) have demonstrated quantum correlations between Stokes and anti-Stokes photons generated with a controllable time delay by resonant Raman scattering on atomic ensembles. The intermediate storage and subsequent retrieval of correlated photons in atomic ensembles are essential ingredients of the proposal of Duan and co-workers for a quantum repeater (Duan et al., 2001), which is an important tool for long-distance quantum communication.

D. Few-photon cavity-EIT

It is well known in quantum optics that presence of high-finesse cavities can be used to enhance photon-photon interactions for example for the purpose of quantum computation (Niehse and Chuang, 2000). It is therefore natural to consider few-photon EIT inside a cavity, i.e. when the probe or the coupling field is replaced by a single quantized radiation mode. We will show in the present subsection that electromagnetically induced transparency combined with cavity quantum electrodynamics can lead to a number of interesting linear and nonlinear effects.

Let us first discuss linear phenomena associated with intracavity EIT. Consider a single three-level atom placed inside a resonator where the quantized resonator mode acts as the coupling field between the internal states \( |2 \) and \( |3 \). As shown by Field (1993) electromagnetically induced transparency on the probe transition is possible already with few resonator photons or even vacuum, provided that the atom and the resonator are strongly coupled. To see this we note that the fully quantum interaction Hamiltonian

\[
H_{\text{int}} = -\hbar \frac{\Omega_p}{2} |3\rangle \langle 1| - \hbar \frac{g}{2} \hat{a}^\dagger |3\rangle \langle 2| + \text{h.c.} \tag{100}
\]

separates into effective three-level systems

\[
|1, n\rangle \leftrightarrow |2, n+1\rangle \tag{101}
\]
where $1, 2, 3$ denote the internal state of the atom and $n$ and $n+1$ the number of photons in the resonator mode. If $g$, which characterizes the atom - cavity coupling, is sufficiently large and the atom is initially in state $|1\rangle$, EIT can be achieved for the probe field even for $n = 0$. Eq.(101) suggests another interesting application: The effective three-level systems have dark states. For example

$$|D\rangle = \cos \theta(t)|1, 0\rangle - \sin \theta(t)|2, 1\rangle$$  \hspace{1cm} (102)

with $\tan \theta(t) = \Omega_g(t)/g$. Thus stimulated Raman adiabatic passage from state $|1, 0\rangle$ to state $|2, 1\rangle$ induced by the probe field offers the possibility of a controlled generation of a single cavity photon. The potential usefulness of this effect in cavity QED has first been pointed out by Parkins et al. (Parkins et al., 1993). Law and Eberly proposed an application for the generation of a single photon in a well-defined wavepacket (Law and Eberly, 1996). Recently such a "photon pistol" was experimentally realized (Henrich et al., 2000; Kuhn et al., 2002).

The process of transferring excitation from atoms to a field mode is reversible and allows the opposite process of mapping cavity-mode fields onto atomic ground-state coherences. In this case the resonator mode takes on the role of the probe field, i.e.

$$H_{\text{int}} = -\hbar \frac{g^2}{2} \hat{a}^\dagger |3\rangle \langle 1| - \hbar \frac{\Omega_c}{2} |3\rangle \langle 2| + \text{h.c.}$$  \hspace{1cm} (103)

and we have the coupling

$$ |1, n\rangle \xrightarrow{g} |3, n - 1\rangle \xrightarrow{\Omega_c(t)} |2, n - 1\rangle $$  \hspace{1cm} (104)

The dark state for $n = 0$ is identical to that given in Eq. (102), except $\tan \theta(t) = g/\Omega_c(t)$ in the present case. The mapping provides a possibility for measuring cavity fields (Parkins et al., 1995). It was shown furthermore by Pellizzari et al. (1995) that a quantum-state transfer from one atom to a second atom via a shared cavity mode can be used to implement a quantum gate (Pellizzari et al., 1995). Finally Cirac et al. (Cirac et al., 1997) proposed an application to transfer quantum information between atoms at spatially separated nodes of a network.

All of the above discussed effects require however the strong coupling of the single atom to the resonator mode. This experimentally challenging requirement can partly be alleviated if an optically thick ensemble of three level atoms is used. When an ensemble of $N$ three-level atoms interacts with a single resonator (probe) mode and a classical (coupling) field according to

$$H_{\text{int}} = -\frac{\hbar}{2} \sum_{j=1}^{N} [g \hat{a} \hat{a}^\dagger |j\rangle \langle j+1| + h \Omega_c |j\rangle \langle j+1| + \text{h.c.}] $$  \hspace{1cm} (105)

only symmetric collective states are coupled to the initial state $|1, 1, ... 1\rangle$, where all $N$ atoms are in the ground state $|1\rangle$:

$$|1^N\rangle \equiv |1...1\rangle, \hspace{1cm} (106)$$

$$|1^{N-1}2\rangle \equiv \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |1...2, j...1\rangle, \hspace{1cm} (107)$$

$$|1^{N-2}2^2\rangle \equiv \frac{1}{\sqrt{2N(N-1)}} \sum_{i \neq j=1}^{N} |1...2, j...2, i...1\rangle, \hspace{1cm} (108)$$

etc.

If the initial photon number is $n = 1$ again a simple three-level coupling scheme emerges

$$|1^N, 1\rangle \xrightarrow{g\sqrt{N}} |1^{N-1}3, 0\rangle \xrightarrow{\Omega_c(t)} |1^{N-1}2, 0\rangle. \hspace{1cm} (109)$$

Due to the symmetric interaction of all $N$ atomic dipoles with the resonator mode, the coupling constant is however collectively enhanced

$$g \rightarrow g \sqrt{N}. \hspace{1cm} (110)$$

Thus controlled dark-state Raman adiabatic passage from a state with a single photon in the resonator to a single collective excitation and vice versa is possible without the requirement of a strong coupling limit of cavity-QED, defined here as $g^2 \gg \kappa \Gamma$, where $\kappa$ is the cavity decay rate. For higher photon numbers more complicated coupling schemes emerge. All of them do possess however dark states which allow a parallel transfer of arbitrary photon states with $n \ll N$ to collective excitations, which has important applications for quantum memories for photons and quantum networking (Lukin et al., 2000b).

As discussed in previous subsections, EIT can be used to achieve a resonant enhancement of nonlinear interactions leading e.g. to a cross-phase modulation. Large phase shifts produced by the interaction of individual photons are very appealing for their potential use in quantum information processing. Imamoğlu et al. (1997) showed that if a medium with sufficiently large Kerr nonlinearity is put into an optical cavity, it can lead to a photon blockade effect. When a photon enters a previously empty cavity, the induced refractive index detunes the cavity resonance. If this frequency shift is larger than the cavity linewidth a second photon cannot enter and will be reflected. This can be employed e.g. to build a controllable single-photon source or a controlled-phase gate (Duan and Kimble, 2003). Imamoğlu et al. suggested the use of a large ensemble of atoms in the resonator to enhance the photon blockade effect. It was later shown (Gheri et al., 1999) however, that the large dispersion of the EIT medium does not allow for a collective enhancement of the nonlinear atom-cavity coupling. While photon blockade can be achieved using either single atom or multi-atom EIT systems (Werner and Imamoğlu, 1999), the strong coupling limit of cavity-QED appears to be required in both cases. It must be emphasized however that in contrast to a many-atom cavity-EIT medium, a collection of $N \gg 1$ two-level atoms in a cavity is a nearly ideal linear system, exhibiting vanishingly small photon blockade effect. The survival of the nonlinearity in the $N \gg 1$
atom cavity-EIT is related to the reduction of the cavity-linewidth, or equivalently the width of the atom-cavity dark state (Lukin et al., 1998a; Werner and Imamoglu, 1999).

VII. SUMMARY AND PERSPECTIVES

EIT has undoubtedly made a mark upon optical science. Hopefully we have succeeded in explaining the relationship between EIT and the earlier related ideas of coherent preparation of atoms by fields and especially linked it with the concept of the dark-state. We stress again the distinct feature of EIT and related phenomena, in contrast to the earlier spectroscopic tools such as coherent population trapping, is that they occur in media that are optically dense and cause not only a modification of the excitation state of the matter but also significant changes to the optical fields themselves. It is this latter point that is crucial to understanding the importance of EIT in optics. EIT provides a new means to change the optical characteristics of matter, for instance the degree of absorption or refraction or the group velocity, and so provides a way to alter the propagation of optical fields and to enhance the generation of new fields.

At the time of writing (2004) we are some fourteen years on from the first experimental demonstration of the phenomenon of EIT. Much research activity has been undertaken in the period since this discovery to understand the phenomenon and its various implications. More importantly there has been a concerted effort in many laboratories to harness the effect for applications. It is informative to review the extent to which this field has grown rapidly in these years, and has so remained a healthy and active subject for the last decade with no sign of declining activity. There are constantly new and interesting proposals and demonstrations that extend EIT to fresh applications. It has become a standard tool in the kit for researchers studying the optical properties of atomic and molecular gases. For these reasons of durability and utility we believe that EIT has earned a significant place in optical physics that make it a valuable topic of study for new research students and experienced researchers alike.

We have tried in this review to cover the main ideas behind the application of EIT and related dark-state phenomena to optics. It is inevitable in a work such as this that there have been omissions. Much work has indeed been published in this field, especially in the theoretical area, and it was simply not possible to cover or cite every single contribution. We humbly apologize to anyone we have failed to cite. Our intention was primarily to provide the reader new to the field with an accessible and comprehensive framework that will support them when they make further more detailed explorations of this topic. We hope that in this we have succeeded and that we have provided a useful introduction to the theoretical ideas that underpin the subject.

Where possible we have tried to examine the results of real experiments. Again we have been selective choosing the most convenient illustrative examples rather than attempting a comprehensive review of all work. The details of the experiments discussed here are unfortunately rather sparse due to the constraints of space. Indeed we have not explored in any structured way the experimental techniques that are required to achieve robust EIT and thus to investigate these phenomena. This omission is regrettable, but comes about since there are in fact many types of situation where EIT can be observed, e.g. in dense gases with transform limited high power pulsed lasers, with CW lasers interacting with vapsours using Doppler free geometries or indeed with CW lasers in cold gases. It is thus not practicable to cover so many diverse experimental situations and to understand the types of experiments the reader must consult the cited literature and where necessary other reviews and textbooks to obtain a fuller understanding of the required experimental methods.

One feature of the work in this field is that the vast majority of investigations have been carried out in the gas phase. This trend runs somewhat contrary to a general tendency for non-linear optics to move towards employing solid-state media. The reason for this is that gas phase media still offer the physicist unique features; small homogeneous and inhomogeneous linewidths compared to most solid state media, transparency over much of the IR-VUV range, possibilities to laser cool and high thresholds against optical damage. Moreover a problem normally identified for gas phase non-linear optical media, the low value of the non-linear susceptibility compared to many solid-density optical crystals, is circumvented by EIT since it permits the physicists to use resonance to hugely enhance the susceptibility.

Having reviewed the subject and examined a number of the applications that have already been thoroughly investigated we now turn to examine the future. It is of course a rather risky undertaking to make any predictions about what new applications are likely to emerge and likely as not we will be wrong about much of what follows. Nevertheless we feel it is of use to other scientists in this field at least to offer some thoughts on this subject in the hope of stimulating further ideas. For safety we will confine ourselves to areas where there is at least some current activity and so some evidence upon which to base our extrapolations.

The potential of EIT and maximal coherence to generate high brightness coherent fields in the short wavelength range remains only partially explored. EIT has been shown to enhance frequency conversion efficiencies to the 0.01 level in up-conversion to the 100nm frequency range. By careful choice of the atomic system and through judicious optimisation of the density and length of the medium and the use of transform-limited lasers to drive all the fields it may be possible to improve the conversion efficiency by a substantial further factor. This could result in the generation of transform limited nano-second pulses (spectral linewidths of \(\sim 100\text{MHz}\))
of VUV radiation with energies of 10’s of microjoules. These could be of utility in a number of applications in non-linear spectroscopy, for instance if combined with a second longer wavelength tunable laser to excite two-photon transitions to highly excited states, or perhaps in Raman spectroscopy. The fixed frequency nature of the output would appear to preclude a wider range of applications.

Whilst EIT-type four-wave mixing enhancement may be of limited utility since it results in a fixed frequency output this is not, in principle, a limit for off resonant schemes with maximal coherence. The high conversion efficiencies already demonstrated in the far-UV may be extended into the VUV. There is a problem in the Raman like excitations since there is a limit to the degree of up-conversion as the initial excitation is via difference frequency mixing of the applied fields. There is therefore an inherent limit set at around 170nm for the shortest wavelength that can be generated due to the energy level structure of the atoms and the shortest available wavelengths from lasers. For shorter wavelength extensions an important problem that must be solved will be how to excite maximal coherence between the ground-state and a very highly excited upper state. This should use sum-frequency excitation by the applied lasers. The creation of maximal coherence in this case cannot use the conventional STIRAP type adiabatic excitation schemes. Recently a promising alternative has been demonstrated, Stark Chirped Rapid Adiabatic Passage (SCRAP), with the potential for efficient excitation of maximal coherence between the ground-state and a very highly excited state of an atom (Rickes et al., 2000). A recent first demonstration of the use of this technique showed a degree of enhancement to XUV generation via third-harmonic generation (Rickes et al., 2003), but further work is needed in this area.

Besides the potential for efficient generation of coherent radiation in new frequency domains, resonant nonlinear optics based on EIT will be of growing importance for controlled nonlinear interactions at the few photon level. The above mentioned limitations concerning tunability and accessible frequencies are of no relevance here. Photons are ideal carriers of quantum information, yet processing of this information e.g. in a quantum computer is quite difficult. The reason for this is the weakness of photon-photon interactions in usual nonlinear media. Here EIT may offer new directions. Although some theoretical proposals exist, the full potential of EIT based nonlinear optics for these applications is still not explored. In particular on the experimental side little has been done in this direction.

Another feature of EIT, which makes it very attractive for future applications in quantum information is the possibility to transfer coherence and quantum states from light to collective atomic spin excitations. Quantum memories for photons (Fleischhauer and Lukin, 2002; Lukin et al., 2000b) as discussed in Sec. III are just one potential avenue, quantum networks based on them including quantum repeaters are another (Kuzmich et al., 2003; van der Wal et al., 2003). Combining the state mapping techniques with atom-atom interactions in mesoscopic samples may provide new tools to generate photon wave-packets with tailored quantum states or to manipulate these states. A first proposal in this direction using a dipole-blockade of Rydberg excitations was given in (Lukin et al., 2001). The controlled coupling of a photon to many atomic spins in EIT could be of interest for the preparation or the probing of entangled many particle states. We have just begun to explore the role of entanglement in many-body physics and quantum information. Laser manipulation of spin ensembles via EIT could provide a very useful tool in this quest.

EIT also offers new possibilities in linear and nonlinear matter-wave optics. As pointed out in Sec. IV slow light in atomic gases corresponds to a quasi-particle which is a mixed electromagnetic-matter excitation. For very low group velocities almost all excitation is concentrated in atomic spins while the propagation properties are still mostly determined by the electromagnetic part. Thus slow light in ultra-cold atomic gases provides a new way to control the propagation of matter waves with potential applications in matter-wave interferometry (Zimmer and Fleischhauer, 2004) or if atom-atom interactions are included in nonlinear matter-wave optics (Masalas and Fleischhauer, 2004).

Ultra-fast measurements have recently entered the attosecond domain (Drescher et al., 2002). The possibility of employing the highly efficient multi-order Raman generation that results from an adiabatically prepared maximal vibrational coherence has already been investigated. A key problem to solve before this can be widely employed for ultra-fast measurements will be the synchronisation of the train of sub-femtosecond pulses that are generated with external events. One possibility (suggested by A. Sokolov) is to use the pulse train to “measure” processes in the modulated molecules themselves so that the synchronisation is automatically satisfied.

As we have emphasized throughout this article, coherent preparation techniques that are at the heart of EIT can be easily implemented in optically dense atomic samples in the gas phase. It is generally argued however, that the range of its applications can be significantly enlarged if EIT is implemented in solid-state media. The obvious roadblock in this quest are the ultrashort coherence times of optical transitions in solids: even for transitions that are not dipole-allowed, the coherence times are typically in the sub-nanosecond timescales. Among the many different physical mechanisms contributing to this fast dephasing, perhaps the most fundamental one is the interaction of electrons with lattice vibrations (phonons). While cooling the samples to liquid helium temperatures reduces the average number of phonons and thereby phonon-induced-dephasing, the coherence times still remain significantly shorter than their atomic counterparts.

Fortunately, there is an exception to the general rule of ultrashort dephasing times in solids: electron spin
degrees-of-freedom in a large class of solid-state materials have relatively long coherence times, ranging from 1μsec in optically active direct-band-gap bulk semiconductors such as GaAs (Wolf et al., 2001), to 1msec in rare-earth ion doped (insulating) crystals (Kuznetsova et al., 2002). In both cases, the ratio of the coherence relaxation rates of the dipole-allowed and spin-flip transitions can be as large as 10^4, indicating that EIT can be implemented efficiently.

Most of the experimental efforts aimed at demonstrating EIT in solid-state spins have so far focused on rare-earth ion doped crystals (Ham et al., 1997a; Turukhin et al., 2002) and nitrogen-vacancy centers in diamond (Wei and Manson, 1999). In the experiments carried out using Pr doped Y_2SiO_5 at cryogenic temperatures (T= 5K), Turukhin and co-workers have observed group velocities as slow as 45 m/s and a group delay of 66μsec, for light pulses with a pulsewidth of 50μsec (Turukhin et al., 2002). Storage and retrieval of these pulses have also been demonstrated (Turukhin et al., 2002). While the ratio of the group delay to the pulsewidth in these experiments is on the order of unity, this is an impressive and promising development for solid-state EIT.

Optoelectronics and photonics technology is largely based on semiconductor heterostructures formed out of column III-V semiconductors. The conduction band electrons of these elements have predominantly s-type wavefunctions with small spin-orbit coupling. As a result, the spin coherence times are 4 orders of magnitude longer than the radiative recombination rate of excitons. The possibility of observing efficient EIT and slow light propagation in a doped GaAs quantum well in the quantum Hall regime has been discussed (Imamoglu, 2000). While this particular realization requires low temperatures and high magnetic fields (B ~ 10 Tesla), long spin coherence times in GaAs have been observed even at room temperature and with vanishing magnetic fields (Wolf et al., 2001). In fact, there is substantial activity in the emerging field of spintronics (Wolf et al., 2001) and it is plausible to argue that EIT can play a role by providing efficient photon-counter: after mapping the quantum state of a propagating light pulse onto collective hyperfine excitations of a trapped cold atomic gas, it is possible to monitor the resonance fluorescence induced by an additional laser field that only couples to the hyperfine excited state (Imamoglu, 2002; James and Kwiat, 2002). Even with a fluorescence collection/detection efficiency as low as 10%, it can then be possible to achieve photon counting with efficiency exceeding 99%. Such a device could be of great use in quantum optical information processing (Knill et al., 2001).

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