Electromagnetically induced transparency: Optics in coherent media

Michael Fleischhauer

Fachbereich Physik, Technische Universität Kaiserslautern, D-67663 Kaiserslautern, Germany

Atac Imamoglu

Institute of Quantum Electronics, ETH-Hönggerberg, HPT G12, CH-8093 Zürich, Switzerland

Jonathan P. Marangos

Quantum Optics and Laser Science Group, Blackett Laboratory, Imperial College, London SW7 2BW, United Kingdom

(Published 12 July 2005)

Coherent preparation by laser light of 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, which have placed gas-phase systems at the center of recent advances in the development of media with radically new optical properties. This article reviews these advances and the new possibilities they offer for nonlinear optics and quantum information science. As a basis for the theory of electromagnetically induced transparency the authors consider the atomic dynamics and the optical response of the medium to a continuous-wave laser. They then discuss pulse propagation and the adiabatic evolution of field-coupled states and show how coherently prepared media can be used to improve frequency conversion in nonlinear optical mixing experiments. The extension of these concepts to very weak optical fields in the few-photon limit is then examined. The review concludes with a discussion of future prospects and potential new applications.

CONTENTS

I. INTRODUCTION

Advances in optics have frequently arisen through the development of new materials with optimized optical properties. For instance, the introduction of new optical crystals in the 1970s and 1980s led to substantial increases in nonlinear optical conversion efficiencies to the ultraviolet (UV). Another example has been the development of periodically poled crystals that permit quasiphase matching in otherwise poorly phase-matched nonlinear optical processes, which greatly enhance nonlinear frequency-mixing efficiencies. As we shall see, coherent preparation is a new avenue that produces remarkable changes in the optical properties of a gas-phase atomic or molecular medium.

The cause of the modified optical response of an atomic medium in this case is the laser-induced coherence of atomic states, which leads to quantum interference between the excitation pathways that control the optical response. We can in this way eliminate the absorption and refraction (linear susceptibility) at the resonant frequency of a transition. This was termed electromagnetically induced transparency (EIT) by Harris and

co-workers (Harris *et al.*, 1990). The importance of EIT stems from the fact that it gives rise to greatly enhanced nonlinear susceptibility in the spectral region of induced transparency of the medium and is associated with steep dispersion. For some readable general accounts of earlier work in the field, see, for example, Harris (1997) or Scully (1992). Other more recent reviews on specific aspects of EIT and its applications can be found in the articles of Lukin, Hemmer, and Scully (2000); Matsko, Kocharovskaya, *et al.* (2001); Vitanov *et al.* (2001), as well as in the topical Colloquium by Lukin (2003). It should be emphasized that the modification of atomic properties due to quantum interference has been studied extensively for 25 years; see, for example, 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 in which 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 $\mathrm{Im}[\chi^{(1)}]$ determines the dissipation of the field by the atomic gas (absorption), while the real part $\mathrm{Re}[\chi^{(1)}]$ determines the refractive index. The form of $\mathrm{Im}[\chi^{(1)}]$ 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 $\mathrm{Re}[\chi^{(1)}]$ follows the familiar dispersion profile, with anomalous dispersion (decrease in $\mathrm{Re}[\chi^{(1)}]$ with field frequency) in the central part of the absorption profile within the linewidth. Figure 1 illustrates both the conventional form of $\chi^{(1)}$ and the modified form that results from EIT, as will be discussed shortly.

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 referred to Shore (1990).

For a two-level system, the result of coherent evolution is characterized by oscillatory population transfer (Rabi flopping). The generalization of this coherent situation to driven three-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 within the basis of either bare atomic states or new eigenstates, which diagonalize the complete atom-field interaction Hamiltonian. In both cases we shall see that interference between alternative excitation pathways between atomic states leads to modified optical response.

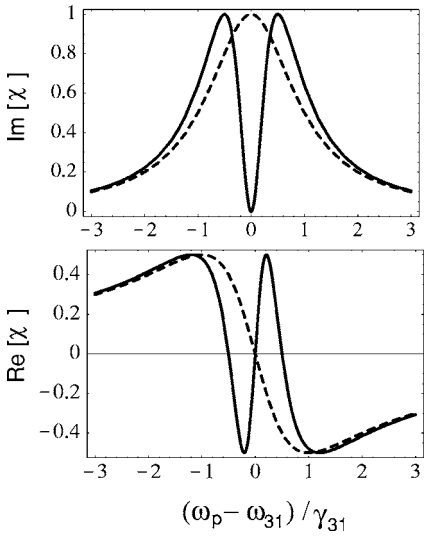

FIG. 1. Susceptibility as a function of the frequency ω_p of the applied field relative to the atomic resonance frequency ω_{31}, for a radiatively broadened two-level system with radiative width γ_{31} (dashed line) and an EIT system with resonant coupling field (solid line): top, imaginary part of $\chi^{(1)}$ characterizing absorption; bottom, real part of $\chi^{(1)}$ determining the refractive properties of the medium.

The linear and nonlinear susceptibilities of a Λ-type three-level system driven by a coherent coupling field will be derived in Sec. III. Figure 1 shows the imaginary and real parts 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 nonlinear susceptibility. Inspection of these frequency-dependent dressed susceptibilities reveals immediately several important features. One recognizes that $\mathrm{Im}[\chi^{(1)}]$ undergoes destructive interference

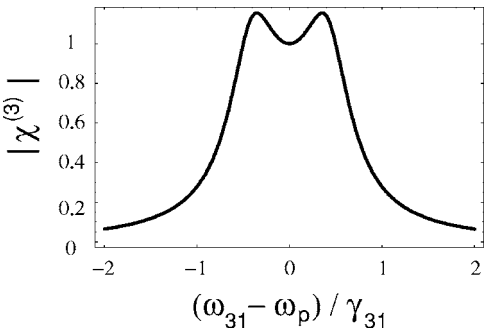

FIG. 2. Absolute value of nonlinear susceptibility for sum-frequency generation $|\chi^{(3)}|$ as a function of ω_p, in arbitrary units. The parameters are identical to those used in Fig. 1.

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 where there is a high degree of transmission the nonlinear 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 nonlinear effects. It is most significant that the refractive index passes through the vacuum value and the dispersion is steep and linear exactly where absorption is small. This gives rise to effects such as ultraslow group velocities, longitudinal pulse compression, and storage of light. Furthermore, through the destructive (constructive) interference in $\mathrm{Im}\chi^{(1)}$ (χ^3) and the elimination of the effect of resonance upon the refractive index, the conditions for efficient nonlinear mixing are met.

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 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, Hemmer, and Shahiar, 1997; Schmidt *et al.*, 1997; Zhao *et al.*, 1997; Wei and Manson, 1999; Serapiglia *et al.*, 2000). Atomic gases remain very important in optical applications for several key reasons; for example, they are often transparent in the vacuum UV and infrared, and there exist techniques to cool them to ultracold 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 cutoff 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 ushered in a renaissance in activity in the area of gas-phase nonlinear optics. New opportunities also arise due to the extremely spectrally narrow features that can result from coherent preparation; in particular, this gives ultralow group velocities and ultralarge nonlinearities. These are of potential importance to new techniques in optical information storage and also in nonlinear optics at the few-photon level, both of which may be important to quantum information processing.

Throughout this review we shall 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 nonlinear optical response due to electromagnetically induced transparency and related phenomena. We do not, however, cover the related topic of lasing without inversion (Kocharovskaya and Khanin, 1988; Gornyi *et al.*, 1989; Harris, 1989; Scully *et al.*, 1989) and the interested reader is advised to look elsewhere for reviews on that subject (Knight, 1990; Kocharovskaya, 1992; Mandel, 1993; Scully and Zubairy, 1997). In the following section we shall introduce the underlying physical concepts of EIT through the coupling of near-resonant laser fields with the states of a three-level system. In Sec. III we discuss in detail the dynamics in three-level atoms coupled to the applied laser fields and determine the optical response. In Sec. IV we treat the topic of pulse propagation in EIT and review the developments that have culminated in the demonstration of ultraslow group velocities down to a few meters per second and even the "stopping" and "storing" of a pulse of light. The utility of coherent preparation in enhancing the efficiency of frequency conversion processes is discussed in Sec. V. Then in Sec. VI we shall 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 future prospects in quantum optics and atom optics as a result of EIT.

II. PHYSICAL CONCEPTS UNDERLYING 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 photoionization of multielectron atoms. Due to the existence of doubly excited states the photoionization spectrum of multielectron 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 degenerate continuum states, with lifetimes in the picosecond to subpicosecond range. For the reason that they decay naturally to the continuum, these states are called *autoionizing states*. Fano introduced to atomic physics the concept of the interference between the excitation channels to the continuum that exist for a single autoionizing resonance coupled to a flat continuum [Fano, 1961; Fig. 3(a)]. In the vicinity of the autoionizing resonance the final continuum state can be reached via either direct excitation (channel 1) or through 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-

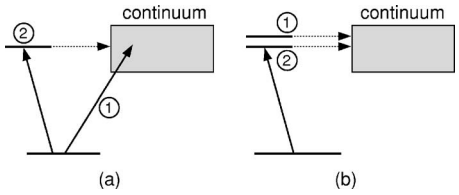

FIG. 3. Fano interferences of excitation channels into a continuum: (a) for a single autoionizing resonance; (b) for two autoionizing states.

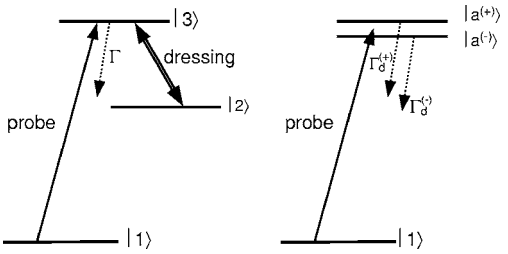

FIG. 4. Interference generated by coherent coupling: left, coherent coupling of a metastable state $|2\rangle$ to an excited state $|3\rangle$ by the dressing laser generates (right) interference of excitation pathways through the doublet of dressed states $|a^{\pm}\rangle$ (Autler-Townes doublet) provided the decay out of state $|2\rangle$ is negligible compared to that of state $|3\rangle$.

dependent suppression or enhancement in the photoionization cross section.

Experiments by Madden and Codling (1965) showed the resulting transparency windows in the autoionizing spectrum of helium. Sometime later the autoionizing 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).[1] In this wave-mixing experiment the absorption was eliminated in spectral regions where the nonlinear response remained large, hence an improved efficiency for frequency conversion was reported.

During the 1960s autoionizing 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 provided 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 [Fig. 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 (1990) showed how this situation could be used to enhance four-wave mixing. In experiments in zinc vapor that showed significantly increased nonlinear mixing, one of the fields was tuned to a transition between an excited bound state and a pair of closely spaced autoionizing 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 while leaving stimulated emission from the states unchanged. Although we shall 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) when it was realized 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 (Fig. 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 could 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 Fig. 1, while the nonlinear 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 vapor, pointed 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 (1989), in which 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 $|2\rangle$. The amplitude for $|3\rangle$, 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 $|1\rangle$-$|3\rangle$ pathway, or indirectly via the $|1\rangle$-$|3\rangle$-$|2\rangle$-$|3\rangle$ pathway (or 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 path-

[1]See the improved $\chi^{(3)}$ fit of Armstrong and Beers, 1975.

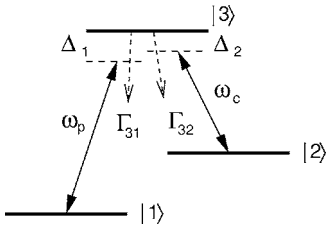

FIG. 5. Generic system for EIT: lambda-type scheme with probe field of frequency ω_p and coupling field of frequency ω_c. $\Delta_1=\omega_{31}-\omega_p$ and $\Delta_2=\omega_{32}-\omega_c$ denote field detunings from atomic resonances and Γ_{ik} radiative decay rates from state $|i\rangle$ to state $|k\rangle$.

way, but for resonant fields it is of opposite sign.

B. Dark state of the three-level Λ-type atom

The use in laser spectroscopy of externally applied electromagnetic fields to change the system Hamiltonian of course predates the idea of using this in nonlinear 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 1960s. Hänsch and Toschek (1970) recognized the existence of these types 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 nonlinear 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 three-level atom. We illustrate the three possible coupling schemes in Figs. 5 and 6. For consistency states are labeled so that the $|1\rangle$-$|2\rangle$ transition is always the dipole-forbidden transition. Of these prototype schemes we shall be most concerned with the lambda configuration in 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.

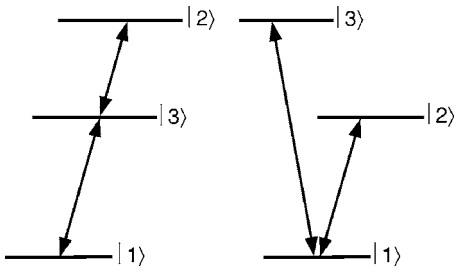

FIG. 6. Ladder (left) and vee-type (right) three-level schemes. These do not show EIT in the strict sense because of the absence of a (meta)stable dark state.

The physics underlying 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 shall therefore review briefly the concept of *dark states*. Alzetta *et al.* (1976) made the earliest observation of the phenomenon of *coherent population trapping* (CPT) followed shortly by theoretical studies of Whitley and Stroud (1976). Arimondo and Orriols (1976) and Gray *et al.* (1978) explained these observations using the notion of coherent population trapping in a dark eigenstate of a three-level lambda medium (see Fig. 5). In this process a pair of near-resonant fields are coupled to the lambda system and result in the Hamiltonian $H=H_0+H_{\rm int}$, where the Hamiltonian for the bare atom is H_0 and that for the interaction with the fields is $H_{\rm int}$. The Hamiltonian H has a new set of eigenstates when viewed in a proper rotating frame (see below), one of which has the form $|a^0\rangle=\alpha|1\rangle-\beta|2\rangle$, which contains no amplitude of the bare state $|3\rangle$ and has amplitudes α and β proportional to the fields such that it is 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 remained there since the excitation probability of this dark state is canceled via interference. An early account on the effect of CPT on the propagation of laser fields was given by Kocharovskaya and Khanin (1986). 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 eigenstates of a three-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 $H_{\rm int}=\boldsymbol{\mu}\cdot\boldsymbol{E}$ is often expressed in terms of the *Rabi coupling* (or Rabi frequency) $\Omega=\boldsymbol{\mu}\cdot\boldsymbol{E}_0/\hbar$, with $\boldsymbol{E}_0$ being the amplitude of the electric field $\boldsymbol{E}$, and $\boldsymbol{\mu}$ the transition electronic dipole moment. After introducing the rotating-wave approximation, we can represent the Hamiltonian of the three-level atom interacting with a coupling laser with real Rabi frequency Ω_c and a probe laser with Rabi frequency Ω_p (Fig. 5) in a rotating frame as

$$H_{\rm int}=-\frac{\hbar}{2}\begin{bmatrix}0 & 0 & \Omega_p\\ 0 & -2(\Delta_1-\Delta_2) & \Omega_c\\ \Omega_p & \Omega_c & -2\Delta_1\end{bmatrix}. \tag{1}$$

Here $\Delta_1=\omega_{31}-\omega_p$ and $\Delta_2=\omega_{32}-\omega_c$ are the detunings of the probe and coupling laser frequencies ω_p and ω_c from the corresponding atomic transitions.

A succinct way of expressing the eigenstates of the interaction Hamiltonian (1) is in terms of the "mixing angles" θ and ϕ 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 Fig. 5). For two-photon resonance ($\Delta_2=\Delta_1$, or $\delta=0$) the mixing angles are given by

$$\tan\theta=\frac{\Omega_p}{\Omega_c}, \tag{2}$$

$$\tan 2\phi=\frac{\sqrt{\Omega_p^2+\Omega_c^2}}{\Delta}. \tag{3}$$

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

$$|a^+\rangle=\sin\theta\sin\phi|1\rangle+\cos\phi|3\rangle+\cos\theta\sin\phi|2\rangle, \tag{4}$$

$$|a^0\rangle=\cos\theta|1\rangle-\sin\theta|2\rangle, \tag{5}$$

$$|a^-\rangle=\sin\theta\cos\phi|1\rangle-\sin\phi|3\rangle+\cos\theta\cos\phi|2\rangle. \tag{6}$$

The reader's attention is drawn to the following features: While 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 $\hbar\omega^\pm$,

$$\hbar\omega^\pm=\frac{\hbar}{2}(\Delta\pm\sqrt{\Delta^2+\Omega_p^2+\Omega_c^2}). \tag{7}$$

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 Fig. 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 counterintuitive pulse sequence. This was first demonstrated by Bergmann and co-workers (Kuklinski *et al.*, 1989; Gaubatz *et al.*, 1990), 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 ω_c is first applied. This is similar to the EIT situation since $\Omega_p\ll\Omega_c$ and the dark state indeed corresponds exactly to $|1\rangle$ with negligible population in either $|2\rangle$ or $|3\rangle$. But now Ω_p is gradually (i.e., adiabatically) increased and at the same time Ω_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 shall assume continuous-wave (cw) classical fields interacting with a single atom that can be modeled as a lambda system (Fig. 5). For the derivation of linear and nonlinear 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 nonperturbative field effects, a single-atom wave-function approach is simpler and more illustrative; we shall 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 shall 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 shall 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 nonzero electric dipole coupling to both $|1\rangle$ and $|2\rangle$. A (near) resonant nonperturbative electromagnetic field of frequency ω_c, termed the coupling field, is applied on the $|2\rangle$-$|3\rangle$ transition. A probe field of frequency ω_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

$$H_{\rm int} = -\frac{\hbar}{2}[\Omega_p(t)\hat{\sigma}_{31}e^{i\Delta_1 t} + \Omega_c(t)\hat{\sigma}_{32}e^{i\Delta_2 t} + \text{H.c.}], \tag{8}$$

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 laser-driven atomic systems are governed by the master equation for the atomic density operator:

$$\begin{aligned}\frac{d\rho}{dt} &= \frac{1}{i\hbar}[H_{\rm int},\rho] + \frac{\Gamma_{31}}{2}[2\hat{\sigma}_{13}\rho\hat{\sigma}_{31} - \hat{\sigma}_{33}\rho - \rho\hat{\sigma}_{33}] \\ &+ \frac{\Gamma_{32}}{2}[2\hat{\sigma}_{23}\rho\hat{\sigma}_{32} - \hat{\sigma}_{33}\rho - \rho\hat{\sigma}_{33}] \\ &+ \frac{\gamma_{2\rm deph}}{2}[2\hat{\sigma}_{22}\rho\hat{\sigma}_{22} - \hat{\sigma}_{22}\rho - \rho\hat{\sigma}_{22}] \\ &+ \frac{\gamma_{3\rm deph}}{2}[2\hat{\sigma}_{33}\rho\hat{\sigma}_{33} - \hat{\sigma}_{33}\rho - \rho\hat{\sigma}_{33}],\end{aligned} \tag{9}$$

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 Γ_{31} and Γ_{32}, respectively. We assume here that the thermal occupancy of the relevant radiation field modes is completely negligible. For optical frequencies this approximation is easily justified. A detailed derivation of this master equation is given by Cohen-Tannoudji *et al.* (1992). We have also introduced energy-conserving dephasing processes with rates $\gamma_{3\rm deph}$ and $\gamma_{2\rm deph}$; the latter determines one of the fundamental time scales for practical EIT systems, as we shall 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 $\gamma_{31}=\Gamma_3+\gamma_{3\rm deph}$, $\gamma_{32}=\Gamma_3+\gamma_{3\rm deph}+\gamma_{2\rm deph}$, and $\gamma_{21}=\gamma_{2\rm deph}$.

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

$$\begin{aligned}\vec{P}(t) &= -\sum_i \langle e\boldsymbol{r}_i\rangle/V \\ &= \frac{N_{\rm atom}}{V}[\boldsymbol{\mu}_{13}\rho_{31}e^{-i\omega_{31}t} + \boldsymbol{\mu}_{23}\rho_{32}e^{-i\omega_{32}t} + \text{c.c.}].\end{aligned} \tag{10}$$

To obtain the expression in the second line, we assumed that all $N_{\rm atom}$ 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 $\boldsymbol{\mu}_{13}=\mu_{13}\hat{\boldsymbol{z}}$ and $\boldsymbol{\mu}_{23}=\mu_{23}\hat{\boldsymbol{z}}$, we let $\varrho=N_{\rm atom}/V$ and obtain $P_z(t)=P(t)$ as

$$P(t) = \varrho[\mu_{13}\rho_{31}e^{-i\omega_{31}t} + \mu_{23}\rho_{32}e^{-i\omega_{32}t} + \text{c.c.}]. \tag{11}$$

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

$$\begin{aligned}\rho_{32} &= \frac{i\Omega_c e^{i\Delta_1 t}}{(\gamma_{32}+i2\Delta_2)}\rho_{12}, \\ \rho_{12} &= -\frac{i\Omega_c e^{i\Delta_2 t}}{\gamma_{21}+i2(\Delta_2-\Delta_1)}\rho_{13}, \\ \rho_{31} &= \frac{i\Omega_p e^{i\Delta_1 t}}{(\gamma_{31}+i2\Delta_1)} + \frac{i\Omega_c e^{i\Delta_2 t}}{(\gamma_{31}+i2\Delta_1)}\rho_{21}.\end{aligned} \tag{12}$$

As in Sec. I we define the single-photon detuning as $\Delta=\Delta_1=\omega_{31}-\omega_p$ and two-photon detuning as $\delta=\Delta_1-\Delta_2=\omega_{21}-(\omega_p-\omega-c)$. Keeping track of the terms that oscillate with $\exp[-i\omega_p t]$, we obtain

$$\begin{aligned}\chi^{(1)}(-\omega_p,\omega_p) = \frac{|\mu_{13}|^2\varrho}{\epsilon_0\hbar} &\times \left[\frac{4\delta(|\Omega_c|^2-4\delta\Delta)-4\Delta\gamma_{21}^2}{||\Omega_c|^2+(\gamma_{31}+i2\Delta)(\gamma_{21}+i2\delta)|^2}\right. \\ &\left. + i\frac{8\delta^2\gamma_{31}+2\gamma_{21}(|\Omega_c|^2+\gamma_{21}\gamma_{31})}{||\Omega_c|^2+(\gamma_{31}+i2\Delta)(\gamma_{21}+i2\delta)|^2}\right].\end{aligned} \tag{13}$$

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 nonperturbative 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 Fig. 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 $\text{Im}[\chi]$ over Δ is conserved as Ω_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 γ_{31}. In this latter case, it becomes apparent that the modifications in the linear susceptibility call for an explanation based on quantum interference phenomena

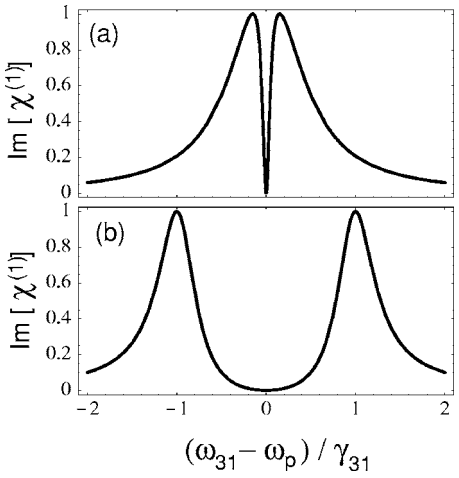

FIG. 7. EIT absorption spectrum for different values of coupling field and $\gamma_{21}=0$: (a) $\Omega_c=0.3\gamma_{31}$; (b) $\Omega_c=2\gamma_{31}$.

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 coworkers (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 $\gamma_{31}\sim 2\times 10^{11}\ \mathrm{s}^{-1}$. The decay rate of the lower state coherence $\gamma_{21}\sim 4\times 10^{9}\ \mathrm{s}^{-1}$ is determined by the collisional broadening for an atomic density of $\varrho\sim 5\times 10^{15}\ \mathrm{cm}^{-3}$.

The transparency obtained at two-photon resonance is independent of the detuning of the probe field from the bare $|1\rangle$-$|3\rangle$ transition (Δ). As Δ 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 become highly asymmetric for $\Delta > \gamma_{31}$. Figure 9 shows contour plots of $\mathrm{Im}[\chi^{(1)}]$ as a function of the detunings Δ_1 and Δ_2 of the two fields as well as the single-photon (Δ) and two-photon (δ) photon detunings. It is evident that for large detuning Δ_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 ultraslow group velocities for light pulses can be obtained in transparent media, which will be discussed extensively in the following section.

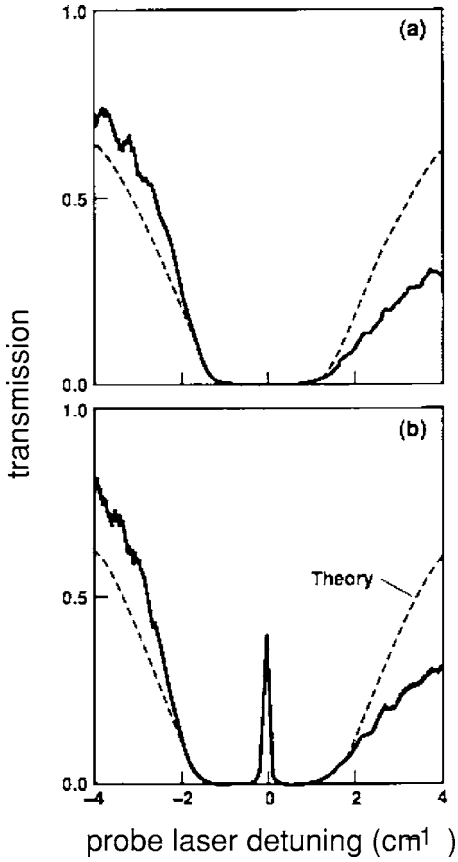

FIG. 8. First experimental demonstration of EIT in Sr vapor by Boller *et al.*, 1991: top, transmission through cell without coupling field; bottom, with coupling field on. From Boller *et al.*, 1991.

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 have been carried out using ultracold 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 nonideal effects.

In actual atomic systems, the dephasing rate of the forbidden $|1\rangle$-$|2\rangle$ transition is nonzero due to atomic collisions. All the important features of EIT remain observable even when $\gamma_{21}\neq 0$, provided that the coupling field Rabi frequency satisfies

$$|\Omega_c|^2 \gg \gamma_{31}\gamma_{21}. \tag{14}$$

Figure 10 shows the imaginary part of $\chi^{(1)}(-\omega_p,\omega_p)$ for $\gamma_{21}=0$ [Fig. 10(a)], $\gamma_{21}=0.1\gamma_{31}$ [Fig. 10(b)], and $\gamma_{21}=10\gamma_{31}$ [Fig. 10(c)]. In all plots, we take $\Omega_c=0.5\gamma_{31}$. We first note from Fig. 10(b) 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

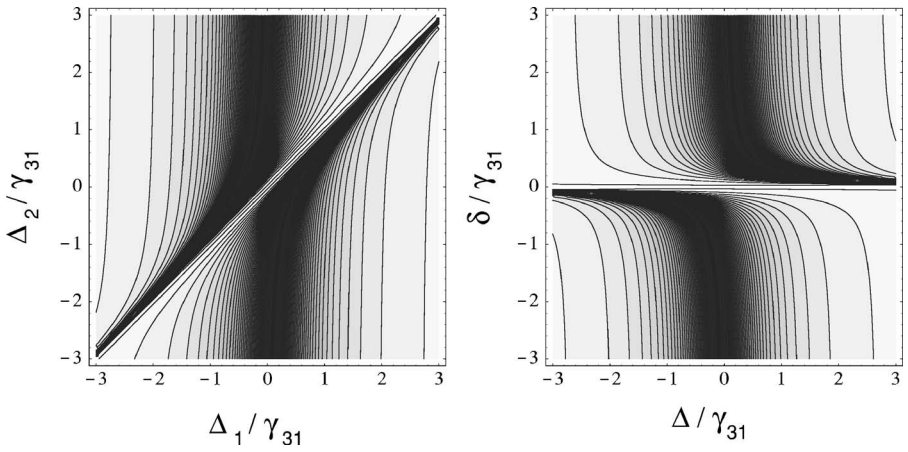

FIG. 9. Contour plot of imaginary part of susceptibility $\mathrm{Im}[\chi^{(1)}]$: left, as a function of detunings Δ_1 and Δ_2; right, as a function of single-photon detuning Δ and two-photon detuning δ in units of γ_{31}. White areas correspond to low absorption, dark to large absorption. The insensitivity of the induced transparency at $\delta=0$ on the single-photon detuning is apparent.

absorption due to γ_{21} provides a fundamental limit for many of the EIT applications. For the $\gamma_{21} \geqslant \gamma_{31}$ case depicted in Fig. 10(c), 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. 3(a)–3(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).

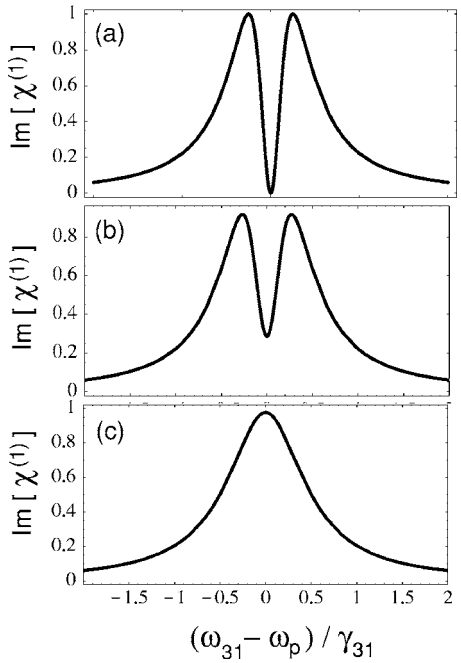

FIG. 10. Absorption spectrum for nonvanishing decay of $|2\rangle$-$|1\rangle$ transition in arbitrary units: (a) $\gamma_{31}=1$, $\gamma_{21}=0$; (b) $\gamma_{31}=1$, $\gamma_{21}=0.1$; (c) $\gamma_{31}=1$, $\gamma_{21}=10$. In all cases $\Omega_c=0.5$.

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 that 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 γ_{21} and γ_{32} (Imamoglu, 1991). In contrast, for a coupling field with large amplitude fluctuations, the absorption profile could be smeared out. In applications where a nonperturbative 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 EIT is preserved to first order.

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 copropagating 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 $\Delta\nu_{\mathrm{Dopp}} > \Omega_c$ will wash out the level splitting and the interference. EIT can be recovered by increasing the coupling field intensity so as to satisfy $\Omega_c > \Delta\nu_{\mathrm{Dopp}}$; in this limit we obtain

$$\mathrm{Im}[\chi^{(1)}(-\omega_p,\omega_p)]_{\omega_p=\omega_{31}} \propto \frac{\Delta\nu_{\mathrm{Dopp}}^2\gamma_{31}}{\Omega_c^4}. \tag{15}$$

This strong dependence on reciprocal Ω_c is a direct consequence of robust quantum interference. We emphasize that in this large coupling field limit, we can consider Ω_c as the effective detuning from dressed-state resonances. As a result of quantum interference, this effective detuning can be used to suppress absorption much more strongly than in noninterfering 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 (Dalibard *et al.*, 1992; Gardiner *et al.*, 1992; Carmichael, 1993). 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 nonunitary 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 in which 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 repopulation 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 the population decay of state $|2\rangle$ (at rate Γ_2) to mimic dephasing. We therefore introduce an additional anti-Hermitian decay term to obtain an effective Hamiltonian in the rotating frame

$$H_{\text{eff}} = -\frac{\hbar}{2}[\Omega_p\hat{\sigma}_{31} + \Omega_c\hat{\sigma}_{32} + \text{H.c.}] + \hbar\left(\Delta - \frac{i}{2}\Gamma_3\right)\hat{\sigma}_{33} + \hbar\left(\delta - \frac{i}{2}\Gamma_2\right)\hat{\sigma}_{22}. \tag{16}$$

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

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

with an eigenenergy $\hbar\omega_0=0$ independent of the values of the Rabi frequencies and the single-photon detuning. We here have assumed real-valued Rabi frequencies, which are always possible as long as propagation effects are not considered. In the time-dependent response, we shall see that the emergence of EIT can be understood as a result of the system's 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 $\delta=0$ moves 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 Ω_c is nonperturbative. 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 $|\Psi(t)\rangle$:

$$|\Psi_d(t)\rangle = U|\Psi(t)\rangle, \tag{18}$$

where

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

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

$$\tilde{H}_{\text{eff}} = -\frac{\hbar}{2}\begin{bmatrix} 0 & \Omega_p\sin\phi & \Omega_p\cos\phi \\ \Omega_p\sin\phi & -2\Delta_- + i\Gamma_- & -\frac{i}{2}\Gamma_{+-} \\ \Omega_p\cos\phi & -\frac{i}{2}\Gamma_{+-} & -2\Delta_+ + i\Gamma_+ \end{bmatrix}, \tag{20}$$

where Δ_- and Δ_+ 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 $\tilde{H}_{\text{eff}}$ in the limit of perturbative Ω_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

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

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.

$\tilde{H}_{\text{eff}}$ 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 $\kappa_{23}=\kappa_{32}=0.5(\Gamma_3-\Gamma_2)\sin 2\phi$ that gives rise to quantum interference in absorption. When $\Gamma_2=0$, we find that $\kappa_{23}=\sqrt{\Gamma_{2d}\Gamma_{3d}}$, 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 (1961). Let us 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 $|f\rangle$. To reach this final state, the atom can virtually excite either of the two dressed states; since excitations are virtual, the probability amplitudes corresponding to each have to be added, leading to quantum interference. State $|f\rangle$ could either be a fourth atomic state not coherently coupled to the others or it could be the state $|1\rangle$ itself. In the latter case, the absorption process is Rayleigh scattering of probe photons.

Having seen the simplicity of the analysis based on effective non-Hermitian Hamiltonians, we next address the question of nonlinear susceptibilities. Since the latter are by definition calculated in the limit of near-unity ground-state occupancy, $H_{\rm eff}$ of Eq. (16) provides a natural starting point. In this section, we shall focus on sum-frequency generation to highlight the different role quantum interference effects play in nonlinear susceptibilities as compared to linear ones. To this end, we introduce an additional driving term $\Omega_{\rm eff}$ 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 ω_a and ω_b) and a set of intermediate (nonresonant) states $|i\rangle$:

$$\Omega_{\rm eff}=\sum_i \frac{\Omega_a\Omega_b}{2}\left[\frac{1}{\omega_{i1}-\omega_a}+\frac{1}{\omega_{i1}-\omega_b}\right]. \tag{22}$$

Here $\Omega_a=\mu_{1i}E_a/\hbar$ and $\Omega_b=\mu_{i2}E_b/\hbar$.

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

$$H_{\rm eff}^{\rm sfg}=-\frac{\hbar}{2}[\Omega_{\rm eff}\hat{\sigma}_{21}+\Omega_p\hat{\sigma}_{31}+\Omega_c\hat{\sigma}_{32}+{\rm H.c.}] +\hbar\left(\Delta-\frac{i}{2}\Gamma_3\right)\hat{\sigma}_{33}+\hbar\left(\delta-\frac{i}{2}\Gamma_2\right)\hat{\sigma}_{22}. \tag{23}$$

To obtain the nonlinear susceptibility, we assume both $\Omega_{\rm eff}$ and Ω_p to be perturbative. The generated polarization at ω_p is given by

$$P(t)=\varrho\mu_{13}a_3(t)\exp(-i\omega_p t)+{\rm c.c.}, \tag{24}$$

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

$$\chi^{(3)}(-\omega_p,\omega_a,\omega_b,\omega_c) =\frac{2\mu_{23}\mu_{31}\varrho}{3\epsilon_0\hbar^3}\frac{1}{\Omega_c^2+(\gamma_{31}+i2\Delta)(\gamma_{21}+i2\delta)} \times\sum_i \mu_{1i}\mu_{i2}\left[\frac{1}{\omega_{i1}-\omega_a}+\frac{1}{\omega_{i1}-\omega_b}\right]. \tag{25}$$

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 $\chi^{(1)}(-\omega_p,\omega_p)=0$.

It is perhaps illustrative to compare $\chi^{(3)}$ obtained for the EIT system with the usual third-order nonlinear susceptibility for nonresonant 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 nonresonant susceptibility $(1/\Delta\omega_2\Delta\omega_3)$ are 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

$$T(\omega_p,z)=\exp\{ikz\chi^{(1)}(-\omega_p,\omega_p)/2\}, \tag{26}$$

with $k=2\pi/\lambda$ the resonant wave number 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 Eq. (26), one finds that the transmittivity near resonance $\delta=0$ is a Gaussian with width

$$\Delta\omega_{\rm trans}=\frac{\Omega_c^2}{\sqrt{\Gamma_{31}\gamma_{31}}}\frac{1}{\sqrt{\varrho\sigma z}} \quad (\varrho\sigma z\gg 1). \tag{27}$$

Here $\sigma=3\lambda^2/2\pi$ is the absorption cross section of an atom and ϱ is the atom number density. One recognizes a power broadening of the resonance with increasing Rabi frequency Ω_c or intensity of the coupling field,

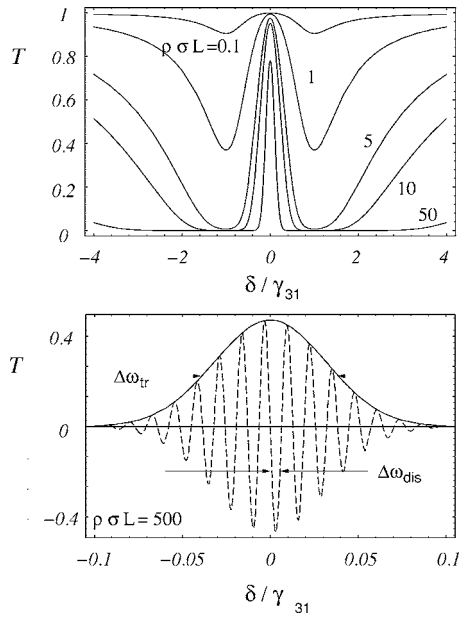

FIG. 11. Transmission spectrum through an EIT medium: top, dependence on density length products $\varrho\sigma z$; bottom, illustration of dispersive width.

which can, however, be partially compensated for by increasing the density-length product $\varrho\sigma z$. This important property is illustrated in Fig. 11, where a finite decay rate γ_{21} 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 χ on δ, can be used to detect shifts of the EIT resonance by phase-sensitive detection. Let us define $\Delta\omega_{\mathrm{dis}}$ as the frequency variation associated with an accumulated phase shift of 2π for the propagation through the medium. This is given by

$$\Delta\omega_{\mathrm{disp}} = 2\pi\frac{\Omega_c^2}{\Gamma_{31}}\frac{1}{\varrho\sigma z} \quad (\varrho\sigma z \gg 1), \tag{28}$$

much smaller than $\Delta\omega_{\mathrm{trans}}$. This effect, which is also illustrated in Fig. 11, plays an important role for applications of EIT in optical detection of magnetic fields (Scully and Fleischhauer, 1992; Fleischhauer and Scully, 1994).

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 characteristic 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 happen either 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. Optical pumping may not, however, be well suited to achieve electromagnetically induced transparency in optically thick media. Here radiation trapping can lead to a dramatic slowdown 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 nonrecoverable loss of photons from the probe field, which could be fatal if the probe field were 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 that does not involve spontaneous emission at all. The process that achieves this is called stimulated Raman adiabatic passage (STIRAP) and was discovered and developed by Bergmann *et al.* (see also Kuklinski *et al.*, 1989 and Gaubatz *et al.*, 1990) 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 now a widely used method in atomic and molecular physics for the preparation of specific quantum states and for controlling chemical reactions. It also has 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 the rotating-wave approximation and in a proper rotating frame is

$$H(t) = -\frac{\hbar}{2}\begin{bmatrix} 0 & 0 & \Omega_p(t) \\ 0 & 0 & \Omega_c(t) \\ \Omega_p(t) & \Omega_c(t) & -2\Delta \end{bmatrix}. \tag{29}$$

We assume that the relative phase between the two fields is arbitrary but constant and thus both Ω_p and Ω_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 trans-

form 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 $|2\rangle$ or vice versa without populating the intermediate excited state $|3\rangle$ 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

$$|a^0(t)\rangle = \cos\theta(t)|1\rangle - \sin\theta(t)|2\rangle, \quad \tan\theta = \frac{\Omega_p(t)}{\Omega_c(t)} \tag{30}$$

is the adiabatic dark state, which has no overlap with the excited state $|3\rangle$ and thus does not lead to spontaneous emission.

An 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 requirements besides the correct initial conditions are that the time rate of change of the fields be sufficiently slow, i.e., adiabatic, and that the relative phase between the fields be 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,

$$\tilde{H} = -\hbar \begin{bmatrix} \omega^+(t) & \frac{i}{2}\dot{\theta} & 0 \\ -\frac{i}{2}\dot{\theta} & 0 & -\frac{i}{2}\dot{\theta} \\ 0 & \frac{i}{2}\dot{\theta} & \omega^-(t) \end{bmatrix}, \tag{31}$$

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 $|\omega^\pm|$ (7):

$$\left| \left\langle \frac{d}{dt} a_0(t) \middle| a_\pm(t) \right\rangle \right| \ll |\omega^\pm(t)|, \tag{32}$$

which for the case of single-photon resonance $\Delta=0$ simplifies to

$$\Omega(t) \equiv \sqrt{\Omega_p^2(t) + \Omega_c^2(t)} \gg |\dot{\theta}(t)|. \tag{33}$$

In lowest order of the nonadiabatic 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)

$$\Gamma_{\text{eff}} = \Gamma_3 \frac{|\dot{\theta}(t)|^2}{\Omega^2(t)}. \tag{34}$$

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 nonadiabatic losses due to this effect.

If all atoms are initially in state $|1\rangle$, the coupling field (Ω_c) is applied before the probe field (Ω_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,

$$E_{\text{prep}} = \hbar\omega_p N \frac{|\Omega_p|^2}{|\Omega_c|^2}. \tag{35}$$

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 shall see later in Sec. 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, especially 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 ω_{c0} is sufficiently close to the frequency ω_0 of electromagnetically induced transparency, the presence of the medium will lead to a very strong pulling of the resonance frequency ω_c of the combined atom-cavity system towards this value (Lukin, Fleischhauer, *et al.*, 1998). At the same time the resonance width is substantially reduced (Lukin, Fleischhauer, *et al.*, 1998). 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 ω. For simplicity we assume a unidirectional ring resonator of round-trip length L with a single in-out coupling mirror. Then

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

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

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

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 Eq. (36) that the width of the cavity resonance is changed according to

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

The first term accounts for a small enhancement of the cavity width due to the additional losses induced by the medium. Much more important, however, is 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 by 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, Fleischhauer, *et al.*, 1998).

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 nonlinear 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.

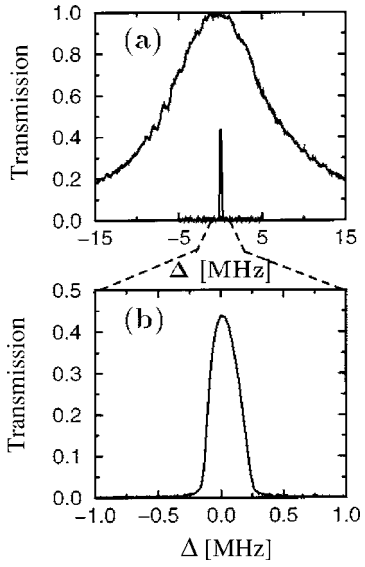

FIG. 12. Experimental demonstration of linewidth narrowing: (a) transmission spectrum of empty cavity and with EIT medium; (b) magnification of transmission spectrum with EIT. From Müller *et al.*, 1997.

Extensive theoretical work was carried out by Scully and co-workers (Scully, 1991; Fleischhauer *et al.*, 1992a, 1992b; Scully and Zhu, 1992; Rathe *et al.*, 1993) 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 (Moseley *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 has led to the suggestion that very sensitive interferometers and magnetometers be used, since tiny variations in frequency couple to large changes in refractive index (Scully and Fleischhauer, 1992; Fleischhauer and Scully, 1994; Budker *et al.*, 1998, 1999; Nagel *et al.*, 1998; Sautenkov *et al.*, 2000; Stahler *et al.*, 2001; Affolderbach *et al.*, 2002). In the magnetometer schemes this greatly increases the sensitivity of the measurement of the Zeeman shifts induced by a magnetic field.

As mentioned earlier, lasing 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 (Gao *et al.*, 1992; Fry *et al.*, 1993; Nottleman *et al.*, 1993) as well as actual demonstrations of laser oscillation without population inversion (Zibrov *et al.*, 1995; Padmabandu *et al.*, 1996). An application of lasing without inversion to short-wavelength 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 already contains all necessary information, it is worthwhile to devote a separate section to this issue, the more so as the almost perfect transparency at certain frequencies, characteristic of 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 often have 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 ultraslow 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

$$\mathrm{Re}[\chi^{(1)}] = \eta\frac{2\Gamma_{31}\delta}{\Omega_c^2} + \mathcal{O}(\delta^2), \tag{39}$$

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

$$v_{\mathrm{gr}} \equiv \left.\frac{d\omega_p}{dk_p}\right|_{\delta=0} = \frac{c}{n+\omega_p(dn/d\omega_p)}, \tag{40}$$

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

$$v_{\mathrm{ph}} \equiv \left.\frac{\omega_p}{k_p}\right|_{\delta=0} = \frac{c}{n} = c. \tag{41}$$

An important property of the EIT system is that the second-order term in Eq. (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 Eq. (39) yields at two-photon resonance

$$v_{\mathrm{gr}} = \frac{c}{1+n_{\mathrm{gr}}} \quad \text{with } n_{\mathrm{gr}} = \varrho\sigma c\frac{\Gamma_{31}}{\Omega_c^2}, \tag{42}$$

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

$$\tau_d = L\left(\frac{1}{v_{\mathrm{gr}}}-\frac{1}{c}\right) = \frac{Ln_{\mathrm{gr}}}{c} = \varrho\sigma L\frac{\Gamma_{31}}{\Omega_c^2}. \tag{43}$$

It should be noted that under nonideal conditions, i.e., if the dark resonance is not perfectly stable, for example 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 γ_{31} and γ_{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 ϱ.

Due to the vanishing imaginary part of the susceptibility, i.e., perfect transparency, at $\delta=0$, relatively high atom densities ϱ 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 co-workers in lead vapor a group velocity of $v_{\mathrm{gr}}/c\approx 165$ was observed (Kasapi *et al.*, 1995). In later experiments by Meschede *et al.* a dispersive slope corresponding to a value of $v_{\mathrm{gr}}/c\approx 2000$ was found (Schmidt *et al.*, 1996). Other earlier experiments in which directly or indirectly slow group velocities were measured were performed by Xiao *et al.* (1995) and by Lukin *et al.* (1997). The slowdown 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 slowdown of seven orders of magnitude (Hau *et al.*, 1999). Similarly small values were 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 slowdown of the group velocity was also observed in the solid state by Turukhin *et al.* (2001).

The lossless slowdown 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 because when the pulse enters the sample its front end propagates much more slowly 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 submillimeter 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 a factor $v_{\rm gr}/c$ 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 $v_{\rm gr} \ll c$, 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 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 this picture into a mathematical formalism one can employ a quasiparticle picture first introduced by Mazets and Matisov (1996) and independently by Fleischhauer and Lukin (2000), in which this concept was first applied to pulse propagation. One defines dark (Ψ) and bright (Φ) polariton fields according to

$$\Psi(z,t) = \cos\vartheta \mathcal{E}_p(z,t) - \sin\vartheta\sqrt{\varrho}\rho_{21}(z,t)e^{i\Delta kz}, \tag{44}$$

$$\Phi(z,t) = \sin\vartheta \mathcal{E}_p(z,t) + \cos\vartheta\sqrt{\varrho}\rho_{21}(z,t)e^{i\Delta kz}, \tag{45}$$

with the mixing angle determined by the group index

$$\tan^2\vartheta = \frac{\varrho\sigma c\Gamma_{31}}{\Omega_c^2} = n_{\rm gr}. \tag{46}$$

$\mathcal{E}_p$ is the normalized, slowly varying probe field strength, $E_p = \mathcal{E}_p\sqrt{\hbar\omega/2\epsilon_0}$, with ω being the corresponding carrier frequency. $\Delta k = k_c^{\parallel} - k_p$, where k_p is the wave number of the probe field and $k_c^{\parallel}$ is the projection of the coupling-field wave vector along the z axis. ρ_{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, 2001). One finds that only the dark polariton field Ψ is excited, i.e., $\Phi \equiv 0$, and thus

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

$$\rho_{21}(z,t) = -\sin\vartheta\Psi(z,t)\frac{e^{-i\Delta kz}}{\sqrt{\varrho}}. \tag{48}$$

Furthermore, under conditions of single-photon resonance, Ψ obeys the simple shortened wave equation

$$\left[\frac{\partial}{\partial t} + c\cos^2\vartheta\frac{\partial}{\partial z}\right]\Psi(z,t) = 0, \tag{49}$$

which describes a form-stable propagation with velocity

$$v_{\rm gr} = c\cos^2\vartheta. \tag{50}$$

The slowdown of the group velocity of light in an EIT medium can now be given a very simple interpretation: EIT corresponds to the lossless and form-stable propagation of dark-state polaritons. These quasiparticles are a coherent mixture of electromagnetic and atomic spin excitations, the latter referring to an excitation of the $|1\rangle$-$|2\rangle$ coherence. The admixture of the components described by the mixing angle ϑ depends on the strength of the coupling field as well as the density of atoms and determines the propagation velocity. In the limit $\vartheta \to 0$, corresponding to a strong coupling field, the dark-state polariton is almost entirely electromagnetic in nature, and the propagation velocity is close to the vacuum speed of light c. In the opposite limit, $\vartheta \to \pi/2$, the dark-state polariton 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, 2001) 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, to be 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 ($|v| \ll c$) relative to the propagation direction of the light pulse, Galilean transformation rules predict that the group velocity [Eq. (50)] will be modified according to

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

This expression can also be obtained from Eq. (40) if one takes into account that as a result of the Doppler effect the susceptibility or the index of refraction becomes k dependent (spatial dispersion). Light dragging according to Eq. (51) was recently observed by Strekalov *et al.* (2004). It is interesting to note that $v_{\rm gr}$ can now be zero or even negative for $\vartheta \neq \pi/2$ if the atoms move in

the opposite direction to the propagation of light. In fact, since extremely large values of the group index can be achieved in vapor cells, the thermal motion of atoms even at room temperatures can have a significant effect on the effective group velocity. Making use of this effect, Kocharovskaya *et al.* suggested "freezing" a light pulse in a Doppler-broadened medium, where a specific velocity class is selectively excited (Kocharovskaya *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 stopping 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 acceleration of the atoms. 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), as can be observed in vapor cells where the atoms undergo 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 nonlinear processes (Harris and Hau, 1999; Lukin, Yelin, and Fleischhauer, 2000; Lukin and Imamoglu, 2001). It should be noted in this context that, although the number of photons in the medium is reduced by the ratio of the group velocity to the vacuum speed of light, the electric field is continuous at the boundaries and thus nonlinear polarization is not reduced as a result of compression. These applications will be discussed in more detail in Secs. V and VI. Making use of the substantial pulse deformation at the boundary of an EIT medium is also of interest for the storage of information contained in long pulses, which in this way can be compressed to a very small spatial volume.

As noted by Harris, pulse compression is associated with a substantial enhancement of the spatial field gradient in the propagation direction (Harris, 2000), and thus ponderomotive 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 a force enhanced by the ratio c/v_{gr}:

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

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 ultracold quantum gases. For example, 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, 1999b). Stopping of a light pulse at a spatial discontinuity of the control field was used by Dutton *et al.* (2001) to create strongly localized shock waves in a Bose-Einstein condensate and to study their dynamics.

Finally, interesting effects have been predicted by Leonhardt and co-workers when the propagation of ultraslow light is considered in *nonuniformly moving media*. Using an effective classical field theory for slow light, in which 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 and Piwnicki 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, 2000a). This may allow for laboratory studies of some general relativistic effects when adding an inward flow to the vortex (Leonhardt and Piwnicki, 2000b; Visser, 2000). The practical observation of an optical analogy to the event horizon of a black hole will, however, most likely be prevented by absorption and reflection effects.

To assess the potential 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 τ_d of a pulse in an EIT medium to its pulse length τ_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 nonresonant frequency components is nonzero even under ideal conditions of an infinitely long-lived dark state. Thus, in order for the absorption to be negligible, first the decay time of the $|1\rangle$-$|2\rangle$ transition must be very small and second the spectral width of the pulse $\Delta\omega_p$ has to be much smaller than the transparency width $\Delta\omega_{\mathrm{trans}}$:

$$\Delta\omega_p \ll \Delta\omega_{\mathrm{trans}} = \frac{\Omega_c^2}{\sqrt{\Gamma_{31}\gamma_{31}}} \frac{1}{\sqrt{\varrho\sigma L}}. \tag{53}$$

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

$$\Delta\omega_{\mathrm{trans}} = \sqrt{\varrho\sigma L} \frac{1}{\tau_d} \sqrt{\frac{\Gamma_{31}}{\gamma_{31}}}. \tag{54}$$

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 ultraslow pulse by means of atomic motion antiparallel to the flow of light. Making use of $\Delta\omega_p \tau_p \geqslant 1$ one finds an upper limit for the ratio of group delay to pulse length (Harris and Hau, 1999)

$$\frac{\tau_d}{\tau_p} \ll \sqrt{\varrho\sigma L}. \tag{55}$$

The product $\varrho\sigma L$ is the opacity of the medium in the absence of EIT, i.e., $\exp(-\varrho\sigma L)$ is the transmission coefficient at bare atomic resonance. Thus a noticeable

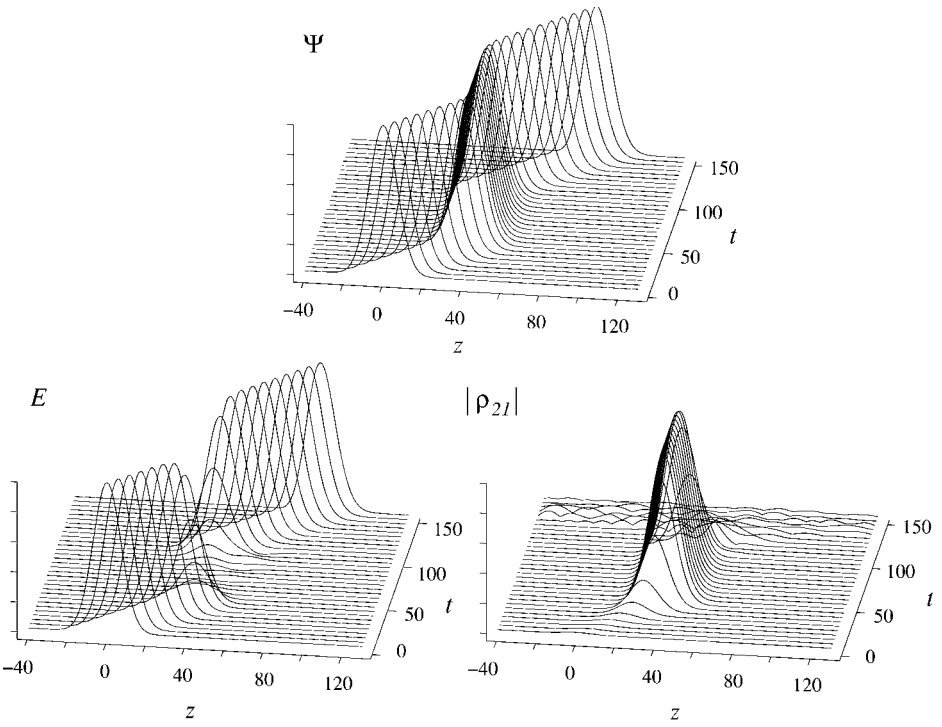

FIG. 13. Numerical simulation of dark-state polariton propagation with envelope $\exp[-(z/10)^2]$. The mixing angle is rotated from 0 to $\pi/2$ and back according to $\cot\vartheta(t) = 100\,(1 - 0.5\tanh[0.1\,(t-15)] + 0.5\tanh[0.1\,(t-125)])$: top, polariton amplitude; bottom left, electric field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

time delay of a pulse by EIT requires an optically thick medium with $\varrho\sigma L > 1$.

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 of transferring spatial excitation distributions of atomic ensembles to light pulses was pointed out before by Csesznegi and Grobe (1997). In the following the "stopping" and "reacceleration" 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 expression "stopping of light" should not be taken literally. As mentioned before, the reduction of the propagation velocity of light in a lossless, 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 ultraslow pulse propagation, in which likewise only a tiny fraction of the original excitation remains in the form of photons.

A key conceptual advance with respect to the stopping of a light pulse occurred when it was realized by Fleischhauer and Lukin (2000) that the bandwidth limitation [Eq. (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,

$$\Delta\omega_p(t) = \Delta\omega_p(0)\frac{v_{\rm gr}(t)}{v_{\rm gr}(0)}, \tag{56}$$

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 $\vartheta \to \vartheta(t)$. Adiabatically rotating $\vartheta(t)$ from 0 to $\pi/2$ leads to a deceleration of the polariton to a full stop. At the same time its character changes from that of an electromagnetic field to that of 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.

Conditions of adiabaticity have been analyzed by Matsko, Rostovtsev, Kocharovskaya, *et al.* (2001) and by Fleischhauer and Lukin (2001). 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 switchoff would lead only to a loss of the very small electromagnetic component of the polariton (Liu *et al.*, 2001; Matsko, Rostovtsev, Kocharovskaya, *et al.*, 2001; Fleischhauer and Mewes, 2002). Reversing the adiabatic rotation of ϑ by increasing the strength of the coupling field leads to a reacceleration of the dark-state polariton associated with a change of character from spinlike to electromagnetic. In Fig. 13 a numerical simulation of the stopping of a dark-state polariton and its successive reacceleration 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

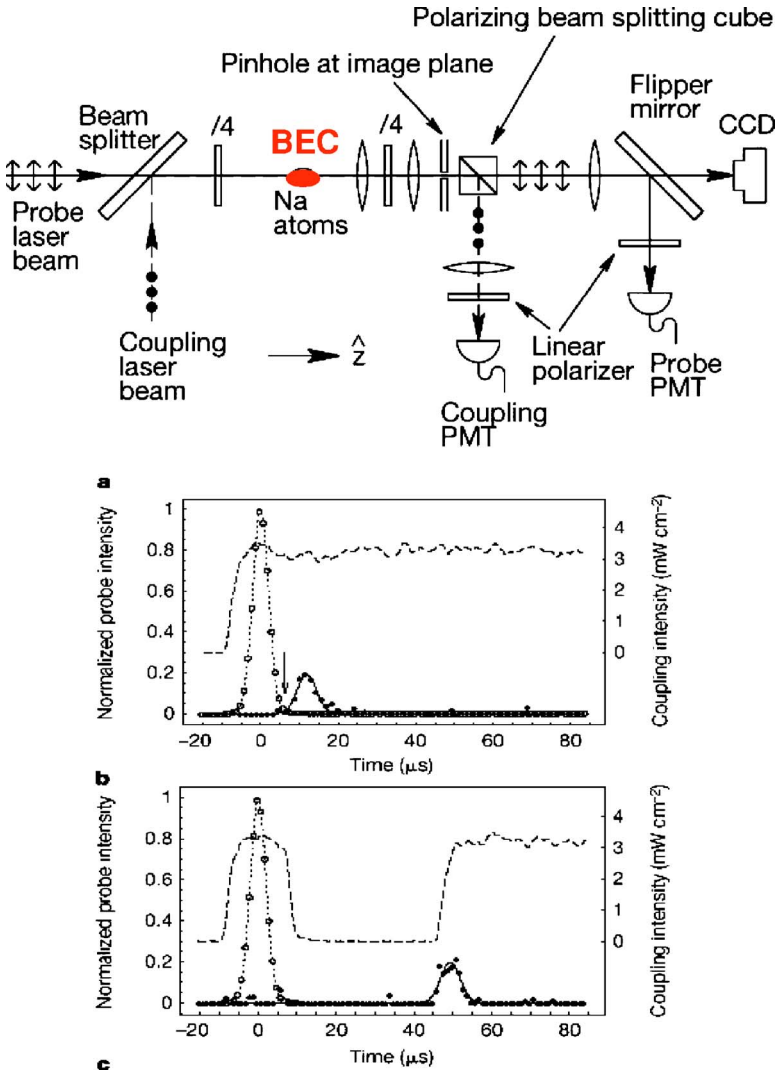

FIG. 14. (Color in online edition) Storage and retrieval of light in ultracold Na atoms: top, experimental setup for observing light storage and retrieval; bottom, measurements of delayed (a) and revived (b) probe pulse after storage. Dashed line shows intensity of coupling fields. For details see Liu *et al.*, 2001. From Liu *et al.*, 2001.

reversibly transferred and stored in long-lived spin coherences. It should be noted that the spin excitation does not in general store the photon energy, as most of it is transferred to the coupling field by stimulated Stokes emission.

The classical aspects of stopping and reaccelerating of light pulses have been experimentally demonstrated by Liu and co-workers in ultracold Na (Liu *et al.*, 2001), by Phillips *et al.* in hot Rb vapor (2001), and in solids by Turukhin and collaborators (Turukhin *et al.*, 2001). The experiment of Liu *et al.* used a similar setup to that used for the demonstration of the group velocity reduction to 17 m/s (Hau *et al.*, 1999). Here a light pulse was slowed and spatially compressed in an ultracold 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 a pulse length of a few μs were achieved. Figure 14 shows the experimental setup as well as the storage and retrieval of a light pulse. The use of ultracold gases has advantages such as the reduction of two-photon Doppler shifts and high densities. Ultracold gases, however, are 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 Doppler shifts, degenerate Zeeman sublevels were used for states $|1\rangle$ and $|2\rangle$. Moreover, to reduce the effects of atomic motion a He buffer gas was employed. Here storage times of up to 0.5 ms could be obtained for a pulse length of several μs. The first successful demonstration of light "storage" in a solid was achieved by Turukhin and co-workers in Pr-doped Y_2SiO_5 after proper preparation of the inhomogeneously broadened material by a special optical hole- and antihole-burning technique (Turukhin *et al.*, 2001). A direct proof of the coherence of the storage mechanism was provided by Mair *et al.* (2002), when 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 moreover performed 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 that 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 $|1\rangle_{\mathrm{ph}}$ is mapped to an ensemble of N three-level atoms according to

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

$$\updownarrow$$

$$|0\rangle_{\mathrm{ph}} \otimes \frac{1}{\sqrt{N}} \sum_{j=1}^{N} |1,1,\ldots,2_j,\ldots,1\rangle. \tag{57}$$

In this way the EIT medium can act as a quantum memory for photons. 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), that 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, 2005), and fluctuations of coupling parameters (Sun *et al.*, 2003). An extension of the model to three spatial dimensions was given by Duan *et al.* (2002).

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 counterpropagating coupling lasers 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 counterpropagating components of the coupling laser regenerated two Stokes pulses with opposite wave vectors. Thus the regenerated light pulses also 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 μs. 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 abovementioned 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, Yelin, and Fleischhauer, 2000). An alternative and often experimentally simpler way to achieve the same effect 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 the creation of 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 for creating 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 nondemolition measurements to prepare quantum states with sub-shot-noise spin fluctuations (Kuzmich *et al.*, 1999, 2000). Duan *et al.* (2000) suggested a detection scheme in which coherent light simultaneously probes 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 co-workers have demonstrated this technique in a remarkable experiment (Julsgaard *et al.*, 2001).

Finally, Imamoglu (2002) and James and Kwiat (2002) have suggested transferring photons to atomic ensembles to build single-photon detectors with high efficiency, which are able to distinguish between single and multiple photons. 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. Now we consider the nonperturbative situation of probe and coupling pulses with comparable strength, which 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 in which 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 with slowly varying probability 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 de-

scribed by a Schrödinger equation for $C_n(z,t)$ 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 Γ_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 a slowly varying amplitude and phase approximation one finds the shortened wave equations for the probe (Ω_p) and coupling pulse (Ω_c),

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

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

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

$$\alpha_p=\frac{\omega_p\varrho|\mu_{12}|^2}{2\epsilon_0\hbar},\quad \alpha_c=\frac{\omega_c\varrho|\mu_{32}|^2}{2\epsilon_0\hbar}, \tag{60}$$

with μ_{ij} being the dipole moments of the corresponding transitions.

If the amplitudes of the fields vary sufficiently slowly that the adiabatic approximation holds, and if the appropriate initial conditions are fulfilled, the atoms stay at all times in the instantaneous dark state, $|a^0(t)\rangle=\cos\theta(t)|1\rangle-\sin\theta(t)|2\rangle$, 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 therefore have no effect at all. Light and matter are exactly decoupled.

As first noted by Harris (1993), there is another case in which bichromatic 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 $\Omega_c(z,t)=\Omega_c f(z,t)$, 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 counterintuitive 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

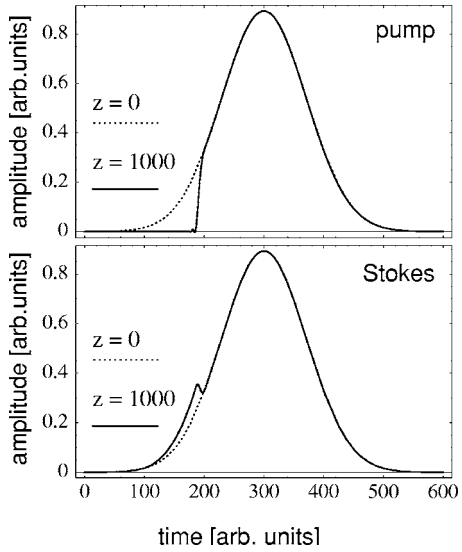

FIG. 15. Comparison of probe field and coupling field: top, amplitude of probe field as function of time in arbitrary units; dotted line, at medium entrance $z=0$; solid line, for $z=1000$; bottom, the same for the coupling field. Distance is measured in units of absorption length and peak values are $\Omega_c=\Omega_p=10\Gamma$.

numerical calculations are used to show the propagation of two matched pulses in an unprepared medium.

Apart from the preparation at the front end, matched pulses are stable solutions of the propagation problem. They should therefore be formed whenever pulses of arbitrary shape are applied to optically thick three-level media. In fact, it has been shown by Harris (1993) that nonequal pairs of pulses with a strong cw carrier,

$$\Omega_p(z,t)=[1+f(z,t)]\Omega_p e^{-i\omega_p(t-z/c)}, \tag{61}$$

$$\Omega_c(z,t)=[1+g(z,t)]\Omega_c e^{-i\omega_c(t-z/c)}, \tag{62}$$

tend to adjust their amplitude modulations $f(z,t)$ and $g(z,t)$ in the course of propagation, i.e.,

$$\left.\frac{f(z,t)}{g(z,t)}\right|_{z\to\infty}=1. \tag{63}$$

Fluctuations in the difference of f and g lead to nonadiabatic transitions out of the dark state, defined by the cw components, and will thus be absorbed. Eventually pulses of identical envelope are formed. The tendency to generate pulses with identical envelopes is not restricted to fields with a strong cw carrier but is also found for arbitrary pulses (pulse matching). The phenomenon of pulse matching causes a correlation of quantum fluctuations in both fields, as discussed, for example, by Agarwal (1993), Fleischhauer (1994), and Jain (1994).

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 reduction in group velocity, it is necessary to include first-order nonadiabatic corrections in the dynamics of the atoms, which lead to small contributions to the excited-state amplitude. Taking these into account, one finds for the state amplitudes

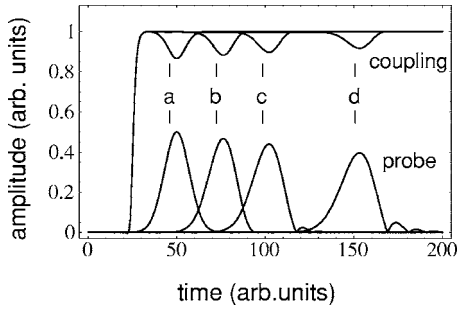

FIG. 16. Propagation of adiabatons: amplitudes of probe and coupling fields as a function of time in a comoving frame for $n_{\mathrm{gr}}z/c$ are (a) 0, (b) 25, (c) 50, and (d) 100 from numerical solution of propagation equations (Fleischhauer and Manka, 1996). Arbitrary space and time units with $c=1$.

$$C_1=\cos\theta, \quad C_2=\frac{2i}{\Omega}\dot{\theta}, \quad C_3=-\sin\theta, \tag{64}$$

where $\Omega=\sqrt{\Omega_p^2+\Omega_c^2}$. It was shown by Grobe *et al.* (1994) that the nonlinear equations of propagation, Eqs. (58) and (59), together with Eq. (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 Ω_p and Ω_c in propagation equations for the rms Rabi frequency Ω and the nonadiabatic coupling $\dot{\theta}$. Under nearly adiabatic conditions, $|\dot{\theta}|\ll\Omega$, 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

$$\left(\frac{\partial}{\partial t}+c\frac{\partial}{\partial z}\right)\Omega(z,t)\approx 0. \tag{65}$$

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

$$\left(\frac{\partial}{\partial t}+c\frac{\partial}{\partial z}\right)\dot{\theta}(z,t)=-\alpha\frac{\partial}{\partial t}\left(\frac{\dot{\theta}}{\Omega^2}\right), \tag{66}$$

which is exactly integrable. The corresponding solutions, called *adiabatons* (Grobe *et al.*, 1994), are particularly simple if Ω is approximately constant over the time interval of interest. In that case the probe and coupling pulses have complementary envelopes and $\dot{\theta}$ propagates without changing form with velocity $v_{\mathrm{gr}}=c/(1+\alpha/\Omega^2)$. The quasi-form-invariant propagation of an adiabaton is 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.

The 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 by Hioe and Grobe (1994). The effect of nonequal coupling strength on adiabaton propagation was studied by Grigoryan and Pashayan (2001). The adiabaton solutions are approximately stable over many single-photon absorption lengths. However, as shown by Fleischhauer and Manka (1996), they eventually decay to matched pulses after sufficiently long propagation distances.

For time scales short enough that decays can be ignored, and for equal coupling strength, exact solutions of the nonlinear propagation problem in V- and Λ-type systems exist even beyond the adiabatic approximation. Konopnicki and Eberly (1981) and Konopnicki *et al.* (1981) found soliton solutions with identical pulse shape, called *simultons*. An interpretation of simultons in terms of solitons corresponding to an effective two-level problem was given by Fleischhauer and Manka (1996). Malmistov (1984) and Bol'shov and Likhanskii (1985) derived simulton solutions in three-level systems using the inverse-scattering method. Several extensions of simultons have been discussed. *N*-soliton solutions were given by Steudel (1988). Inhomogeneously broadened systems were considered by Rahman and Eberly (1998) and by Rahman (1999) and extensions to five-level systems were discussed by Hioe and Grobe (1994).

V. ENHANCED FREQUENCY CONVERSION

A. Overview of atomic coherence-enhanced nonlinear optics

Harris and co-workers identified the enhancement of nonlinear optical frequency conversion as a major benefit of electromagnetically induced transparency in their paper of 1990 (Harris *et al.*, 1990). Hemmer and coworkers (Hemmer *et al.*, 1995) introduced the double-Λ scheme as an important tool in 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. While destructive interference reduces the linear susceptibility of a laser-dressed system, this is not so for the nonlinear 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 that 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 three- or four-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

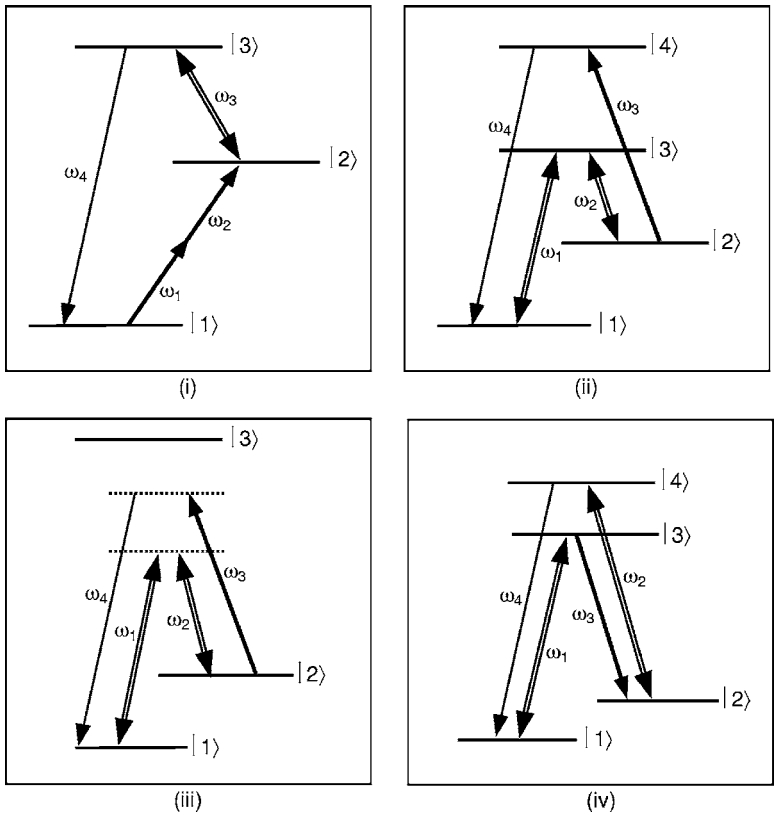

FIG. 17. Different schemes for resonantly enhanced four-wave mixing processes based on EIT and related interference phenomena: (i) sum-frequency generation (ω_4) out of two pump pulses (ω_1 and ω_2) and one strong coherent drive (ω_3); (ii) up-conversion of pump (ω_3) into generated field (ω_4) due to maximum coherence prepared by two strong drive fields (ω_1 and ω_2); (iii) off-resonant variant of (ii); (iv) parametric generation of two fields (ω_3 and ω_4) out of two pump fields (ω_1 and ω_2) in forward- or backward-scattering configuration. See text for details.

schemes illustrated in Fig. 17 the material polarization $P(\omega_4)$ is driven at frequency ω_4 by fields applied at ω_1, ω_2, and ω_3, and will in its turn drive a new electromagnetic field at ω_4. The growth of the new field depends upon the magnitude of the nonlinear source term $P^{\mathrm{nl}}(\omega_4)$ 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

$$\frac{\partial E_4}{\partial z}+\frac{1}{c}\frac{\partial E_4}{\partial t}=\frac{i}{2}\frac{\omega_4}{c\epsilon_0}(P^{\mathrm{nl}}+P^{l}). \tag{67}$$

Here the vector character of the field and 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 polarization. In the framework of the density matrices for a three-level atom we can write the positive frequency component of the polarization as

$$P=\varrho \mathrm{Tr}[\hat{\mu}\rho]=\varrho(\mu_{12}\rho_{21}+\mu_{13}\rho_{31}+\mu_{23}\rho_{32}). \tag{68}$$

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 ω_4 as

$$P^{(1)}(\omega_4)=\epsilon_0\chi^{(1)}E_4, \tag{69}$$

and the nonlinear response as

$$P^{(3)}(\omega_4)=\frac{3}{2}\epsilon_0\chi^{(3)}E_1E_2E_3, \tag{70}$$

where E_j is the electric-field amplitude associated with the frequency component ω_j. The forms of the susceptibilities that can be derived by a full quantum treatment of the atom are extensively discussed in the literature on nonlinear optics [see for, example, Reintjes (1984)]. In general, for nonresonant nonlinear mixing the susceptibility will be characterized by the appearance of three detuning terms in the denominator while the linear susceptibility has only a single detuning term. As was shown in Sec. 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 nonlinear mixing. The forms of the dressed susceptibilities for EIT are given in Sec. 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 dephasing and integration over the Doppler profile are usually required to yield 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:

$$\frac{dE_4}{dz} = i\frac{\omega_4}{4c}\chi^{(3)}E_1E_2E_3e^{i\Delta kz} - \frac{\omega_4}{2c}\,\mathrm{Im}[\chi^{(1)}]E_4 + i\frac{\omega_4}{2c}\,\mathrm{Re}[\chi^{(1)}]E_4, \tag{71}$$

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 nonlinear polarization reaches a large value then the source of the new field will be strong, and 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 that drives it. This phase-matching condition is essential and dictates (at resonance where $\mathrm{Re}[\chi^{(1)}]=0$) 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 Δk will be finite, resulting in a limited length over which the field grows before slipping out of phase with the driving polarization $l_{\mathrm{coh}}=1/\Delta k$. 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 nonresonant transitions to other levels contribute to the wave-vector mismatch.

Let us examine the four-wave mixing schemes shown in Fig. 17 in which EIT or related effects have been found to be advantageous. 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 in which a two-photon and a single-photon resonance are used for the applied fields, generating a field in resonance with an atomic transition [Fig. 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 nonlinear susceptibility 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 is applied resonantly in a lambda-configured three-level system close to both Raman and single-photon resonance [Fig. 17(ii)]. The presence of EIT eliminates absorption and dispersion in the propagation of both fields. 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 and so is restricted to high-power pulsed lasers or to Doppler-free cw laser experiments (Harris and Jain, 1997; Merriam *et al.*, 1999, 2000).

(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 Δ; Fig. 17(iii)] provided that they are strong enough so that the condition $\Omega_1\Omega_2 > \Delta\gamma_{21}$ is satisfied (where γ_{21} 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 (ii). If the maximal coherence is excited between molecular vibrational or rotational states the medium is very suited to high-order stimulated Raman sideband 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-Λ scheme, which can enhance the mixing further. An important variant of the double-Λ scheme with three applied fields is that explored by Hemmer *et al.* (1995) and by Zibrov and co-workers, in which only two fields are injected [Zibrov *et al.*, 1999; Fig. 17(iv)]. These fields are applied close to single-photon resonance with transitions in each of the two Λ systems formed in alkali-metal atoms. This scheme with cw laser fields has remarkable properties including, if the applied fields are copropagating, the growth of both pairs of fields and, if the applied fields are counterpropagating, mirrorless oscillation, as experimentally demonstrated by Zibrov *et al.* (1999).

The advantages of all of these schemes lie in the reduction of unwanted absorption and dispersion for the generated and the driving fields while 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 de-

velopment is the demonstration of high-efficiency multiorder Raman sideband 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 sidebands to synthesize ultrashort pulses was first predicted by 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, Rostovtsev, Fleischhauer, and Scully, 2001).

B. Nonlinear 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. 17(i) 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 discussed 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 nonlinear susceptibility.

Taking the linear and nonlinear susceptibilities derived in Sec. 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

$$I_4 = \frac{3n\omega^2}{8Z_o c^2}|\chi^{(3)}|^2|E_1|^2|E_2|^2|E_3|^2 F(\Delta k, z, R, I), \tag{72}$$

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 $\mathrm{Re}[\chi^{(1)}]$ and $\mathrm{Im}[\chi^{(1)}]$, respectively, and c is the speed of light in vacuum. Here we have introduced the phase-matching factor $F(\Delta k, z, R, I)$, which depends upon the details of the mixing scheme and the focal geometry. This general result indicates the importance of phase matching (and hence Δk) for the generated field intensity.

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

$$I_4 = \frac{3n\omega^2}{8Z_o c^2}|\chi^{(3)}|^2|E_1|^2|E_2|^2|E_3|^2 \times \frac{1 + e^{-\omega I z/c} - 2e^{-\omega I z/2c}\cos\left[\left(\Delta k + \frac{\omega R}{2c}\right)z\right]}{\left[\frac{\omega I}{2c}\right]^2 + \left[\Delta k + \frac{\omega R}{2c}\right]^2}. \tag{73}$$

It is implicit that $\chi^{(3)}$, $\mathrm{Re}[\chi^{(1)}]$, and $\mathrm{Im}[\chi^{(1)}]$ are the laser-dressed expressions introduced in Sec. 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

$$I_4 \sim \left|\frac{\chi^{(3)}}{\chi^{(1)}}\right|^2, \tag{74}$$

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 Fig. 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 inhomogenous spectral widths in the system (e.g., Doppler width). For example, a laser intensity of above 1 MW cm^{-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 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 ultraviolet (VUV). This is in large part because atomic species with high-ionization potentials (>10 eV) 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 nonlinear optical processes using EIT (Hemmer *et al.*, 1995; Babin *et al.*, 1996). 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 values of the susceptibilities at a given frequency are obtained, and these quantities can be used to calculate the generated field. 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 Ω_c manifest themselves most clearly. As discussed by Harris *et al.* (1990) there is expected to be a steplike increase in the conversion efficiency when the magnitude of Ω_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 that drive EIT, since a multimode 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 (Sokolov *et al.*, 2001) has highlighted that when pulsed laser fields are used additional considerations must be made. Kasapi and coworkers showed (Kasapi *et al.*, 1995) that the group velocity of a 20-ns 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 Sec. 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 Fig. 17(i)] in which the EIT was created on the $3p$-$1s$ transition at 103 nm in a lambda scheme by the application of a field at 656 nm on the $2s$-$3p$ transition (Fig. 18). A field at 103 nm 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 photionization 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 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 2×10^{-4} were reported (Zhang *et al.*, 1995).

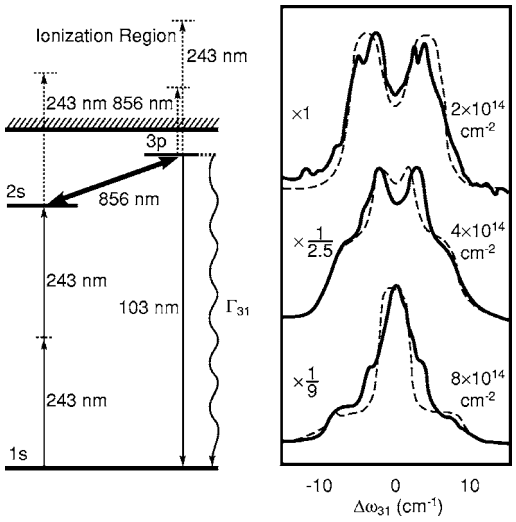

FIG. 18. Experimental demonstration of sum-frequency generation in H: left, level and coupling scheme; right, calculated (dashed) and measured (solid line) nonlinear susceptibility as function of detuning for the $3p$-$1s$ transition for different density-length products of the medium (nL). Only under the least dense condition is the Autler-Townes split structure apparent in the generated signal. At higher density strong resonant enhancement is seen. From 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 nonlinear susceptibility. This is one of the most important factors limiting the conversion efficiency in the hydrogen scheme (Hakuta, 2004). The requirements on a minimum value of $\Omega_c > \Delta_{\mathrm{Doppler}}$ constrain the conversion efficiency that can be achieved because of a scaling factor by $1/\Omega_c^2$, which 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 utilizing 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, in which 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 Δk due to the other atomic levels rather than absorption in the medium (Dorman *et al.*, 2000a).

In a Doppler-free medium the effect of constructive interference for the nonlinear term is most apparent. In a system without inhomogeneous broadening perfect transparency can be induced with $\Omega_c \ll \gamma_{31}$. As Ω_c is small relative to γ_{31}, the nonlinear 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 Fig. 7). The spectral widths of these features are typically subnatural 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 nonlinear susceptibilities in this case can be extremely large, as there is a constructive interference. Nonlinear optics at very low light levels, i.e., at the few-photon limit, are possible in this regime. Associated with their first dramatic measurements of ultraslow light, Hau *et al.* (1999) reported a nonlinear refractive index in an ultracold Na vapor that was 0.18 $\mathrm{cm}^2\ \mathrm{W}^{-1}$. 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 nonlinearity in a typical solid-state system. A discussion of this process in the few-photon limit is given in Sec. VI.

C. Nonlinear 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 three-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 nondriven transition. Considering the system illustrated in Fig. 17(ii), we can imagine that both applied fields are now strong. Under appropriate adiabatic conditions (see Sec. 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 "counterintuitive" 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 Fig. 17(iii)] provided that the condition $\Omega_1\Omega_2 > |\Delta|\gamma_{21}$ is met, where Δ 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 pulses that are sufficiently energetic to prepare all of the atoms in the beam path. The ρ_{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

$$\rho_{12} = -\frac{\Omega_1\Omega_2^*}{\sqrt{\Omega_1^2+\Omega_2^2}}\exp\{i(\omega_1-\omega_2)t-(k_1-k_2)z\} = |\rho_{12}|e^{-i\delta kz}e^{i(\omega_1-\omega_2)t}. \tag{75}$$

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 maximal coherence requires that the pulse energy exceed the following value:

$$E_{\mathrm{prep}} = \frac{f_{13}}{f_{23}}\varrho AL\hbar\omega, \tag{76}$$

where f_{ij} are the oscillator strengths of the transitions and ϱL is 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 that all atoms are in the appropriate dressed state. It should be noted that the preparation energy is much smaller for nonlinear 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:

$$\frac{dE_4}{dz} = -i\eta_4\hbar\omega_4 n[(a_4\rho_{11}+d_4\rho_{22})E_4 + b_4\rho_{12}E_3], \tag{77}$$

with 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 ρ_{12} is large. This significantly relaxes the constraints usually set by phase matching in nonlinear optics. Recently near-unity conversion efficiencies to the far UV were reported in an atomic lead system where maximum coherence had been created (Jain *et al.*, 1996; Harris *et al.*, 1997; Merriam *et al.*, 1999, 2000). An example of high-conversion-efficiency four-

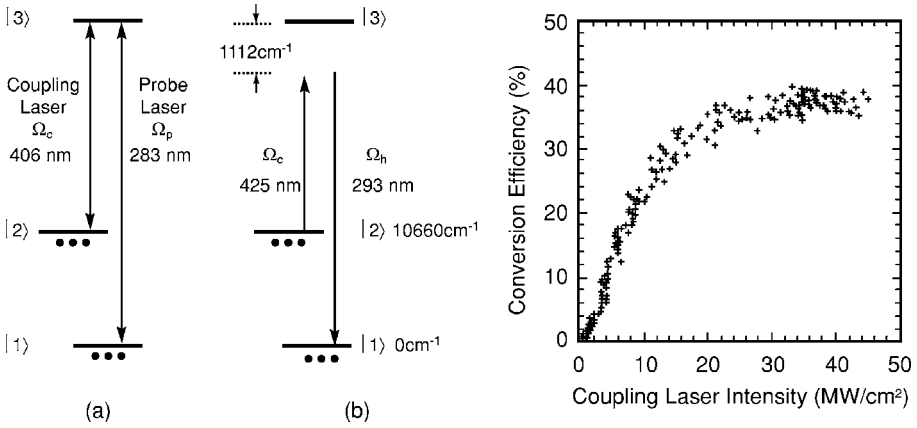

FIG. 19. Parametric up-conversion with maximum coherence: (a) a pair of (near) resonant coupling (Ω_c) and probe pulses (Ω_p) generate maximum coherence between states $|1\rangle$ and $|2\rangle$. From Harris *et al.*, 1997. (b) The coherently prepared medium then leads to efficient up-conversion of second coupling pulse Ω_c into the Ω_h. Also shown is the conversion efficiency from 425 to 293 nm as a function of coupling laser intensity. From Jain *et al.*, 1996.

wave mixing is shown in Fig. 19 (adapted from Jain *et al.*, 1996).

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 Λ system, Eq. (12), one recognizes that the polarization at ω_4 proportinal to ρ_{31} is a sum of two terms, the first being proportional to the linear susceptibility and the second to the nonlinear susceptibility. The drive fields E_1 and E_2 are now implicitly included in ρ_{12}, which has the maximal value of 0.5. These drive fields are resonant so the structure of $\chi^{(3)}$ simplifies to a single nonresonant term in the denominator. The nonlinear susceptibility therefore shares this single nonresonant term in the denominator with the linear susceptibility and so these two quantities are of comparable size. This is a very unusual situation in nonlinear 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 nonlinear 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 while the nonlinearity remains large.

In a molecular medium large coherence between vibrational or rotational levels has also been achieved using adiabatic pulse pairs in gas-phase hydrogen and deuterium (Sokolov *et al.*, 2000). Efficient multiorder Raman sideband 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 Fig. 17(iii). This superposition then mixes with the applied fields to form a broad spectrum of sidebands through multiorder 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 (infrared 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 sideband generation point the way to synthesizing very-short-duration light pulses, since the broadband Raman sideband 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-Λ systems

Four-wave mixing with all the optical fields close to resonance is achieved in atoms with four 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 (Buckle *et al.*, 1986; Arimondo, 1996). Experiments in the cw limit have been carried out in sodium vapor that show the phase sensitivity of EIT in a double-lambda scheme (Korsunsky *et al.*, 1999).

First we shall consider the case in which 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 or refraction of the generated field but also that in the applied fields themselves. The situation of four-wave mixing in a double-lambda system has been treated theoretically in detail

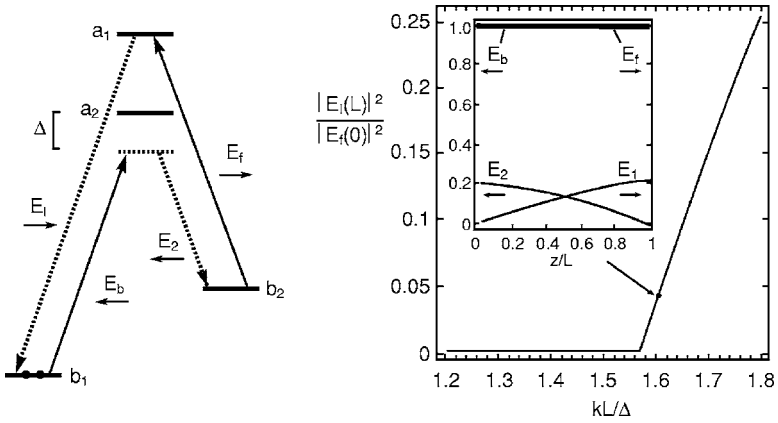

FIG. 20. Generation of fields E_1 and E_2 by resonant four-wave mixing of two counterpropagating pump fields E_f and E_b: left, level and coupling scheme; right, calculated threshold behavior of output intensity of field E_1 as a function of medium length L. The inset shows the spatial distributions of the fields above threshold of parametric oscillation at the indicated point. Adapted from Fleischhauer *et al.*, 2000.

by Korsunsky and Kosachiov (1999), who found the relative phases of the four fields to evolve with propagation so as to support 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 lossless conditions.

An experimental realization of up-conversion in a double-lambda scheme driven by three resonantly tuned pulsed fields, with the new field generated in the far UV at 186 nm, was achieved by Merriam *et al.* (2000) in lead vapor. High conversion efficiencies, more than 0.3 from the shortest-wavelength applied field at 233 nm to the 186 nm field, were indeed observed. The experiments confirm that the double-lambda scheme supports lossless 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 nonlinear 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 nonlinear 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 vapor the applied fields counterpropagate, and the resulting nonlinear gain and efficient intrinsic feedback in this configuration lead to mirrorless self-oscillation at extremely low applied field strengths (at the μW 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 nonclassical radiation (Lukin *et al.*, 1999; Fleischhauer, Lukin, *et al.*, 2000).

VI. EIT WITH FEW PHOTONS

In the absence of a strongly polarizable medium, photons are essentially noninteracting particles. In communications 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 have failed. For the latter, one needs strong, dissipation-free light-light interactions. In the present 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, for example, to a Kerr effect or, equivalently, a cross-phase modulation in which 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 π rad, 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 π rad using two light fields, each containing a single photon. In addition to new possibilities for addressing fundamental issues in quantum theory, such strong nondissipative interactions could be used to realize a controlled-NOT gate between two quantum bits (qubits), defined, for example, by the polarization state of a single-photon pulse. However, given the weakness of nonresonant optical nonlinearities and the dominant role of absorption in resonant processes, the combination of a large third-order Kerr susceptibility and small linear susceptibility appears to be incompatible.

In this section, we show how EIT can overcome these seemingly insurmountable obstacles. We have already seen how the nonreciprocity between linear and nonlinear susceptibilities in EIT media can be used to enhance

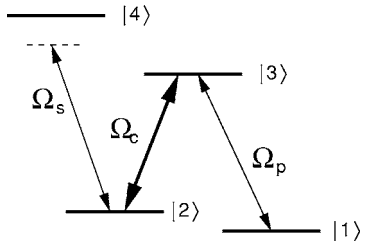

FIG. 21. Level scheme for giant Kerr effect.

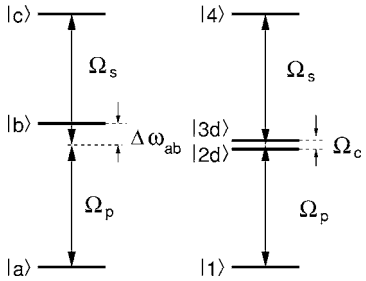

FIG. 22. Cross-phase modulation with and without giant Kerr effect: left, level configuration for cross-phase modulation in an ordinary three-level scheme; right, dressed-state representation of giant Kerr scheme.

generation of radiation at new frequencies. We shall see in this subsection how dissipation-free strong photon-photon interactions can be induced using EIT, due to a constructive interference in Kerr nonlinearity at two-photon resonance ($\delta=0$), where linear susceptibility experiences a complete cancellation. 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 ω_s is applied on this transition, with a Rabi frequency Ω_s. The cross-phase modulation nonlinearity that we are interested in is between this signal field and the probe field applied on the $|1\rangle$-$|3\rangle$ transition. We assume that the applied fields couple only to the respective transitions indicated in Fig. 21. Throughout this section, we assume that the coupling field with Rabi frequency Ω_c is nonperturbative, remains unchanged, and can be treated classically.

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

$$\begin{aligned} H_{\text{eff}} &= -\frac{\hbar}{2}[\Omega_p\sigma_{31}+\Omega_c\sigma_{32}+\Omega_s\sigma_{42}+\text{H.c.}] \\ &\quad +\hbar\left(\Delta-\frac{i}{2}\Gamma_3\right)\sigma_{33}+\hbar\left(\delta-\frac{i}{2}\Gamma_2\right)\sigma_{22} \\ &\quad +\hbar\left(\Delta\omega_{42}-\frac{i}{2}\Gamma_4\right)\sigma_{44} \end{aligned} \tag{78}$$

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 Γ_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 evaluate the linear and nonlinear components of the polarization at ω_p:

$$\begin{aligned} P(t) &= N\mu_{13}a_3(t)e^{-i\omega_p t}+\text{H.c.} \\ &= \epsilon_0\chi^{(1)}(-\omega_p,\omega_p)E_p e^{-i\omega_p t} \\ &\quad +\epsilon_0\chi^{(3)}(-\omega_p,\omega_s,-\omega_s,\omega_p)|E_s|^2E_p e^{-i\omega_p t}+\text{H.c.} \end{aligned} \tag{79}$$

Similarly, we can obtain the linear and nonlinear susceptibilities at ω_s. For simplicity, we consider only the resonant case in which $\Delta=\delta=0$.

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

$$\chi^{(3)}_{\text{xpm}}=\frac{|\mu_{13}|^2|\mu_{24}|^2\varrho}{2\epsilon_0\hbar^3}\frac{1}{\Omega_c^2}\left[\frac{1}{\Delta\omega_{42}}+i\frac{\Gamma_4}{2\Delta\omega_{42}^2}\right]. \tag{80}$$

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 ($\chi^{(3)}_{3-\text{level}}$) depicted in Fig. 22(a). For this system consisting of the ground state $|a\rangle$, an intermediate state of opposite parity $|b\rangle$, and a final state $|c\rangle$, we find

$$\text{Re}[\chi^{(3)}_{3-\text{level}}]=\frac{|\mu_{ab}|^2|\mu_{bc}|^2\varrho}{2\epsilon_0\hbar^3}\frac{1}{\Delta\omega_{ab}^2\Delta\omega_{bc}}, \tag{81}$$

where μ_{ij} and $\Delta\omega_{ij}$ denote the dipole matrix element of and the detuning from the transition $|i\rangle$-$|j\rangle$, respectively ($i,j=a,b,c$). When we compare the two expressions, we observe that they have practically identical forms: in the EIT cross-phase modulation scheme, $1/4\Delta\omega_{ab}^2$ is replaced by $1/\Omega_c^2$. Even though one needs to have $\Delta\omega_{ab}^2\geqslant\Gamma_b^2$ in a three-level scheme to avoid absorption, $\Omega_c^2\ll\Gamma_3^2$ is possible in the EIT cross-phase modulation 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).

We can understand the origin of this enhancement using the dressed-state picture depicted in Fig. 22(b). 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 interme-

diate states $|2d\rangle$ and $|3d\rangle$. The two paths contributing to virtual excitation of state $|4\rangle$ experience a constructive interference for ω_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 Ω_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 $\Omega_c^2 \ll \Gamma_3^2$. In this limit, small changes in two-photon detuning δ that are caused, for example, by a shift in the energy of state $|2\rangle$ can give rise to a drastic increase in $\mathrm{Re}[\chi^{(1)}]$ 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 ω_p that is proportional to Ω_s^2.

Finally, we note that by optical pumping and by choosing the polarization of the lasers appropriately, one can realize the energy-level diagram depicted in Fig. 21 practically for all alkali-metal atoms. It has been estimated that the enhancement of $\chi_{\mathrm{xpm}}^{(3)}$ 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 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 $\Delta\omega_{42}=0$ (Braje *et al.*, 2003); this is the purely dissipative limit of $\chi_{\mathrm{xpm}}^{(3)}$ 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 $\Omega_c^2 \ll \Gamma_3^2$, 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 $\Gamma_3\Gamma_2 \ll \Omega_c^2 \ll \Gamma_3^2$, $\Delta=0$ and concentrate on the limit of small $|\delta|$. For this system, we have a probe-field group velocity

$$v_p = \frac{\Omega_c^2}{\Gamma_3}\frac{1}{\varrho\sigma_{13}}, \tag{82}$$

where $\sigma_{13}=3\lambda_p^2/2\pi$ is the peak absorption cross section of the $|1\rangle$-$|3\rangle$ 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,

$$\tau_d = \frac{\Gamma_3}{\Omega_c^2}\varrho\sigma_{13}L, \tag{83}$$

referred to as the *group delay time* (Harris and Yamamoto, 1998; Harris and Hau, 1999), can be much longer than the traversal time of the signal field $T_s=L/c$ and the pulse widths of the probe (τ_p) and signal ($\tau_s=\tau_p$) 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 τ_p, i.e.,

$$L_h = v_p\tau_p = \frac{\Omega_c^2}{\Gamma_3}\frac{1}{\varrho\sigma_{13}}\tau_p, \tag{84}$$

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).

$$\begin{aligned}\frac{\partial E_p}{\partial z} + \left(\frac{1}{v_p}+\frac{1}{c}\right)\frac{\partial E_p}{\partial t} &= i\frac{\pi}{\lambda_p}\chi_{\mathrm{xpm}}^{(3)}\left|E_s\left(t-\frac{z}{c}\right)\right|^2 E_p \\ &= \left(\frac{i}{4(\Delta\omega_{42}+i\Gamma_4/2)v_p}\right) \\ &\quad\times\left|\Omega_s\left(t-\frac{z}{c}\right)\right|^2 E_p,\end{aligned} \tag{85}$$

where we have used Eq. (80).

When the group delay is negligible compared to τ_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

$$\phi_{\mathrm{xpm}} \to \left(\frac{\Gamma_4}{\Delta\omega_{42}+i\Gamma_4/2}\right)\frac{n_s}{A}\frac{\sigma_{24}}{2}\sqrt{\frac{\ln 2}{\pi}}\frac{\tau_d}{\tau_p}, \tag{86}$$

where n_s is the number of photons in the pulse and A its cross-sectional area. Equation (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

$$\phi_{\mathrm{xpm}} \to \left(\frac{\Gamma_4}{\Delta\omega_{42}+i\Gamma_4/2}\right)\frac{n_s}{A}\frac{\sigma_{24}}{8}. \tag{87}$$

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 Ω_c, ϱ, and L (Harris and Hau, 1999), and is on the order of 0.1 rad for

single-photon pulses focused into an area $A \sim \sigma_{24}$. This remarkably simple result assumes that $\Delta\omega_{42} \sim \Gamma_4/2$, 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 ultraslow 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 in which two different light pulses, centered at the two transparency frequencies, travel with the identical ultraslow 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 $|2\rangle$-$|4\rangle$ transition of species A. By choosing this pulse to be the signal pulse, one can realize a giant Kerr interaction between two ultraslow light pulses (Lukin and Imamoglu, 2000). An alternative scheme based on only a single atomic species was recently proposed by Petrosyan and Kuritzki (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

$$\phi_{\mathrm{xpm}} = \frac{\Gamma_4}{\Delta\omega_{42} + i\Gamma_4/2} \frac{n_s}{A} \frac{\sigma_{24}}{2} \sqrt{\frac{\ln 2}{\pi}} \frac{\tau_d}{\tau_p}, \tag{88}$$

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 $\tau_d \geqslant \tau_p$ (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 $\Delta\omega_{\mathrm{trans}} = \Omega_c^2/\Gamma_3\sqrt{\varrho\sigma_{13}L}$. If we set $\tau_p = \Delta\omega_{\mathrm{trans}}^{-1}$, we find

$$\frac{\tau_d}{\tau_p} \leqslant \sqrt{\varrho\sigma_{13}L}. \tag{89}$$

Since the other factors on the right-hand side of Eq. (88) can be of order unity, we can obtain $\phi_{\mathrm{xpm}} \geqslant \pi$, provided that we have $\varrho\sigma_{13}L \geqslant 10$. The 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 ϕ_{xpm}-bandwidth product that is independent of the optical depth (Lukin and Imamoglu, 2000).

Next, we examine the validity of this conclusion using a full quantum field-theoretic approach (Lukin and Imamoglu, 2000). To this end, we replace the classical electric fields describing the signal and probe fields with operators $\hat{E}_i(z,t) = \hat{E}_i^{(+)}(z,t) + \hat{E}_i^{(-)}(z,t)$, where

$$\hat{E}_i^{(+)}(z,t) = \sum_k \sqrt{\frac{\hbar kc}{2\epsilon_0 V}} \hat{a}_{ki}(z,t) e^{ik(z-ct)}. \tag{90}$$

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 degrees of freedom, we also need to impose a finite bandwidth Δ_q on 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 $\tau_p^{-1} < \Delta_q \ll \Delta\omega_{\mathrm{trans}}$ 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 to that given in Eq. (85), provided we replace c by v_s on the right-hand side. These equations can then be solved to yield

$$\begin{aligned}
\hat{E}_p^{(+)}(z,t) &= \hat{E}_p^{(+)}(0,t') \\
&\quad \times \exp\left[i\frac{\mu_{24}^2}{\hbar^2} \frac{\tau_d}{4\pi\Delta\omega_{42}} \hat{E}_s^{(-)}(0,t') \hat{E}_s^{(+)}(0,t') \right], \\
\hat{E}_s^{(+)}(z,t) &= \hat{E}_s^{(+)}(0,t') \\
&\quad \times \exp\left[i\frac{\mu_{13}^2}{\hbar^2} \frac{\tau_d}{4\pi\Delta\omega_{42}} \hat{E}_p^{(-)}(0,t') \hat{E}_p^{(+)}(0,t') \right],
\end{aligned} \tag{91}$$

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

$$|1_i\rangle = \sum_k \xi_k \hat{a}_{ki}^\dagger |0\rangle \tag{92}$$

for both fields, where we require $\Sigma_k |\xi_k|^2 = 1$ to ensure proper normalization. The spatiotemporal wave function of each pulse is given by the single-photon amplitude, $\Psi_i(z,t) = \langle 0|\hat{E}_i^{(+)}(z,t)|1_i\rangle$. To evaluate the correla-

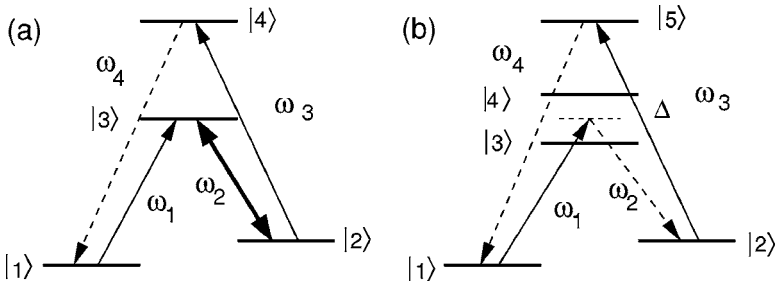

FIG. 23. Four-wave mixing schemes for the generation of (a) one field of frequency ω_4, and (b) two fields ω_2 and ω_4.

tions induced by the interaction, we concentrate on the two-photon amplitude

$$\Psi_{sp}(z,t;z',t') = \langle 0|\hat{E}_s^{(+)}(z,t)\hat{E}_p^{(+)}(z',t')|1_s,1_p\rangle. \tag{93}$$

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

$$\Psi_{sp}(z,t;z,t) = \Psi_s(0,t)\Psi_p(0,t)e^{i\phi}, \tag{94}$$

where $\phi=\tau_d\Delta_q\sigma_{24}\Gamma_4/4\pi A\Delta\omega_{42}$. Since the equal space and time commutation relations are proportional to Δ_q, there is explicit dependence on this quantity. The nonlinear phase shift ϕ is the counterpart of the classical phase ϕ_{xpm} obtained earlier in Eq. (88); the equivalence is obtained by replacing τ_p^{-1} with the quantization bandwidth Δ_q (Lukin and Imamoglu, 2000).

A large single-photon nonlinear phase shift ϕ that exceeds π rad 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 greater detail in the four-wave mixing schemes shown in Fig. 23. In the first scheme discussed by Harris and Hau (1999), photons from two pulses (ω_1) and (ω_3) are converted into an electromagnetic-field pulse at ω_4 in the presence of a strong monochromatic drive field at ω_2 [Fig. 23(a)]. In the second scheme [Fig. 23(b)], discussed by Johnsson *et al.* (2002) for copropagating pump fields and by Fleischhauer and Lukin (2000) and by Lukin *et al.* (1999) for counterpropagating pump fields, the field ω_2 is initially absent and spontaneously generated along with ω_4.

In the first scheme [see Fig. 23(a)], the input pulse ω_1 experiences a group delay due to EIT on the $|1\rangle$-$|3\rangle$ transition, while the second one, ω_3, moves almost with the vacuum speed of light. Harris and Hau showed that within an undepleted pump approximation the conversion efficiency, i.e., the ratio of the number of generated photons at ω_4 to the number of input photons at ω_1, is given by a dimensionless function $\Phi(\eta,\kappa L)$ multiplied by the number of input photons per atomic cross section:

$$\frac{n_4^{\mathrm{out}}}{n_1^{\mathrm{in}}} = \Phi(\eta,\kappa L)\frac{n_3^{\mathrm{in}}}{A_3}\sigma_{24}. \tag{95}$$

Here κ is the (E-field) absorption coefficient at the $|1\rangle$-$|4\rangle$ transition and L is the medium length. $\eta=L/L_h$ is the ratio of the length L_h 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 $\kappa L\to 0$ and of a small group delay within the medium $\eta\to 0$, the usual result of perturbative long-pulse nonlinear optics is obtained, which for Gaussian pulses reads

$$\Phi(\eta,\kappa L) = \left[\frac{\ln(2)}{\pi}\right]^{1/2}\frac{T_1}{\sqrt{T_1^2+T_3^2}}\eta\kappa L. \tag{96}$$

Here the T_μ's denote the temporal length of the pulses. In the limit of $\eta\to\infty$, i.e., if the group velocity reduction of ω_1 is large, so that the ω_3 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 η^{-1}. A maximum of Φ 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 ten photons per atomic cross section are sufficient to obtain perfect conversion.

In the second scheme, where the field ω_2 is generated [see Fig. 23(b)], a finite detuning from level $|3\rangle$ is needed to suppress absorption. Furthermore, in order to cancel ac-Stark shifts the frequencies ω_1 and ω_2 can be tuned midway between two states with appropriate transition matrix elements (Johnsson *et al.*, 2002). Efficient frequency conversion (Hemmer *et al.*, 1994; Ham, Shariar, *et al.*, 1997; Babin *et al.*, 1999; Hinze *et al.*, 1999), generation of squeezing (Lukin *et al.*, 1999), as well as the possibility of mirrorless oscillations (in counterpropagating geometry) have been predicted (Lukin, Hemmer, *et al.*, 1998) and in part experimentally observed (Zibrov *et al.*, 1999). It has been shown by Fleischhauer and Lukin

(2000) that an extremely low input photon flux $\phi_{\rm thr}$ is sufficient to reach the threshold of mirrorless oscillations:

$$\phi_{\rm thr} = fN\gamma_{12}, \tag{98}$$

where N is the number of atoms in the beam path and γ_{12} is the coherence decay rate of the $|1\rangle$-$|2\rangle$ transition. f is a numerical prefactor 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):

$$H_{\rm int} = \frac{\hbar g c}{2\Delta}\int dz \frac{\hat{\Omega}_1^\dagger \hat{\Omega}_3^\dagger \hat{\Omega}_2 \hat{\Omega}_4 + \text{H.c.}}{\hat{\Omega}_3^\dagger \hat{\Omega}_3 + \hat{\Omega}_4^\dagger \hat{\Omega}_4}, \tag{99}$$

where $\hat{\Omega}_\mu$ are the operators corresponding to the (complex) Rabi frequencies of the fields and where a common coupling constant $g=\varrho\mu^2\omega/2\hbar\varepsilon_0 c$ of all transitions was assumed. One recognizes that, in contrast to the case of ordinary nonresonant nonlinear optics, the sum of the intensities of the resonant fields appears in the denominator. As a consequence, in a copropagating geometry, the conversion length, i.e., the length after which a pair of photons in the input fields ω_1 and ω_3 is completely converted into a pair of photons in the fields, ω_2 and ω_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 very recently that the first experimental demonstrations of the predicted effects have been seen. Kuzmich and co-workers (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 the presence of high-finesse cavities can be used to enhance photon-photon interactions, for example, for the purpose of quantum computation (Nielsen 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 shall show in this 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 $|2\rangle$ and $|3\rangle$. As shown by Field (1993), electromagnetically induced transparency on the probe transition is already possible 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

$$H_{\rm int} = -\hbar\frac{\Omega_p}{2}|3\rangle\langle 1| - \hbar\frac{g}{2}\hat{a}|3\rangle\langle 2| + \text{H.c.} \tag{100}$$

separates into effective three-level systems,

$$|1,n\rangle \overset{\Omega_p}{\leftrightarrow} |3,n\rangle \overset{g\sqrt{n+1}}{\leftrightarrow} |2,n+1\rangle, \tag{101}$$

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 $n=0$. Equation (101) suggests another interesting application. The effective three-level systems have dark states. For example,

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

with $\tan\theta(t)=\Omega_p(t)/g$. 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 was first pointed out by Parkins *et al.* (1993). Law and Eberly proposed an application for the generation of a single photon in a well-defined wave packet (Law and Eberly, 1996). Recently such a "photon pistol" was experimentally realized (Hennrich *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.,

$$H_{\rm int} = -\hbar\frac{g}{2}\hat{a}|3\rangle\langle 1| - \hbar\frac{\Omega_c}{2}|3\rangle\langle 2| + \text{H.c.} \tag{103}$$

and we have the coupling

$$|1,n\rangle \overset{g\sqrt{n}}{\leftrightarrow} |3,n-1\rangle \overset{\Omega_c(t)}{\leftrightarrow} |2,n-1\rangle. \tag{104}$$

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.* (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 be partly met 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

$$H_{\rm int} = -\frac{\hbar}{2}\sum_{j=1}^{N}[g\hat{a}|3\rangle_{jj}\langle 1| + \hbar\Omega_c|3\rangle_{jj}\langle 2| + \text{H.c.}] \tag{105}$$

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

$$|\mathbf{1}^N\rangle \equiv |1,\ldots,1\rangle,$$

$$|\mathbf{1}^{N-1}\mathbf{2}\rangle \equiv \frac{1}{\sqrt{N}}\sum_{j=1}^{N}|1,\ldots,2_j,\ldots,1\rangle,$$

$$|\mathbf{1}^{N-2}\mathbf{2}^2\rangle \equiv \frac{1}{\sqrt{2N(N-1)}}\sum_{i\neq j=1}^{N}|1,\ldots,2_i,\ldots,2_j,\ldots,1\rangle,$$

$$\text{etc.} \tag{106}$$

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

$$|\mathbf{1}^N,1\rangle \overset{g\sqrt{N}}{\leftrightarrow} |\mathbf{1}^{N-1}\mathbf{3},0\rangle \overset{\Omega_c(t)}{\leftrightarrow} |\mathbf{1}^{N-1}\mathbf{2},0\rangle. \tag{107}$$

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

$$g \rightarrow g\sqrt{N}. \tag{108}$$

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 $g^2 \gg \kappa\Gamma_3$, where κ 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, Yelin, and Fleischhauer, 2000).

As discussed in previous subsections, EIT can be used to achieve a resonant enhancement of nonlinear interactions leading, for example, 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. Imamoglu *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 could be employed, for example, to build a controllable single-photon source or a controlled-phase gate (Duan and Kimble, 2004). Imamoglu *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 multiatom EIT systems (Werner and Imamoglu, 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 a vanishingly small photon blockade effect. The survival of nonlinearity in $N \gg 1$ atom cavity EIT is related to the reduction of the cavity linewidth, or equivalently the width of the atom cavity dark state (Lukin, Fleischhauer, *et al.*, 1998; Werner and Imamoglu, 1999).

VII. SUMMARY AND PERSPECTIVES

EIT has had a profound impact 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 that 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 14 years on from the first experimental demonstration of the phenomenon of EIT. Much research activity has been undertaken in the period since this discovery, in an effort 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, and has remained a healthy and active subject with no sign of declining activity. There continue to be new and interesting proposals and demonstrations that extend EIT to fresh applications. It has become a standard tool for the study of 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. Some omissions were inevitable. Much work has 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 would support further, more detailed, explorations of this topic.

Where possible we have tried to connect the theory with the results of real experiments. Again we have been selective, choosing illustrative examples rather than attempting a comprehensive review of all work. Because of space constraints we have not explored in any structured way the experimental techniques used in these experiments, either to achieve robust EIT or to investigate EIT phenomena. This omission is regrettable, but the situations in which EIT can be observed are too diverse to be covered in depth here. The reader is urged to consult the cited literature and where necessary other reviews and textbooks to obtain a fuller understanding of the 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 nonlinear 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, the possibility of using laser cooling, and high thresholds against optical damage. Moreover, a problem normally identified with gas-phase nonlinear optical media, the low value of the nonlinear susceptibility compared to many solid-density optical crystals, is circumvented by EIT since it permits the use of resonance to enhance the susceptibility.

Having reviewed the subject and examined a number of applications that have already been thoroughly investigated, we now turn to the future. While predictions are always risky, we offer some thoughts on what new applications are likely to emerge in this field in the hope of stimulating further ideas. We shall 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 100-nm frequency range. By careful choice of the atomic system and through judicious optimization 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 nanosecond pulses (spectral linewidths of $\sim$100 MHz) of VUV radiation with energies of tens of microjoules. These could be of use in a number of applications in nonlinear 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.

While 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 with Raman-like excitations: a limit to the degree of up-conversion as the initial excitation is via difference-frequency mixing of the applied fields. The inherent limit is set at around 170 nm 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 is how to excite maximal coherence between the ground state and a very highly excited upper state. One approach is to 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 extreme-UV 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, for example, in a quantum computer, is quite difficult. The reason for this is the weakness of photon-photon interactions in the 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 has still not been explored, especially on the experimental side.

Another feature of EIT, which makes it very attractive for future applications in quantum information, is the possibility of transferring coherence and quantum states from light to collective atomic spin excitations. Quantum memories for photons (Lukin, Yelin, and Fleischhauer, 2000; Fleischhauer and Lukin, 2001), 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 for generating photon wave packets with tai-

lored quantum states or for manipulating these states. A first proposal in this direction using a dipole blockade of Rydberg excitations was given by 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 quasiparticle, 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 ultracold 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).

Ultrafast measurements have recently entered the attosecond domain (Drescher *et al.*, 2002). The possibility of employing the highly efficient multiorder Raman generation that results from an adiabatically prepared maximal vibrational coherence has already been investigated. A key problem to solve before this approach can be widely employed for ultrafast measurements will be the synchronization of the train of subfemtosecond pulses that are generated with external events. One possibility (suggested by Sokolov, 2002) is to use the pulse train to "measure" processes in the modulated molecules themselves so that the synchronization is automatically satisfied.

As we have emphasized throughout this review, 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 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 on subnanosecond time scales. 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 1 μs in optically active direct-band-gap bulk semiconductors such as GaAs (Wolf *et al.*, 2001) to 1 ms 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 could 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, Hemmer, and Shahiar, 1997; Turukhin *et al.*, 2001) and nitrogen vacancy centers in diamond (Wei and Manson, 1999). In experiments carried out using Pr-doped Y_2SiO_5 at cryogenic temperatures ($T=5$ K), Turukhin and co-workers have observed group velocities as slow as 45 m/s and a group delay of 66 μs for light pulses with a pulse width of 50 μs (Turukhin *et al.*, 2001). Storage and retrieval of these pulses have also been demonstrated (Turukhin *et al.*, 2001). While the ratio of the group delay to the pulse width in these experiments is on the order of unity, this is an impressive and promising development for solid-state EIT.

Optoelectronics and photonics technology are largely based on semiconductor heterostructures formed out of column-III–V semiconductors. The conduction-band electrons of these elements have predominantly *s*-type wave functions with small spin-orbit coupling. As a result, the spin coherence times are four 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 ($B\sim 10$ T), 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; Zutic *et al.*, 2004), and it is plausible to argue that EIT could play a role by providing long optical delays and/or enabling "spin-photon" information transfer.

Yet another interesting extension of gas-phase EIT to plasmas was suggested by Harris (1996). Here classical interference effects in parametric wave interactions which are analogous to interference effects in three-level atoms lead to an induced transparency window below the plasma frequency. This could have interesting applications for magnetic fusion, plasma heating, and plasma diagnostics. For some recent work on this see, Litvak and Tokman (2002) and Shvets and Wurtele (2002).

Even though EIT is by now a well-established technique, new applications continue to appear regularly. It was recently proposed, for example, that a "dynamical photonic band-gap" structure could be realized in an EIT medium by spatially modifying the refractive index with a standing-wave optical field (Andre and Lukin, 2002). This could be achieved by making use of the large absorption-free dispersion around the transparency frequency. EIT may also allow for the realization of a high-efficiency photon counter. After mapping the quantum state of a propagating light pulse onto collective hyperfine excitations of a trapped cold atomic gas, it is pos-

sible 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 should be possible to achieve photon counting with an efficiency exceeding 99%. Such a device could be of great use in quantum optical information processing (Knill *et al.*, 2001).

ACKNOWLEDGMENTS

We would like to express our gratitude to Steve Harris, who has been a source of inspiration through his innovative ideas, insights, and encouragement. Our numerous discussions and collaborations with Klaas Bergmann, Peter Knight, Misha Lukin, Marlan Scully, Moshe Shapiro, and Bruce Shore have played a key role in shaping our understanding of various aspects of EIT. A.I. would also like to thank John Field for truly enlightening discussions dating from the very early days of EIT. Finally, we acknowledge K. Boller, K. Hakuta, L. Hau, O. Kocharovskaya, and H. Schmidt.

REFERENCES

Affolderbach, C., M. Stahler, S. Knappe, and R. Wynands, 2002, Appl. Phys. B: Lasers Opt. **75**, 605.
Agarwal, G. S., 1993, Phys. Rev. Lett. **71**, 1351.
Alzetta, G., A. Gozzini, L. Moi, and G. Orriols, 1976, Nuovo Cimento Soc. Ital. Fis., B **36**, 5.
Andre, A., and M. Lukin, 2002, Phys. Rev. Lett. **89**, 143602.
Arimondo, E., 1996, Prog. Opt. **35**, 259.
Arimondo, E., and G. Orriols, 1976, Lett. Nuovo Cimento Soc. Ital. Fis. **17**, 333.
Armstrong, J. A., and J. J. Wynne, 1974, Phys. Rev. Lett. **33**, 1183.
Armstrong, Lloyd, Jr., and Brian Lee Beers, 1975, Phys. Rev. Lett. **34**, 1290.
Autler, S. H., and C. H. Townes, 1955, Phys. Rev. **100**, 703.
Babin, S., U. Hinze, E. Tiemann, and B. Wellegehausen, 1996, Opt. Lett. **21**, 1186.
Babin, S. A., E. V. Podivilov, D. A. Shapiro, U. Hinze, E. Tiemann, and B. Wellegehausen, 1999, Phys. Rev. A **59**, 1355.
Bajcsy, M., A. S. Zibrov, and M. D. Lukin, 2003, Nature (London) **426**, 638.
Bergmann, K., H. Theuer, and B. W. Shore, 1998, Rev. Mod. Phys. **70**, 1003.
Boller, K. J., A. Imamoglu, and S. E. Harris, 1991, Phys. Rev. Lett. **66**, 2593.
Bol'shov, L. A., and V. V. Likhanskii, 1985, Sov. J. Quantum Electron. **15**, 889.
Braje, D. A., V. Balic, G. Y. Yin, and S. E. Harris, 2003, Phys. Rev. A **68**, 041801(R).
Buckle, S. J., S. M. Barnett, P. L. Knight, M. A. Lauder, and D. T. Pegg, 1986, Opt. Acta **33**, 2473.
Budker, D., D. F. Kimball, S. M. Rochester, and V. V. Yashchuk, 1999, Phys. Rev. Lett. **83**, 1767.
Budker, D., V. Yashchuk, and M. Zolotorev, 1998, Phys. Rev. Lett. **81**, 5788.
Buffa, R., M. P. Anscombe, and J. P. Marangos, 2003, Phys. Rev. A **67**, 033801.
Carmichael, H. J., 1993, *An Open Systems Approach to Quantum Optics: Lectures Presented at the Université Libre de Bruxelles, October 28 to November 4, 1991* (Springer, Berlin).
Cerboneschi, E., and E. Arimondo, 1995, Phys. Rev. A **52**, R1823.
Cirac, J. I., P. Zoller, H. J. Kimble, and H. Mabuchi, 1997, Phys. Rev. Lett. **78**, 3221.
Cohen-Tannoudji, C., J. Dupont-Roc, and G. Grynberg, 1992, *Atom-Photon Interactions* (Wiley, New York).
Csesznegi, J. R., and R. Grobe, 1997, Phys. Rev. Lett. **79**, 3162.
Dalibard, J., Y. Castin, and K. Molmer, 1992, Phys. Rev. Lett. **68**, 580.
Dalton, B. J., and P. L. Knight, 1982, Opt. Commun. **41**, 411.
Dorman, C., I. Kucukkara, and J. P. Marangos, 2000a, Opt. Commun. **180**, 263.
Dorman, C., I. Kucukkara, and J. P. Marangos, 2000b, Phys. Rev. A **61**, 013802.
Drescher, M., M. Hentschel, R. Kienberger, M. Uiberacker, V. Yakovlev, A. Scrinizi, T. Westerwalbesloh, U. Kleineberg, U. Heinzmann, and F. Krausz, 2002, Nature (London) **419**, 803.
Duan, L. M., J. I. Cirac, and P. Zoller, 2002, Phys. Rev. A **66**, 023818.
Duan, L. M., J. I. Cirac, P. Zoller, and E. S. Polzik, 2000, Phys. Rev. Lett. **85**, 5643.
Duan, L. M., and H. J. Kimble, 2004, Phys. Rev. Lett. **92**, 127902.
Duan, L. M., M. D. Lukin, J. I. Cirac, and P. Zoller, 2001, Nature (London) **414**, 413.
Dutton, Z., M. Budde, C. Slowe, and L. V. Hau, 2001, Science **293**, 5530.
Eberly, J. H., M. L. Pons, and H. R. Haq, 1994, Phys. Rev. Lett. **72**, 56.
Faist, J., F. Capasso, C. Sirtori, K. W. West, and L. Pfeiffer, 1997, Nature (London) **390**, 589.
Fano, U., 1961, Phys. Rev. **124**, 1866.
Fano, U., and J. Cooper, 1965, Phys. Rev. **137**, 1364.
Field, J. E., 1993, Phys. Rev. A **47**, 5064.
Field, J. E., K. Hahn, and S. E. Harris, 1991, Phys. Rev. Lett. **67**, 3733.
Fleischhauer, M., 1994, Phys. Rev. Lett. **72**, 989.
Fleischhauer, M., 1999, Opt. Express **4**, 107.
Fleischhauer, M., C. H. Keitel, M. O. Scully, and C. Su, 1992a, Opt. Commun. **87**, 238.
Fleischhauer, M., C. H. Keitel, M. O. Scully, C. Su, B. T. Ulrich, and S. Y. Zhu, 1992b, Phys. Rev. A **46**, 1468.
Fleischhauer, M., and M. D. Lukin, 2000, Phys. Rev. Lett. **84**, 5094.
Fleischhauer, M., and M. D. Lukin, 2001, Phys. Rev. A **64**, 022314.
Fleischhauer, M., M. D. Lukin, A. B. Matsko, and M. O. Scully, 2000, Phys. Rev. Lett. **84**, 3558.
Fleischhauer, M., and A. S. Manka, 1996, Phys. Rev. A **54**, 794.
Fleischhauer, M., and C. Mewes, 2002, in *Proceedings of the International School of Physics "Enrico Fermi": Experimental Quantum Computation and Information*, edited by F. De Martini and C. Monroe (IOS, Amsterdam/Washington, D.C.), pp. 511–529.
Fleischhauer, M., and M. O. Scully, 1994, Phys. Rev. A **49**, 1973.
Fry, E. S., X. Li, D. Nikonov, G. G. Padmabandu, M. O. Scully, A. V. Smith, F. K. Tittel, C. Wang, S. R. Wilkinson, and S.-Y. Zhu, 1993, Phys. Rev. Lett. **70**, 3235.
Gao, J. C., C. Guo, G. Jin, P. Wang, J. Zhao, H. Zhang, Y. Jiang, D. Wang, and D. Jiang, 1992, Opt. Commun. **93**, 323.

Gardiner, C. W., A. S. Parkins, and P. Zoller, 1992, Phys. Rev. A **46**, 4363.
Garton, W. R. S., 1966, Adv. At. Mol. Phys. **2**, 93.
Gaubatz, U., P. Rudecki, S. Schiemman, and K. Bergmann, 1990, J. Chem. Phys. **92**, 5363.
Gheri, K. M., W. Alge, and P. Grangier, 1999, Phys. Rev. A **60**, R2673.
Gornyi, M. B., B. G. Matisov, and Y. V. Rozhdestvenskii, 1989, Sov. Phys. JETP **68**, 728.
Gray, H. R., R. M. Whitley, and C. R. Stroud, 1978, Opt. Lett. **3**, 218.
Grigoryan, G. G., and Y. T. Pashayan, 2001, Phys. Rev. A **64**, 013816.
Grobe, R., F. T. Hioe, and J. H. Eberly, 1994, Phys. Rev. Lett. **73**, 3183.
Hahn, K. H., D. A. King, and S. E. Harris, 1990, Phys. Rev. Lett. **65**, 2777.
Hakuta, K., 2004, private communication.
Hakuta, K., M. Suzuki, M. Katsuragawa, and J. Z. Li, 1997, Phys. Rev. Lett. **79**, 209.
Hald, J., J. L. Sorensen, C. Schori, and E. S. Polzik, 1999, Phys. Rev. Lett. **83**, 1319.
Ham, B. S., P. R. Hemmer, and M. S. Shahiar, 1997, Opt. Commun. **144**, 227.
Ham, B. S., M. S. Shariar, M. K. Kim, and P. R. Hemmer, 1997, Opt. Lett. **22**, 1849.
Hänsch, T. W., and P. Toschek, 1970, Z. Phys. **236**, 213.
Harris, S. E., 1989, Phys. Rev. Lett. **62**, 1033.
Harris, S. E., 1993, Phys. Rev. Lett. **70**, 552.
Harris, S. E., 1994a, Phys. Rev. Lett. **72**, 52.
Harris, S. E., 1994b, Opt. Lett. **19**, 2018.
Harris, S. E., 1996, Phys. Rev. Lett. **77**, 5357.
Harris, S. E., 1997, Phys. Today **50** (7), 36.
Harris, S. E., 2000, Phys. Rev. Lett. **85**, 4032.
Harris, S. E., J. E. Field, and A. Imamoglu, 1990, Phys. Rev. Lett. **64**, 1107.
Harris, S. E., J. E. Field, and A. Kasapi, 1992, Phys. Rev. A **46**, R29.
Harris, S. E., and L. V. Hau, 1999, Phys. Rev. Lett. **82**, 4611.
Harris, S. E., and M. Jain, 1997, Opt. Lett. **22**, 636.
Harris, S. E., and Z.-F. Luo, 1995, Phys. Rev. A **52**, R928.
Harris, S. E., and A. V. Sokolov, 1997, Phys. Rev. A **55**, R4019.
Harris, S. E., and Y. Yamamoto, 1998, Phys. Rev. Lett. **81**, 3611.
Harris, S. E., G. Y. Yin, M. Jain, H. Xia, and A. J. B. Merriam, 1997, Philos. Trans. R. Soc. London, Ser. A **355**, 2291.
Hau, L. V., S. E. Harris, Z. Dutton, and C. H. Behroozi, 1999, Nature (London) **397**, 594.
Hemmer, P. R., K. Z. Cheng, J. Kierstead, M. S. Shariar, and M. K. Kim, 1994, Opt. Lett. **19**, 296.
Hemmer, P. R., D. P. Katz, J. Donoghue, M. Cronin-Golomb, M. S. Shariar, and P. Kumar, 1995, Opt. Lett. **20**, 769.
Hennrich, M., T. Legero, A. Kuhn, and G. Rempe, 2000, Phys. Rev. Lett. **85**, 4872.
Hinze, U., L. Meyer, B. N. Chichkov, E. Tiemann, and B. Wellegehausen, 1999, Opt. Commun. **166**, 127.
Hioe, F. T., and R. Grobe, 1994, Phys. Rev. Lett. **73**, 2559.
Imamoglu, A., 1991, Ph.D. thesis (Stanford University).
Imamoglu, A., 2000, Opt. Commun. **179**, 179.
Imamoglu, A., 2002, Phys. Rev. Lett. **89**, 163602.
Imamoglu, A., and S. E. Harris, 1989, Opt. Lett. **14**, 1344.
Imamoglu, A., H. Schmidt, G. Woods, and M. Deutsch, 1997, Phys. Rev. Lett. **79**, 1467.
Jain, M., 1994, Phys. Rev. A **50**, 1899.
Jain, M., A. J. Merriam, A. Kasapi, G. Y. Yin, and S. E. Harris, 1995, Phys. Rev. Lett. **75**, 4385.
Jain, M., H. Xia, G. Y. Yin, A. J. Merriam, and S. E. Harris, 1996, Phys. Rev. Lett. **77**, 4326.
James, D. F. V., and P. Kwiat, 2002, Phys. Rev. Lett. **89**, 183601.
Johnsson, M., E. Korsunsky, and M. Fleischhauer, 2002, Opt. Commun. **212**, 335.
Johnsson, M. T., and M. Fleischhauer, 2002, Phys. Rev. A **66**, 043808.
Julsgaard, B., A. Kozhekin, and E. S. Polzik, 2001, Nature (London) **413**, 400.
Juzeliunas, G., and H. J. Carmichael, 2002, Phys. Rev. A **65**, 021601(R).
Juzeliunas, G., M. Masalas, and M. Fleischhauer, 2003, Phys. Rev. A **67**, 023809.
Kasapi, A., M. Jain, G. Y. Yin, and S. E. Harris, 1995, Phys. Rev. Lett. **74**, 2447.
Kasapi, A., G. Y. Yin, M. Jain, and S. E. Harris, 1996, Phys. Rev. A **53**, 4547.
Kash, M. M., V. A. Sautenkov, A. S. Zibrov, L. Hollberg, G. R. Welch, M. D. Lukin, Y. Rostovtsev, E. S. Fry, and M. O. Scully, 1999, Phys. Rev. Lett. **82**, 5229.
Kien, F. E., J. Q. Liang, M. Katsuragawa, K. Ohtsuki, K. Hakuta, and A. V. Sokolov, 1999, Phys. Rev. A **60**, 1562.
Knight, P. L., 1990, Phys. Rep. **190**, 1.
Knill, E., R. Laflamme, and G. J. Milburn, 2001, Nature (London) **409**, 46.
Kocharovskaya, O., 1992, Phys. Rep. **219**, 175.
Kocharovskaya, O., Y. Rostovtsev, and M. O. Scully, 2001, Phys. Rev. Lett. **86**, 628.
Kocharovskaya, O. A., and Y. I. Khanin, 1986, Sov. Phys. JETP **63**, 945.
Kocharovskaya, O. A., and Y. I. Khanin, 1988, JETP Lett. **48**, 630.
Konopnicki, M. J., P. D. Drummond, and J. H. Eberly, 1981, Opt. Commun. **36**, 313.
Konopnicki, M. J., and J. H. Eberly, 1981, Phys. Rev. A **24**, 2567.
Korsunsky, E. A., and D. V. Kosachiov, 1999, Phys. Rev. A **60**, 4996.
Korsunsky, E. A., N. Leinfellner, A. Huss, S. Baluschev, and L. Windholz, 1999, Phys. Rev. A **59**, 2302.
Kozhekin, A. E., K. Molmer, and E. Polzik, 2000, Phys. Rev. A **62**, 033809.
Kozlov, V. V., S. Wallentowitz, and S. Raghavan, 2002, Phys. Lett. A **296**, 210.
Kozuma, M., D. Akasmatsu, L. Denf, E. W. Hagley, and M. G. Payne, 2002, Phys. Rev. A **66**, 031801(R).
Kuhn, A., M. Hennrich, and G. Rempe, 2002, Phys. Rev. Lett. **89**, 067901.
Kuklinski, J. R., U. Gaubatz, T. F. Hioe, and K. Bergmann, 1989, Phys. Rev. A **40**, 6741.
Kuzmich, A., W. P. Bowen, A. D. Boozer, A. Bocha, C. W. Chou, L. M. Duan, and H. J. Kimble, 2003, Nature (London) **423**, 731.
Kuzmich, A., L. Mandel, and N. P. Bigelow, 2000, Phys. Rev. Lett. **85**, 1594.
Kuzmich, A., L. Mandel, J. Janis, Y. E. Young, R. Ejnisman, and N. P. Bigelow, 1999, Phys. Rev. A **60**, 2346.
Kuzmich, A., K. Molmer, and E. S. Polzik, 1997, Phys. Rev. Lett. **79**, 4782.
Kuznetsova, E., O. Kocharovskaya, P. Hemmer, and M. O.

Scully, 2002, Phys. Rev. A **66**, 063802.
Law, C. K., and J. H. Eberly, 1996, Phys. Rev. Lett. **76**, 1055.
Leonhardt, U., and P. Piwnicki, 1999, Phys. Rev. A **60**, 4301.
Leonhardt, U., and P. Piwnicki, 2000a, Phys. Rev. Lett. **84**, 822.
Leonhardt, U., and P. Piwnicki, 2000b, Phys. Rev. Lett. **85**, 5253.
Litvak, A., and M. D. Tokman, 2002, Phys. Rev. Lett. **88**, 095003.
Liu, C., Z. Dutton, C. H. Behroozi, and L. V. Hau, 2001, Nature (London) **409**, 490.
Lounis, B., and C. Cohen-Tannoudji, 1992, J. Phys. II **2**, 579.
Lukin, M. D., 2003, Rev. Mod. Phys. **75**, 457.
Lukin, M. D., M. Fleischhauer, R. Cote, L. M. Duan, D. Jaksch, J. I. Cirac, and P. Zoller, 2001, Phys. Rev. Lett. **87**, 037901.
Lukin, M. D., M. Fleischhauer, M. O. Scully, and V. L. Velichansky, 1998, Opt. Lett. **23**, 295.
Lukin, M. D., M. Fleischhauer, A. S. Zibrov, H. G. Robinson, V. L. Velichansky, L. Hollberg, and M. O. Scully, 1997, Phys. Rev. Lett. **79**, 2959.
Lukin, M. D., P. Hemmer, and M. O. Scully, 2000, Adv. At., Mol., Opt. Phys. **42**, 347.
Lukin, M. D., P. R. Hemmer, M. Löffler, and M. O. Scully, 1998, Phys. Rev. Lett. **81**, 2675.
Lukin, M. D., and A. Imamoglu, 2000, Phys. Rev. Lett. **84**, 1419.
Lukin, M. D., and A. Imamoglu, 2001, Nature (London) **273**, 273.
Lukin, M. D., A. B. Matsko, M. Fleischhauer, and M. O. Scully, 1999, Phys. Rev. Lett. **82**, 1847.
Lukin, M. D., S. F. Yelin, and M. Fleischhauer, 2000, Phys. Rev. Lett. **84**, 4232.
Madden, R. B., and K. Codling, 1965, Astrophys. J. **141**, 364.
Mair, A., J. Hager, D. F. Phillips, R. L. Walsworth, and M. D. Lukin, 2002, Phys. Rev. A **65**, 031802(R).
Malmistov, A. I., 1984, Sov. J. Quantum Electron. **14**, 385.
Mandel, P., 1993, Contemp. Phys. **34**, 235.
Masalas, M., and M. Fleischhauer, 2004, Phys. Rev. A **69**, 061801(R).
Matsko, A. B., O. Kocharovskaya, Y. Rostovtsev, A. S. Z. G. R. Welch, and M. O. Scully, 2001, Adv. At., Mol., Opt. Phys. **46**, 191.
Matsko, A. B., Y. V. Rostovtsev, M. Fleischhauer, and M. O. Scully, 2001, Phys. Rev. Lett. **86**, 2006.
Matsko, A. B., Y. V. Rostovtsev, O. Kocharovskaya, A. S. Zibrov, and M. O. Scully, 2001, Phys. Rev. A **64**, 043809.
Mazets, I. E., and B. G. Matisov, 1996, JETP Lett. **64**, 515.
Merriam, A. J., S. J. Sharpe, M. Shverdin, D. Manuszak, G. Y. Yin, and S. E. Harris, 2000, Phys. Rev. Lett. **84**, 5308.
Merriam, A. J., S. J. Sharpe, H. Xia, D. Manuszak, G. Y. Yin, and S. E. Harris, 1999, Opt. Lett. **24**, 625.
Mewes, C., and M. Fleischhauer, 2002, Phys. Rev. A **66**, 033820.
Mewes, C., and M. Fleischhauer, 2005, Phys. Rev. A (to be published).
Moiseev, S. A., and N. S. Kroll, 2001, Phys. Rev. Lett. **87**, 173601.
Moseley, R. R., S. Shepherd, D. J. Fulton, B. D. Sinclair, and M. H. Dunn, 1995, Phys. Rev. Lett. **74**, 670.
Müller, G., M. Müller, A. Wicht, R.-H. Rinkleff, and K. Danzmann, 1997, Phys. Rev. A **56**, 2385.
Nagel, A., L. Graf, A. Naumov, E. Mariotti, V. Biancalana, D. Meschede, and R. Wynands, 1998, Europhys. Lett. **44**, 31.
Nielsen, M., and I. Chuang, 2000, *Quantum Computation and Quantum Information* (Cambridge University Press, Cambridge, England).
Nottlemann, A., C. Peters, and W. Lange, 1993, Phys. Rev. Lett. **70**, 1783.
Oreg, J., F. T. Hioe, and J. H. Eberly, 1984, Phys. Rev. A **29**, 690.
Padmabandu, G. G., G. R. Welch, I. N. Shubin, E. S. Fry, D. E. Nikonov, M. D. Lukin, and M. O. Scully, 1996, Phys. Rev. Lett. **76**, 2053.
Parkins, A. S., P. Marte, P. Zoller, O. Carnal, and H. J. Kimble, 1995, Phys. Rev. A **51**, 1578.
Parkins, A. S., P. Marte, P. Zoller, and H. J. Kimble, 1993, Phys. Rev. Lett. **71**, 3095.
Pellizzari, T., S. A. Gardiner, J. I. Cirac, and P. Zoller, 1995, Phys. Rev. Lett. **75**, 3788.
Petch, J. C., C. H. Keitel, P. L. Knight, and J. P. Marangos, 1996, Phys. Rev. A **53**, 543.
Petrosyan, D., and G. Kurizki, 2002, Phys. Rev. A **65**, 033833.
Phillips, D., A. Fleischhauer, A. Mair, R. Walsworth, and M. D. Lukin, 2001, Phys. Rev. Lett. **86**, 783.
Polzik, E. S., 1999, Phys. Rev. A **59**, 4202.
Rahman, A., 1999, Phys. Rev. A **60**, 4187.
Rahman, A., and J. H. Eberly, 1998, Phys. Rev. A **58**, R805.
Rathe, U., M. Fleischhauer, S. Y. Zhu, T. W. Hänsch, and M. O. Scully, 1993, Phys. Rev. A **47**, 4994.
Reintjes, J. F., 1984, *Nonlinear Optical Parametric Processes in Liquids and Gases* (Elsevier Science/Academic, Boston).
Rickes, T., J. P. Marangos, and T. Halfmann, 2003, Opt. Commun. **277**, 133.
Rickes, T., L. P. Yatsenko, S. Steuerwald, T. Halfmann, B. W. Shore, N. V. Vitanov, and K. Bergmann, 2000, J. Chem. Phys. **113**, 534.
Ruostekoski, J., and D. F. Walls, 1999a, Eur. Phys. J. D **5**, 335.
Ruostekoski, J., and D. F. Walls, 1999b, Phys. Rev. A **59**, R2571.
Sautenkov, V. A., M. D. Lukin, C. J. Bednar, I. Novikova, E. Mikhailov, M. Fleischhauer, V. L. Velichansky, G. R. Welch, and M. O. Scully, 2000, Phys. Rev. A **62**, 023810.
Schmidt, H., K. L. Chapman, A. C. Gossard, and A. Imamoglu, 1997, Appl. Phys. Lett. **70**, 3455.
Schmidt, H., and A. Imamoglu, 1996, Opt. Lett. **21**, 1936.
Schmidt, O., R. Wynands, Z. Hussein, and D. Meschede, 1996, Phys. Rev. A **53**, R27.
Schori, C., B. Julsgaard, J. L. Sorensen, and E. S. Polzik, 2002, Phys. Rev. Lett. **89**, 057903.
Scully, M. O., 1991, Phys. Rev. Lett. **67**, 1855.
Scully, M. O., 1992, Phys. Rep. **219**, 191.
Scully, M. O., and M. Fleischhauer, 1992, Phys. Rev. Lett. **69**, 1360.
Scully, M. O., S. Zhu, and A. Gavrielides, 1989, Phys. Rev. Lett. **62**, 2813.
Scully, M. O., and S. Y. Zhu, 1992, Opt. Commun. **87**, 134.
Scully, M. O., and M. S. Zubairy, 1997, *Quantum Optics* (Cambridge University Press, Cambridge, England).
Serapiglia, G. B., E. Paspalakis, C. Sirtori, K. L. Vodopyanov, and C. C. Phillips, 2000, Phys. Rev. Lett. **84**, 1019.
Shapiro, M., 1970, J. Chem. Phys. **56**, 2582.
Shore, B. W., 1967, Rev. Mod. Phys. **39**, 439.
Shore, B. W., 1990, *Theory of Coherent Atomic Excitation* (Wiley, New York).
Shvets, G., and J. S. Wurtele, 2002, Phys. Rev. Lett. **89**, 115003.
Sokolov, A. V., 2002, *KITP Workshop on Quantum Optics*

(University of California Santa Barbara, Santa Barbara, CA).
Sokolov, A. V., D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, 2000, Phys. Rev. Lett. **85**, 562.
Sokolov, A. V., D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, 2001, Phys. Rev. Lett. **87**, 033402.
Stahler, M., S. Knappe, C. Affolderbach, W. Kemp, and R. Wynands, 2001, Europhys. Lett. **54**, 323.
Steudel, H., 1988, J. Mod. Opt. **35**, 193.
Strekalov, D., A. B. Matsko, Nan Yu, and L. Maleki, 2004, Phys. Rev. Lett. **93**, 023601.
Sun, C. P., S. Yi, and L. You, 2003, Phys. Rev. A **67**, 063815.
Tewari, S. P., and G. S. Agarwal, 1986, Phys. Rev. Lett. **56**, 1811.
Turukhin, A. V., V. S. Sudarhanam, M. S. Shahriar, J. A. Musser, B. S. Ham, and P. R. Hemmer, 2001, Phys. Rev. Lett. **88**, 023602.
van der Wal, C., M. D. Eisaman, A. Andre, R. L. Walsworth, D. F. Phillips, A. S. Zibrov, and M. D. Lukin, 2003, Science **301**, 196.
Visser, M., 2000, Phys. Rev. Lett. **85**, 5252.
Vitanov, N. V., M. Fleischhauer, B. W. Shore, and K. Bergmann, 2001, Adv. At., Mol., Opt. Phys. **46**, 55.
Vitanov, N. V., and S. Stenholm, 1997, Phys. Rev. A **56**, 1463.
Wei, C., and N. B. Manson, 1999, Phys. Rev. A **60**, 2540.
Werner, M., and A. Imamoglu, 1999, Phys. Rev. A **61**, 011801(R).
Whitley, R. M., and C. R. Stroud, 1976, Phys. Rev. A **14**, 1498.
Wilson-Gordon, A. D., and H. Friedman, 1992, Opt. Commun. **94**, 238.
Wolf, S. A., D. D. Awschalom, R. A. Buhrman, J. M. Daughton, S. von Molnar, M. L. Roukes, A. Y. Chtchelkanova, and D. M. Treger, 2001, Science **294**, 1488.
Xiao, M., Y. Li, S. Jin, and J. Gea-Banacloche, 1995, Phys. Rev. Lett. **74**, 666.
Zhang, G. Z., K. Hakuta, and B. P. Stoicheff, 1993, Phys. Rev. Lett. **71**, 3099.
Zhang, G. Z., M. Katsuragawa, K. Hakuta, R. I. Thompson, and B. P. Stoicheff, 1995, Phys. Rev. A **52**, 1584.
Zhao, Y., C. Wu, B. S. Ham, M. K. Kim, and E. Awad, 1997, Phys. Rev. Lett. **79**, 641.
Zibrov, A. S., M. D. Lukin, D. E. Nikonov, L. Hollberg, M. O. Scully, V. L. Velichansky, and H. G. Robinson, 1995, Phys. Rev. Lett. **75**, 1499.
Zibrov, A. S., M. D. Lukin, and M. O. Scully, 1999, Phys. Rev. Lett. **83**, 4049.
Zibrov, A. S., A. B. Matsko, O. Kocharovskaya, Y. V. Rostovtsev, G. R. Welch, and M. O. Scully, 2002, Phys. Rev. Lett. **88**, 103601.
Zimmer, F., and M. Fleischhauer, 2004, Phys. Rev. Lett. **92**, 253201.
Zutic, Igor, J. Fabian, and S. Das Sarma, 2004, Rev. Mod. Phys. **76**, 323.