Nonlinear Optics at Low Light Levels

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We show how the combination of electromagnetically induced transparency based nonlinear optics and cold atom technology, under conditions of ultraslow light propagation, allows nonlinear processes at energies of a few photons per atomic cross section. [S0031-9007(99)09290-X]

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The recent observation by Hau and colleagues [1] of ultraslow light propagation (17 m/s) motivates the study of nonlinear optical processes under conditions where slow group velocity is a dominant feature of the problem. The basic idea is to combine electromagnetically induced transparency (EIT) [2–4] and cold atom technology to create a sharp resonance with a transmission linewidth (Fig. 1) which is much less than the natural linewidth of the atoms. A pulse of light propagating in this medium has a phase velocity of c and a group velocity which can be less than $10^{-7}c$. The light pulse also experiences exceedingly large nonlinearities with a magnitude limited only by the pulse width of the incident light and, ultimately, by the dephasing time of the coherence of the transition which is used to create the transparency.

In this Letter we consider the consequences of nonlinear optics using subnatural linewidths and ultraslow group velocities. A dominant feature of the problem is the spatial compression experienced by a pulse of light in the EIT medium. For example, in the experiments of Hau *et al.* [1], a 2.5 μs pulse, which is about 750 m long in free space, compresses into a spatial pulse length of 42 μm in the cold Na vapor. As the pulse propagates, one may use four-frequency nonlinearities to sum or difference with it [5,6] or to modify its refractive index [7] and attenuation constant [8]. The behavior and efficiency of these processes depend on the relative velocities and directions of the propagating pulses. Some striking effects should be possible. For example, by using two-photon absorption at right angles, one may take a picture of the slowly moving spatially compressed pulse and thus, in effect, display information which is accumulated over a longer period of time. Of most interest, these processes occur at energies corresponding to a few photons per atomic cross section or, equivalently, at nJ cm^{-2}.

There is now considerable literature on nonlinear optics with EIT. The essence of all of these effects is a destructive interference in absorption in combination with a constructive interference in the nonlinear susceptibility. In addition to those cited above, other examples include experiments and theory describing EIT-based phase conjugation [9,10], studies of bistability and two-photon absorption [11], efficient frequency summing at maximum coherence [12], new types of nonlinear spectroscopy [13], and studies of correlation and noise at the few quanta level [14]. We also note earlier general studies of double lambda systems [15].

We begin by summarizing the properties of a light pulse of frequency ω_p, which propagates at unity refractive index, and with a slow group velocity, which results from the steep slope of the refractive index $n(\omega_p)$. These are [16]

$$V_g = \frac{c}{(1 + \omega_p \frac{\partial n}{\partial \omega_p})},$$
$$\langle \mathcal{E} \rangle = \frac{1}{2}\epsilon_0 |E|^2 \left(1 + \omega_p \frac{\partial n}{\partial \omega_p}\right), \qquad (1)$$
$$\frac{P}{A} = \langle \mathcal{E} \rangle V_g = \frac{1}{2}(\epsilon_0 c)|E|^2 .$$

Because the refractive index is unity, the electric field and power density P/A are unchanged as the pulse enters the medium. But still, because the group velocity is decreased and the power density is constant, the energy density $\langle \mathcal{E} \rangle$

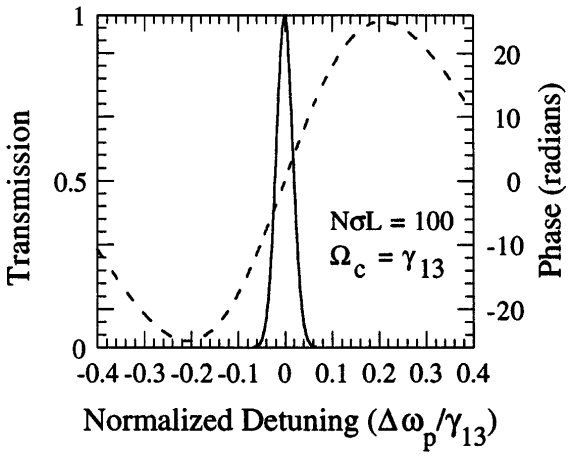

FIG. 1. Transmission and phase of the probe with perfect EIT.

in the medium must increase. This increase occurs by compression in space to a length which we term as the Hau length, L_h. In this Letter we will be especially interested in the case where this length is less than the length of the nonlinear medium.

An energy level diagram is shown in Fig. 2. The coupling laser, tuned to the $|2\rangle \rightarrow |3\rangle$ transition, has Rabi frequency Ω_c and is assumed to be strong and monochromatic. Its magnitude is set so as to control the width of the transmission of Fig. 1 and therefore the bandwidth of the system. The probe pulse ω_p is taken to be on resonance with the $|1\rangle \rightarrow |3\rangle$ transition. It is assumed to be sufficiently weak that the ratio $(\Omega_p/\Omega_c)^2$ is much less than unity, so that in essence all of the atomic population remains in the ground state $|1\rangle$. With the exception of Eq. (2a), we will assume the ideal case of a zero dephasing rate, $\gamma_{12} = 0$, for the $|1\rangle \rightarrow |2\rangle$ transition, and therefore take $\gamma_{23} = \gamma_{13}$. With these assumptions and with the definition of the atomic adsorption cross section $\sigma_{13} = \omega_{13}|\mu_{13}|^2/(c\epsilon_0\hbar\gamma_{13})$, the propagation constants of the slowly moving probe pulse, *relative to vacuum*, are [17,18]

$$\alpha_p = \left(\frac{2\gamma_{12}\gamma_{13}}{|\Omega_c|^2} + \frac{8\gamma_{13}^2\Delta\omega_p^2}{|\Omega_c|^4}\right)N\sigma_{13}\,, \tag{2a}$$

$$\beta_p = \frac{2\gamma_{13}\Delta\omega_p}{|\Omega_c|^2}\,N\sigma_{13}\,, \tag{2b}$$

$$\frac{1}{V_p} = \frac{2\gamma_{13}}{|\Omega_c|^2}\,N\sigma_{13}\,, \tag{2c}$$

$$L_h = \left(\frac{|\Omega_c|^2}{2\gamma_{13}N\sigma_{13}}\right)T_p\,. \tag{2d}$$

The quantities α_p and β_p determine the attenuation and phase shift of the propagating probe pulse. If the probe is monochromatic and exactly on resonance, and with $\gamma_{12} = 0$, $\alpha_p = \beta_p = 0$. For finite pulse bandwidths, to fourth order in $|\Omega_c|$, one obtains the group velocity and the lowest order absorptive term. The reciprocal of the actual (stationary frame) group velocity is equal to $1/c$ plus $1/V_p$. Because the group velocity dispersion is zero at line center, the pulse shape is quite well preserved during propagation. Equation (2d) gives the expression for the Hau length L_h. This length is the distance in which the probe pulse would separate by one pulse length from a pulse of the same temporal length, T_p, which is traveling at c. It is the equivalent of the walk-off length of crystalline nonlinear optics [19]. For group velocities which are slow enough that c may be taken as infinite, this length is the spatial length of the pulse in the medium.

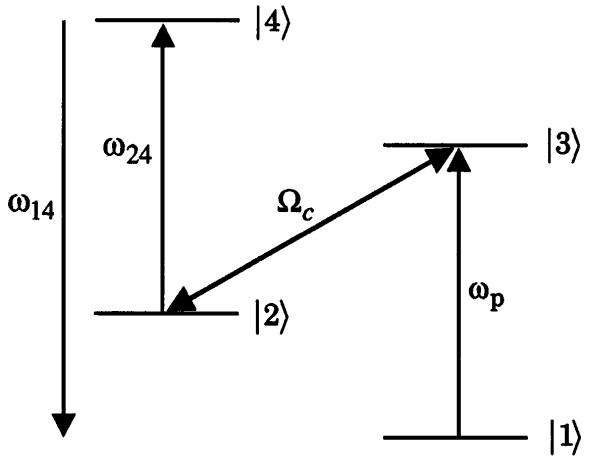

FIG. 2. Energy schematic for the analysis.

The formulas of Eqs. (2) may be combined to establish limits on the minimum pulse width and therefore the minimum L_h. With the dephasing rate $\gamma_{12} = 0$ and a length of nonlinear medium L, the minimum pulse width is determined from Eq. (2a) by setting $\alpha_p L = 0.5$. For a Gaussian shaped pulse, one obtains

$$\begin{aligned} T_{p(\min)} &= 8\ln 2\left(\frac{\gamma_{13}}{|\Omega_c|^2}\right)(N\sigma_{13}L)^{1/2}\,, \\ L_{h(\min)} &= 4\ln 2\left(\frac{L}{N\sigma_{13}}\right)^{1/2}\,, \end{aligned} \tag{3}$$

We first consider collinear sum frequency generation in atoms with the prototype energy level diagram of Fig. 2. We assume applied frequency ω_p, ω_c, and ω_{24}, and a generated frequency ω_{14}. The slowly varying envelope equation for the Rabi frequency Ω_{14} is

$$\begin{aligned} \frac{\partial\Omega_{14}}{\partial z} + \frac{1}{c}\frac{\partial\Omega_{14}}{\partial t} + \alpha_{14}\Omega_{14} &= \kappa_{14}\Omega_c^*\Omega_p\left[t - \left(\frac{1}{V_p} + \frac{1}{c}\right)z\right]\Omega_{24}\left(t - \frac{z}{c}\right), \\ \kappa_{14} &= -\frac{j}{2}\,\frac{\gamma_{14}N\sigma_{14}}{\Delta\tilde{\omega}_{14}|\Omega_c|^2} = -\left(\frac{j}{4\Delta\tilde{\omega}_{14}}\right)\left(\frac{\sigma_{14}\gamma_{14}}{\sigma_{13}\gamma_{13}}\right)\frac{1}{V_p}\,, \\ \alpha_{14} &= \left(\frac{\gamma_{14}^2}{\Delta\omega_{14}^2 + \gamma_{14}^2}\right)\frac{N\sigma_{14}}{2}\,. \end{aligned} \tag{4}$$

$\Delta\omega_{14} = (\omega_p - \omega_c + \omega_{24}) - (\omega_4 - \omega_1)$, $\Delta\tilde{\omega}_{14} = \Delta\omega_{14} - j\gamma_{14}$, and σ_{14} is the absorption cross section of the $|1\rangle \rightarrow |4\rangle$ transition. Equation (4) assumes k-vector matching of the interacting frequencies. This may be attained by very small detunings of the probe laser frequency [20]. Writing $\Omega_p(t,z) = \Omega_p f(t,z)$ and $\Omega_{24}(t,z) = \Omega_{24}g(t,z)$, and working in a frame moving at c, the solution of Eq. (4) is

$$\Omega_{14}(L,t) = \kappa_{14}\Omega_c^*\Omega_p\Omega_{24} \times \int_0^L \exp[\alpha_{14}(z-L)] f\left(t - \frac{z}{V_p}\right) g(t)\, dz\,. \tag{5}$$

We use $|\Omega_{ij}|^2 = (2\gamma_{ij}\sigma_{ij}/\hbar\omega_{ij})(P_{ij}/A)$ and, after some algebra but with no further assumptions, obtain

$$\text{Efficiency} = \frac{(1/\hbar\omega_{14})\int P_{14}(t)\, dt}{(1/\hbar\omega_{24})\int P_p(t)\, dt} = \frac{1}{\hbar\omega_{24}}\left(\frac{P_{24}}{A}T_{24}\right)\sigma_{24}\Phi(\eta,r), \tag{6}$$

where

$$\Phi(\eta,r) = \left(\frac{1}{2T_{24}}\right)\frac{r}{\eta}\int_{-\infty}^{+\infty} \times \left|\int_0^\eta \exp\left[\frac{r}{\eta}(\xi-\eta)\right] f(t-\xi T_p) \times g(t)\, d\xi\right|^2 dt\,.$$

The quantity $\eta = L/L_h$ is the ratio of the material length L to the Hau length L_h. The quantity $r = \alpha_{14}L$ and is equal to the (E-field) loss in a length L. The ratio of the number of photons which are generated at frequency ω_{14} to the number of photons incident at ω_p is equal to a dimensionless factor $\Phi(\eta,r)$ times the number of photons in an atomic cross section σ_{24}.

Figure 3 shows $\Phi(\eta,r)$, for Gaussian shaped pulses, as a function of η for several values of r. In the limit when η and r are small,

$$\Phi(\eta,r) = \left[\frac{\ln(2)}{\pi}\right]^{1/2}\frac{T_p}{\sqrt{T_p^2 + T_{24}^2}}\,\eta r, \qquad \eta \to 0, \quad r \to 0\,. \tag{7a}$$

In this limit the long-pulse, plane-wave formulas of nonlinear optics are regained.

When the length L_h is much less than the length of the nonlinear material, i.e., as η becomes very large, we may

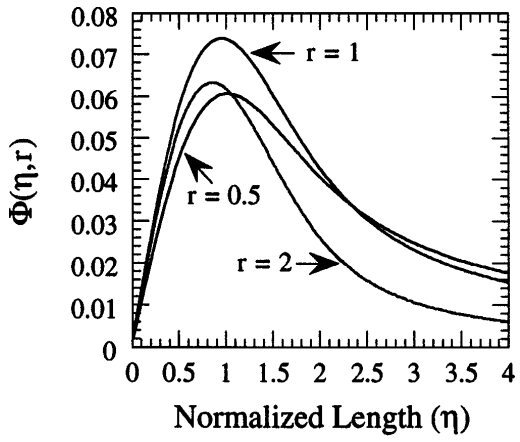

FIG. 3. $\Phi(\eta,r)$ vs normalized length. The function $\Phi(\eta,r)$ is evaluated for Gaussian pulses of equal length.

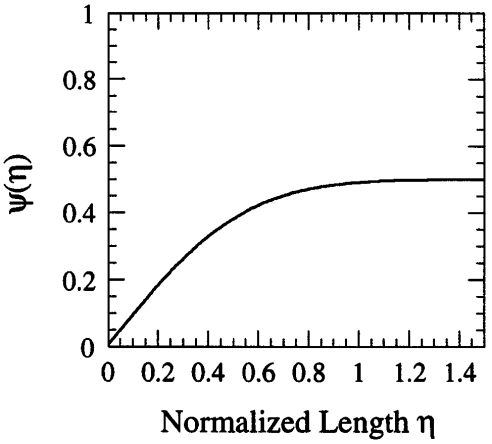

FIG. 4. $\psi(\eta)$ vs normalized length for Gaussian pulses of equal length.

treat this length as a delta function in space and use it to obtain the asymptotic form of $\Phi(\eta,r)$,

$$\Phi(\eta,r) = \left[\frac{\ln(2)}{\pi}\right]^{1/2}\frac{r}{\eta}\exp(-2r), \qquad \eta \to \infty\,. \tag{7b}$$

When the Hau length is short, as compared to the length of the nonlinear medium ($\eta \gg 1$), generation occurs as the ω_{24} pulse sweeps by the almost stationary probe pulse. In this limit, and for constant energy in the ω_{24} pulse, the generation efficiency is independent of the pulse length T_{24}. Because of the more rapid reduction of L_h, an attempt to increase the conversion efficiency by reducing Ω_c, and therefore to increase the nonlinearity, fails. For Gaussian pulses of equal length the maximum value of $\Phi(\eta,r)$ occurs at about $\eta = r = 1$ and is $\cong 0.074$ [Fig. 3]; i.e., four-frequency mixing is optimized by choosing a group velocity such that the material length and the Hau length are equal. We note that the asymptotic form [Eq. (7b)] of $\Phi(\eta,r)$ is useful even as this optimum value is approached.

We next consider the cross-Kerr nonlinearity suggested by Schmidt and Imamoglu and indirectly measured by Hau *et al.* [1,7], and the two-photon absorption process of Harris and Yamamoto [8]. Both occur when the ideal EIT process is modified by the interaction of the field at frequency ω_{24} with the $|2\rangle \to |4\rangle$ transition. With zero detuning in this channel there is absorption and no phase shift at ω_p. At finite detunings, the Stark shift of state $|2\rangle$ results in a nonlinear phase accumulation at ω_p. Both absorption and nonlinear phase shift are present together and, with $\Delta\tilde{\omega}_{24} = \Delta\omega_{24} - j\gamma_{24}$, are described by

$$\frac{\partial\Omega_p}{\partial z} + \left(\frac{1}{V_p} + \frac{1}{c}\right)\frac{\partial\Omega_p}{\partial t} = \kappa_p\left|\Omega_{24}\left(t - \frac{z}{c}\right)\right|^2\Omega_p\,, \qquad \kappa_p = \left(\frac{-j}{4\Delta\tilde{\omega}_{24}}\right)\frac{1}{V_p}\,. \tag{8}$$

The coupling constant κ_p is inversely proportional to the group velocity and leads to the invariance to changing Ω_c

TABLE I. Two-photon absorption, Kerr phase shift, and generation efficiency as a function of incident photons/area.

Process	Photons/area mulitplied by
Two-photon absorption (power loss on resonance)	$\sigma_{24}\psi$
Kerr nonlinearity (phase shift; $\Delta\omega_{24} \gg \gamma_{24}$)	$(\frac{\gamma_{24}}{2\Delta\omega_{24}})\sigma_{24}\psi$
Four-frequency summing (efficiency)	$\sigma_{24}\Phi$

which is noted below. In a frame moving with the probe, the solution of Eq. (8) is

$$\Omega_p(L,t) = \Omega_p(0,t) \times \exp\left[\int_0^L \kappa_p \left|\Omega_{24}\left(t + \frac{z}{V_p}\right)\right|^2 dz\right]. \tag{9}$$

We evaluate the phase shift at the peak of the pulse ($t = 0$) and express the phase shift as a function of the normalized length $\eta = L/L_h$. With no further assumptions,

$$\Omega_p(\eta) = \Omega_p(0)\exp[-jR(\eta)],$$
$$R(\eta) = \left(\frac{P_{24}T_{24}\sigma_{24}}{\hbar\omega_{24}A}\right)\left(\frac{\gamma_{24}}{2\Delta\tilde{\omega}_{24}}\right)\psi(\eta), \tag{10}$$
$$\psi(\eta) = \frac{1}{2}\,\mathrm{erf}\left[\frac{\sqrt{4\ln 2}\,T_p\,\eta}{T_{24}}\right].$$

The propagation factor $R(\eta)$ is equal to the number of photons in an atomic cross section σ_{24} multiplied by the factors $\gamma_{24}/(2\Delta\tilde{\omega}_{24})$ and $\psi(\eta)$. The function $\psi(\eta)$, shown in Fig. 4, has limits

$$\psi(\eta) = \left(\frac{4\ln 2}{\pi}\right)^{1/2}\frac{T_p}{T_{24}}\,\eta, \qquad \eta \to 0,$$
$$= \frac{1}{2}, \qquad \eta \to \infty.$$

As L_h becomes long as compared to the sample length L, $\eta \to 0$, and we regain the nonlinear index of Ref. [7]; i.e., with $n = 1 + n_2(P_{24}/A)$, $n_2 = -(4\ln 2/\pi)^{1/2}|\mu_{13}|^2|\mu_{24}|^2/(c\epsilon_0^2\hbar^3\Delta\omega_{24}|\Omega_c|^2)$ and also the two-photon absorption of Ref. [8]. In the opposite limit when L_h is less than the material length, the phase accumulation and two-photon absorption become independent of the atom density and the length of the medium.

Table I summarizes the previous results. Each of the nonlinear processes require pulse energies corresponding to a single photon per atomic cross section multiplied by an additional factor. For the process of two-photon absorption, when $L_h \ll L$, this additional factor is 0.5. For the Kerr nonlinearity, this factor varies inversely as the ratio of nonlinear phase shifts to nonlinear loss which is desired, and again, in the limit $L_h \ll L$, is independent of the length of the medium. For four-frequency mixing, the factor maximizes when the group velocity is reduced until the Hau length and the material length are approximately equal. Pulse energies of nJ cm^{-2}, i.e., at least 6 orders of magnitude less than previously used, become appropriate for near-resonant cold atom nonlinear optical processes.

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