Refractive-index control with strong fields

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An electromagnetically induced transparency-like effect is described that allows a strong laser to control and reduce to unity the refractive index of a weak probe. A lossless multistate system with all states far from resonance is considered.

Electromagnetically induced transparency (EIT) in a three-state atomic system is now reasonably well understood.[1-4] In this effect a strong controlling laser is used to create a combined Autler–Townes splitting and quantum interference, which allows a probe laser to propagate through what would otherwise be an opaque medium. The refractive index is also modified and, for an ideal three-state system, is unity on resonance and linearly dispersive near resonance.[5,6]

We focus on the use of an EIT-like effect to control the refractive index of a weak probe beam, when far from resonance, in an otherwise nonabsorbing atomic or molecular system. We study a system that has an infinite number of upper states (Fig. 1), which are connected by arbitrary matrix elements to lower states $|1\rangle$ and $|2\rangle$. We are interested in controlling the refractive index of a probe laser with center frequency ω_p by a strong laser with center frequency ω_c. We will assume that the detuning from the upper states $|i\rangle$ is sufficiently large that, in the spirit of the usual (detuned) refractive-index calculation, the loss associated with all the $|1\rangle-|i\rangle$ and $|2\rangle-|i\rangle$ transitions is inconsequential and may be neglected.

We will see that the problem is characterized by a parameter that varies linearly with the power density of the controlling laser. When this parameter is large compared with the linewidth of the $|1\rangle-|2\rangle$ transition, then effective control and reduction to unity of the refractive index of the probe beam become possible. When this parameter is small compared with the linewidth the refractive index is nominally unchanged, and the probe is attenuated by two-photon absorption.

Although we will limit the latter part of this work to the case in which the controlling laser is strong compared with the probe laser, we develop the equations without this assumption. We consider one-dimensional propagation with applied electromagnetic fields

$$E_p(z,t) = \mathrm{Re}[E_p f(z,t)\exp j(\omega_p t - k_p z)],$$
$$E_c(z,t) = \mathrm{Re}[E_c g(z,t)\exp j(\omega_c t - k_c z)]. \quad (1)$$

The quantities $f(z,t)$ and $g(z,t)$ are the (complex) envelopes of the probe and control lasers and vary with space and time. The electric field amplitudes E_p and E_c are real and are independent of space and time. The detunings of the upper states $\Delta\omega_i$ are referenced to the probe frequency (Fig. 1). We allow for a (complex) two-photon detuning, $\Delta\tilde{\omega}_2 = \delta\omega_2 - j(\Gamma_2/2)$, from state $|2\rangle$. When it is optimized, the real part of this detuning, $\delta\omega_2$, permits compensation for the portion of the ac-Stark shift that is not already eliminated by the inherent destructive interference of EIT. When it is set differently from its optimum value, this quantity allows for a simulation of the effects of inhomogeneous broadening. The imaginary part of $\Delta\tilde{\omega}_2$ is used to create a homogenous linewidth for state $|2\rangle$. (Rigorously, this term may be viewed as a decay to other states that are outside the system.)

We will assume that each of the states $|i\rangle$ of Fig. 1 has nonzero matrix elements to both states $|1\rangle$ and $|2\rangle$. For convenience, we take these matrix elements as real and define (real) Rabi frequencies as $\Omega_{1i} = \mu_{1i}E_p/\hbar$ and $\Omega_{2i} = \mu_{2i}E_c/\hbar$. States $|i\rangle$, which couple only to states $|1\rangle$ and $|2\rangle$ but not to both, cause additional ac-Stark shifts and a background refractive index and are not included here. Nonrotating terms are also neglected and may also be included separately. (In most cases, for a weak probe a small

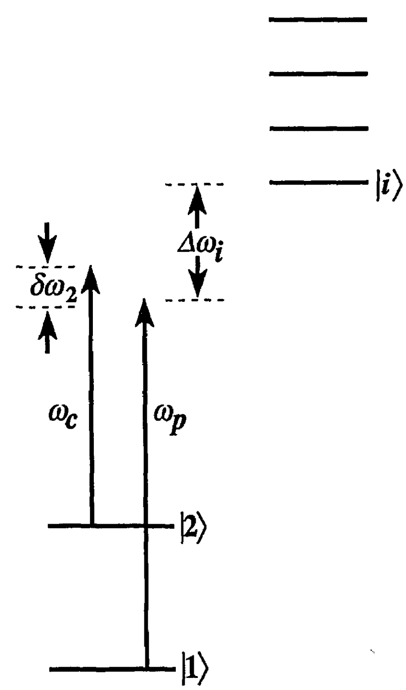

Fig. 1. Energy-level schematic for refractive-index control.

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change in $\delta\omega$ will allow for compensation for all the neglected effects.)

With these approximations, the equations for the probability amplitudes are

$$\frac{\partial a_1}{\partial t} = j\left(\sum_i \frac{\Omega_{1i}}{2} a_i\right) f\,, \tag{2a}$$

$$\frac{\partial a_2}{\partial t} + j\Delta\tilde{\omega}_2 a_2 = j\left(\sum_i \frac{\Omega_{2i}}{2} a_i\right) g\,, \tag{2b}$$

$$\frac{\partial a_i}{\partial t} + j\Delta\omega_i a_i = \frac{j}{2}\Omega_{1i} a_1 f^* + \frac{j}{2}\Omega_{2i} a_2 g^*\,. \tag{2c}$$

We assume that the derivatives of the probability amplitudes of the upper states $|i\rangle$ are small compared with the detuning from these states and can be neglected. We solve Eq. (2c) for a_i in terms of a_1 and a_2 and substitute the result into Eqs. (2a) and (2b). With the definitions[7] of frequencies

$$A = \sum_i \frac{\Omega_{1i}^2}{2\Delta\omega_i} = \frac{E_p^2}{\hbar^2}\sum_i \frac{\mu_{1i}^2}{2\Delta\omega_i}\,,$$
$$D = \sum_i \frac{\Omega_{2i}^2}{2\Delta\omega_i} = \frac{E_c^2}{\hbar^2}\sum_i \frac{\mu_{2i}^2}{2\Delta\omega_i}\,,$$
$$B = C = \sum_i \frac{\Omega_{1i}\Omega_{2i}}{2\Delta\omega_i} = \frac{E_p E_c}{\hbar^2}\sum_i \frac{\mu_{1i}\mu_{2i}}{2\Delta\omega_i}\,, \tag{3}$$

the equations for the probability amplitudes a_1 and a_2 are

$$\frac{\partial a_1}{\partial t} = j\frac{A}{2} a_1 |f|^2 + j\frac{B}{2} a_2 f g^*\,, \tag{4a}$$

$$\frac{\partial a_2}{\partial t} + j\Delta\tilde{\omega}_2 a_2 = j\frac{C}{2} a_1 g f^* + j\frac{D}{2} a_2 |g|^2\,. \tag{4b}$$

We take dipole moments and, from Maxwell's equations, form propagation equations for the envelopes f and g:

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right) f = -j\beta_p\left(|a_1|^2 f + \frac{B}{A} a_1 a_2^* g\right), \tag{4c}$$

$$\left(\frac{\partial}{\partial z} + \frac{1}{c}\frac{\partial}{\partial t}\right) g = -j\beta_c\left(|a_2|^2 g + \frac{C}{D} a_1^* a_2 f\right). \tag{4d}$$

With an atom density N, the quantities $\beta_p = (\mu_0/\varepsilon_0)^{1/2}\hbar\omega_p NA/E_p^2$ and $\beta_c = (\mu_0/\varepsilon_0)^{1/2}\hbar\omega_c ND/E_c^2$ are the real parts of the propagation constants of the probe and control lasers when all atoms are in states $|1\rangle$ and $|2\rangle$, respectively, and with the alternative laser absent.

We study these effects for the case in which the control laser is sufficiently strong compared with the probe laser that almost all the population remains in the ground state ($a_1 = 1$). We also assume that the envelope of the control laser is time independent and, therefore, take $g = 1$. With these approximations, Eqs. (4a) and (4d) are ignored, and the solution of Eq. (4b) is written as

$$a_2 = \sum_{n=1}^{\infty} \frac{j^{(n-1)}}{2} \frac{C}{[\Delta\tilde{\omega}_2 - (D/2)]^n} \frac{\partial^{n-1} f^*}{\partial t^{n-1}}\,. \tag{5}$$

We define the strong-field parameter K as

$$K \equiv \frac{BC}{A} = \frac{E_c^2}{\hbar^2} \frac{\left[\sum_i(\mu_{1i}\mu_{2i}/2\Delta\omega_i)\right]^2}{\sum_i(\mu_{1i}^2/2\Delta\omega_i)}\,. \tag{6}$$

We retain the $n = 1$ and $n = 2$ terms of Eq. (5) and substitute these into Eq. (4c). For a time-independent control field, and to the order of the retained terms, the equation for the propagation of a weak probe pulse is

$$\left(\frac{\partial}{\partial z} + \frac{1}{V_G}\frac{\partial}{\partial t}\right) f = -j\xi\beta_p f\,, \tag{7a}$$

$$\xi = 1 + \frac{K}{2}\frac{1}{[\Delta\tilde{\omega}_2^* - (D/2)]}\,, \tag{7b}$$

$$\frac{1}{V_G} = \frac{1}{c} + \frac{K}{2}\frac{1}{[\Delta\tilde{\omega}_2^* - (D/2)]^2}\beta_p\,. \tag{7c}$$

We first note the ideal case; with $\Gamma_2 = 0$,

$$\delta\omega_2 = 1/2[D - (BC/A)]$$
$$= 1/2(D - K)$$
$$\xi = 0\,, \tag{8}$$

i.e., in the limit of zero linewidth of the $|1\rangle$–$|2\rangle$ transition there is always a detuning $\delta\omega_2$ of the control laser such that $\xi = 0$, and the refractive index of the probe is unity. If there is only a single upper state $|i\rangle$, then $AD - BC = 0$, $\delta\omega_2 = 0$, and the problem reduces to the standard three-state EIT problem.

The parameter K determines the weak- and strong-field limits of ξ. In the weak-field limit, $K \ll \Gamma_2$,

$$\xi_{\text{weak-field}} = 1 - j\frac{K}{\Gamma_2} + \cdots + \,. \tag{9a}$$

Here the probe laser sees the normal refractive index and also two-photon absorption as caused by the control laser. In the strong-field limit, $K \gg \Gamma_2$ and $\delta\omega_2 = (D - K)/2$,

$$\xi_{\text{weak-field}} = -j\frac{\Gamma_2}{K} + \cdots + \,. \tag{9b}$$

Here, increasing the strength of the control laser decreases the absorption and, for sufficiently large K, causes $\xi \to 0$ and the refractive index to approach unity.

We turn next to the group-velocity term of Eq. (7c). This term sets the minimum total energy density for the control laser. The argument is that the control laser pulse (with power density P_c/A) must have a pulse length that is at least as long as the time it takes for the probe pulse (relative to the control pulse) to transit through the medium. For a medium of length L with atom density N, and with $\Gamma_2 = 0$ and $2\delta\omega_2 = D - K$, we obtain

$$\mathcal{E}_c/A = \frac{P_c}{A} L\left(\frac{1}{V_G} - \frac{1}{c}\right)$$
$$= \frac{P_c}{A} L\left(\frac{2\beta_p}{K}\right)$$
$$= R\hbar\omega_c NL\,, \tag{10}$$

where

$$R = \frac{\omega_p}{\omega_c} \frac{\left[\sum_i (\mu_{1i}{}^2/2\Delta\omega_i)\right]^2}{\left[\sum_i (\mu_{1i}\mu_{2i}/2\Delta\omega_i)\right]^2} . \qquad (11)$$

(The dimensionless ratio R is defined so that, for a resonant three-state system with equal oscillator strength on the $|1\rangle-|3\rangle$ and $|2\rangle-|3\rangle$ transitions, $R = 1$.) We thus find the important result that the pulse length of the control laser must be sufficiently long that it has a number of photons equal to the number of atoms through which it will transit.

For a given linewidth of the $|1\rangle-|2\rangle$ transition, Eq. (9b) determines the power density of the control laser that is required for a given reduction in the refractive index. Using it, one may show that to reduce ξ until $|\xi\beta_p L| = 1$ requires a control laser power density of

$$\frac{P_c}{A} = \frac{\Gamma_2}{2} R\hbar\omega_c NL . \qquad (12)$$

For pulses that are long compared with the inverse linewidth, $1/\Gamma_2$, the control laser power density requirement is set by Eq. (12). For pulses that are short compared with this linewidth, the energy density requirement is set by Eq. (10).[8]

We remark that the small probe assumption is necessary for this work but is not necessary when there is only a single upper state. For three-state EIT the Stark shift $2\delta\omega_2 = AD - BC$ is always zero, and the propagation constants at both the probe and the control lasers are equal to their free-space values. For multistate EIT we choose $\delta\omega_2$ so that the probe propagates at its free-space value; but, here, the propagation constant of the control laser approaches its free-space value only as the probe intensity approaches zero.

To observe refractive-index control in gases at large detunings (for example, 10^4 cm^{-1} with a Doppler width of 0.1 cm^{-1}) requires a control laser power density of $\sim 10^{10}$ W/cm^2. To transit through an NL product of 10^{17} atoms/cm^2, for red photons, requires a control laser energy density of >30 mJ/cm^2. Lasers with pulse length of several picoseconds may therefore be most appropriate.

Nonlinear-optical processes based on EIT are already showing promise for applications. To date, experiments have been based on three-state systems operated at or near resonance.[9,10] The results of this Letter suggest the extension to multistate systems operated far from resonance.

This problem shares some common features with the problem of propagation through an asymmetric continuum. In particular, both make use of a frequency offset from two-photon resonance to adjust either the real or the imaginary part of the propagation constant.[11,12]

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