

## Broadband spectral generation with refractive index control

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We describe a method for generating a wide spectrum and at the same time improving propagation through a model inhomogeneous gaseous medium. The method is based on using a small two-photon detuning from a Raman resonance to adiabatically establish an appropriate molecular superposition state. This state, in turn, generates a broad spectrum of sidebands whose refractive index, Rayleigh scatter, and accumulated phase delay are much less than those of a single spectral component, if propagating alone. [S1050-2947(97)50606-8]

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It is desirable to learn to improve the propagation of laser beams through inhomogeneous and turbulent refracting gases and vapors. This will happen if, irrespective of the density of the gas, the refractive index is reduced toward unity. Following the recent results of Jain *et al.* [1], it is expected that, if the nonlinear portion of the dipole moment is of the same magnitude as is the linear portion, then the generation of a very broad spectrum of sidebands should become possible. In this Rapid Communication we describe a technique for accomplishing both of these objectives in a model molecular system.

It turns out that this is surprisingly easy to do: Noting Fig. 1, the idea is to apply two frequencies whose difference is slightly greater than that of a selected Raman transition of the molecule and whose intensity is sufficiently large so as to cause the magnitude of the coherence of this transition to have its maximum value  $|\rho_{ab}|=0.5$ . This coherence mixes with the two incident frequencies to generate a spectrum of Raman sidebands. What is intriguing, and is the central result of this Rapid Communication, is that these sidebands are phased so as to reduce the refractive index and accumulated phase of much of the spectrum. This occurs irrespective of the density of the medium and, also, over a reasonable range of the intensity of the incident fields. This result may also be viewed as broadening the definition of electromagnetically induced transparency (EIT) to include the interaction of an arbitrary number of spectral components that are separated by the frequency of a Raman transition.

In the following paragraphs we first develop a formulation of the problem that is valid in atomic or molecular systems where the detunings of the applied frequencies (Fig. 1) from the electronic states  $|i\rangle$  are large as compared to the Rabi frequencies and where, therefore, the probability amplitudes of these states are small. We will assume that, before the electromagnetic fields are applied, all of the atoms or molecules are in a single (nondegenerate) ground state. We take the two fields that are applied at the input of the medium to be monochromatic and write equations for the generated spectrum as a function of distance. We then give numerical results for incident wavelengths of 355 and 415 nm applied to molecular parahydrogen. These wavelengths set up an antiphased (dark) molecular superposition state of the ( $J=0$ ) ground and first vibrational level.

There is now an extensive literature on EIT in its own right and on its relation to lasers without population inver-

sion. Representative papers are cited in Refs. [2–7]. Of immediate pertinence, Gaubatz *et al.* have studied population trapping and adiabatic preparation in molecules [8]. Recently, and after the completion of the analytical but not the numerical portion of this work, Hakuta described effects in solid molecular  $H_2$  [9]. Earlier, Griskowsky [10] and Oreg *et al.* [11] described adiabatic preparation in two-state and multistate systems.

This work is also related to the work of Kaplan and Shkolnikov on Raman solitons [12]. These authors show that a spectrum of Raman modes of intensity sufficient to form a (two-photon)  $2\pi$  pulse will propagate without variation in area. The essential difference of their work from ours is the same as that of self-induced transparency and electromagnetically induced transparency: The former depends on pulse area and the latter does not.

We proceed with the analysis of this system by including all possible sidebands, whether the sideband frequencies are applied at the input or are generated as part of the propagation process. We denote the envelopes and frequencies of these arbitrarily phased sidebands by  $E_q$  and  $\omega_q$ ,

$$\omega_q = \omega_0 + q(\omega_b - \omega_a - \Delta\omega), \quad (1)$$

where  $\omega_a$  and  $\omega_b$  are the frequencies of the ground and excited molecular states and  $q$  is integer and positive or negative. The quantity  $\Delta\omega$  is the two-photon detuning of any pair of sidebands from the Raman (nonallowed) transition. It is

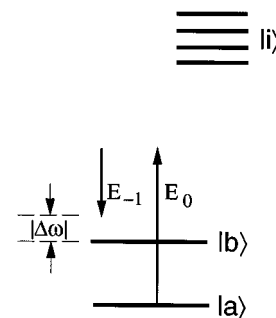


FIG. 1. Energy-level schematic for establishing coherence  $\rho_{ab}$  in a molecular system. Laser fields are applied at the frequencies of the  $q=0$  and  $-1$  sidebands. In the configuration shown, the two-photon detuning  $\Delta\omega$  from the Raman resonance is negative.

taken to be positive when the frequency difference ( $\omega_q - \omega_{q-1}$ ) is smaller than the frequency of the Raman resonance ( $\omega_b - \omega_a$ ). We allow an arbitrary number of virtual states  $|i\rangle$  with energies  $\omega_i$ . The matrix elements from states  $|a\rangle$  and  $|b\rangle$  to these states are  $\mu_{ai}$  and  $\mu_{bi}$ , respectively. We assume that the derivatives of the probability amplitudes of the upper states  $|i\rangle$  are small compared to the detunings from these states and can be neglected. We also assume that the two-photon detuning is small compared to the frequency of the Raman transition. These assumptions allow the problem to be written in terms of an effective  $2 \times 2$  Hamiltonian [13]:

$$H_{\text{eff}} = -\frac{1}{2} \begin{bmatrix} \sum_q a_q |E_q|^2 & \sum_q b_q E_q E_{q-1}^* \\ \sum_q c_q E_q E_{q+1}^* & \sum_q d_q |E_q|^2 - 2\Delta\omega \end{bmatrix} \quad (2a)$$

$$\equiv -\frac{1}{2} \begin{bmatrix} A & B \\ C & D - 2\Delta\omega \end{bmatrix}, \quad (2b)$$

The constants  $a_q$ ,  $b_q$ ,  $c_q$ , and  $d_q$  are

$$\begin{aligned} a_q &= \frac{1}{2\hbar^2} \sum_i \left[ \frac{|\mu_{ai}|^2}{(\omega_i - \omega_a) - \omega_q} + \frac{|\mu_{ai}|^2}{(\omega_i - \omega_a) + \omega_q} \right], \\ d_q &= \frac{1}{2\hbar^2} \sum_i \left[ \frac{|\mu_{bi}|^2}{(\omega_i - \omega_b) - \omega_q} + \frac{|\mu_{bi}|^2}{(\omega_i - \omega_b) + \omega_q} \right], \\ b_q &= \frac{1}{2\hbar^2} \sum_i \left[ \frac{\mu_{ai}\mu_{bi}}{(\omega_i - \omega_a) - \omega_q} + \frac{\mu_{ai}\mu_{bi}}{(\omega_i - \omega_b) + \omega_q} \right], \\ c_q &= b_{q+1}^*. \end{aligned} \quad (3)$$

All rotating and nonrotating, as well as cross-transition, terms are retained within this Hamiltonian. (If the cross-transition and nonrotating terms are neglected, the Hamiltonian reduces to that of Ref. [13].) We also assume the ideal case of zero linewidth for the  $|a\rangle$ - $|b\rangle$  transition.

With the assumption that the envelope of each spectral component varies slowly in space, as compared to its wavelength, the propagation equation for the  $q$ th sideband, in local time, is

$$\begin{aligned} \frac{\partial E_q}{\partial z} &= -j\eta\hbar\omega_q N (a_q \rho_{aa} E_q + d_q \rho_{bb} E_q \\ &+ b_q \rho_{ab} E_{q-1} + c_q \rho_{ab}^* E_{q+1}). \end{aligned} \quad (4)$$

The quantities  $\rho_{ij}$  are the elements of the  $2 \times 2$  density matrix.  $N$  is the number of molecules per volume and  $\eta = (\mu/\epsilon_0)^{1/2}$ . The refractive index of the  $q$ th mode, if alone with all molecules in state  $|a\rangle$  is  $n_q = 1 + (N\hbar a_q/\epsilon_0)$ , and with all molecules in state  $|b\rangle$  is  $1 + (N\hbar d_q/\epsilon_0)$ .

From Eqs. (3) and (4) we obtain conservation relations for photons and power,

$$\frac{\partial}{\partial z} \left( \sum_q \frac{1}{\hbar\omega_q} \frac{|E_q|^2}{2\eta} \right) = 0,$$

$$\frac{\partial}{\partial z} \left( \sum_q \frac{|E_q|^2}{2\eta} \right) = -\frac{N\hbar}{2} (\omega_b - \omega_a) \left[ \frac{\partial}{\partial t} (\rho_{bb} - \rho_{aa}) \right]. \quad (5)$$

Even in the presence of a changing atomic population, the total number of photons remains constant with distance.

In common with other types of electromagnetically induced transparency and population trapping, we must choose an eigenstate of the effective Hamiltonian that evolves smoothly from the ground state  $|a\rangle$ . Writing  $B = |B|e^{j\varphi}$ , this eigenstate and the difference of the eigenvalues of the Hamiltonian of Eq. (2) are

$$|+\rangle = \cos \frac{\theta}{2} e^{j\varphi/2} |a\rangle + \sin \frac{\theta}{2} e^{-j\varphi/2} |b\rangle, \quad (6a)$$

$$E_+ - E_- = \sqrt{\left( \Delta\omega - \frac{D}{2} + \frac{A}{2} \right)^2 + |B|^2}, \quad (6b)$$

where

$$\tan \theta = \frac{2|B|}{2\Delta\omega - D + A}.$$

The phased or antiphased state is selected by the sign of the two-photon detuning. The coherence  $\rho_{ab}$  is then

$$\rho_{ab} = \frac{1}{2} \sin \theta e^{i\varphi}. \quad (7)$$

For molecular systems with large detunings, the Stark shifts  $A$  and  $D$  are approximately equal and  $\theta \approx \tan^{-1}(2|B|/2\Delta\omega)$ . One method of achieving the condition  $\rho_{ab} = -0.5$  at  $z=0$  is to choose  $\Delta\omega$  negative and to increase the product of the two incident fields until  $\theta$  is near  $90^\circ$ . This is done adiabatically with the product of the fields changing slowly, as compared to the separation of the eigenvalues. Instead, at a fixed field, one may allow  $\Delta\omega$  to chirp from an initial negative value toward zero.

Before describing the numerical simulation, we first describe the ideal case that motivated this work: In the hypothetical ideal case where the matrix elements in Eq. (3) and all of the  $E_q$  are equal and where the molecular transition frequency is very small compared to the average energy of the electronic states, the constants  $a_q$ ,  $b_q$ ,  $c_q$ , and  $d_q$  would be approximately equal. With  $\rho_{ab} = -0.5$ , the right-hand side of Eq. (4) is zero and one would expect a small phase variation with distance and thus an effectively small refractive index. This is in the spirit of EIT, where the coherence terms have a sign opposite to the diagonal terms. Conversely, if there were no broad spectral comb of frequencies and only two sidebands were present, then from Eq. (4) we see that, at maximum coherence, the reduction in the refractive index would be only 50%.

A typical real system becomes more complicated and requires a numerical calculation. The reasons for this include the many vibrational states and Franck-Condon factors (or their equivalent) that cause  $b_q c_q < a_q d_q$  and, also, the termination of the spectrum on the low-frequency side, which, in turn, limits its growth [Eq. (5)] on the high-frequency side.

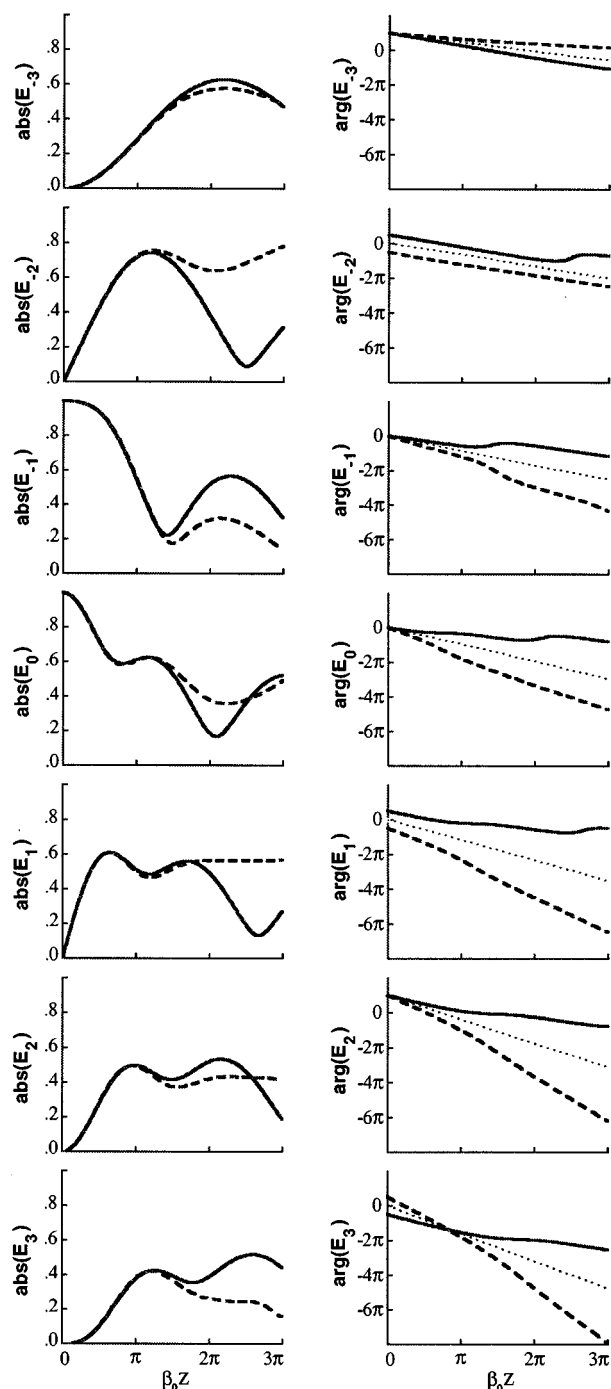


FIG. 2. Normalized amplitude (left) and phase (right) variation, with the distance of the central sidebands generated in parahydrogen. In all plots the solid lines show the sidebands generated by the antiphased (or dark) state. The dashed lines show the sidebands generated by the phased state. For comparison, the phase plots (dotted) show the phase accumulation of a single sideband if propagating alone.

Our numerical simulation is based on parahydrogen with linearly polarized fields with all molecules in the  $J=0$  ground state. The constants  $a_q$ ,  $b_q$ ,  $c_q$ , and  $d_q$  include the contributions of the 0th–36th vibrational transitions of the Lyman band and the 0th–13th transitions of the Werner band. Oscillator strengths and Franck-Condon factors are ob-

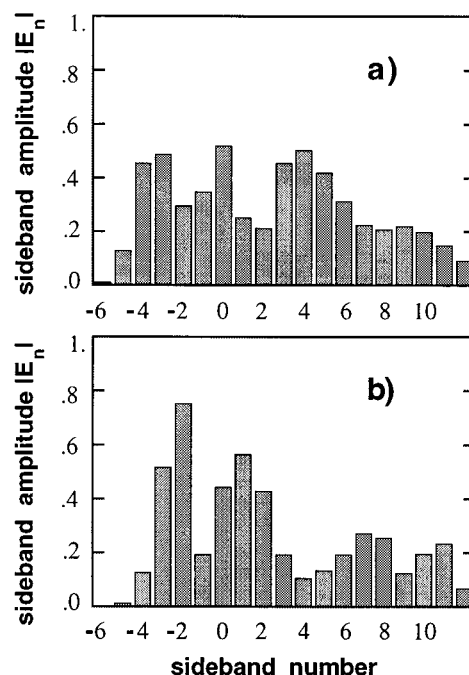


FIG. 3. Normalized sideband amplitudes in parahydrogen: (a) as generated by the antiphased state and (b) as generated by the phased state. In this plot the density length product corresponds to a total phase retardation of  $3\pi$  for the zeroth sideband. At a pressure of 10 atm, this retardation is obtained in a 1-mm-long cell.

tained from Allison and Dalgarno [14] and energies are obtained from Herzberg [15]. The applied laser frequencies are  $\omega_{-1}=24\,100\text{ cm}^{-1}$  (frequency-doubled Ti:sapphire) and  $\omega_0=28\,200\text{ cm}^{-1}$  [frequency-tripled Nd:YAG (neodymium-doped yttrium aluminum garnet)].

We solved Eq. (4) assuming that all the molecules are in the  $|+\rangle$  state [Eq. (6a)]. The power densities of  $E_{-1}$  and  $E_0$  at  $z=0$  are both  $10^{11}\text{ W/cm}^2$ . All other sidebands between  $q=-6$  and 14 have an amplitude of zero at  $z=0$ . Distance is measured in units of  $\beta_0 z$ , which is the phase shift that the zeroth sideband, if alone, would accumulate as it propagates. The density and length are such that this phase accumulation would be  $3\pi$  rad. At a pressure of 10 atm, a  $3\pi$ -rad phase shift is attained in a length of 1.0 mm.

Figure 2 shows numerical results for the amplitude and phase variation with distance of the central sidebands, as driven by either the antiphased or phased states ( $\Delta\omega$  negative or positive, respectively). For comparison, the phase plots also show the phase accumulation of a single sideband if propagating alone. If plots such as these are continued to larger total phase shifts, energy may flow back toward the zeroth sideband and the behavior becomes more complex. However, the average reduction in the refractive index remains roughly the same. Figure 3 shows the amplitudes at  $3\pi$  rad of the spectrum generated by both the antiphased and phased molecular superposition states.

The main conclusion of this work is that the self-protection of the refractive indexes as provided by the generated broadband spectral structure, though not perfect, is still substantial; and the difference in the behavior of the refractive indexes of the spectrum, as generated by the

phased and antiphased molecular superposition states, should be readily observable. By changing only the sign of the detuning one should observe a reduction (or increase) in the right-angle Rayleigh scatter. For the  $q=0$  sideband, the predicted ratio of the Rayleigh scattering,  $\sim(n-1)^2$ , for the phased, as compared to the antiphased state, and averaged over the cell length is 11.

In the past, EIT has only been studied, or experimentally demonstrated [7], for detunings from the states  $|i\rangle$ , which are sufficiently small that the rotating-wave approximation applies. By operating at maximum coherence and allowing for the full generated spectrum, extensions to systems with detunings on the order of  $10^5 \text{ cm}^{-1}$  become possible. Many questions remain: For example, can a single two-photon detuning be used to compensate for many degenerate or near-degenerate rotational states?

We note the relation of this work to the problem of the generation of subfemtosecond time-scale structure: The technique described here should allow the generation of a coherent spectrum of sidebands with a width exceeding  $75\,000 \text{ cm}^{-1}$ . Though the spectrum does not Fourier transform to a train of pulses, there will be periodic structure on a time scale of the inverse of this bandwidth, i.e.,  $\approx 0.1 \text{ fsec}$ . It may also be possible to learn to adjust or compensate for phase variation of the sidebands.

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