

Eigenvectors of a Raman Medium

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We show the existence of discrete sets of Raman sidebands which self-consistently establish a Raman coherence and propagate without change in amplitude and relative phase. Equivalently, there exist periodic femtosecond-time-scale, temporal pulse shapes which propagate without change in shape.

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Two laser beams whose frequency difference is close, but not exactly equal, to that of a Raman resonance in a molecular medium will drive the resonance so as to establish a phased and, in effect, propagating molecular coherence. If the magnitude of this coherence ρ_{ab} is comparable to the magnitude of ρ_{aa} and ρ_{bb} (Fig. 1), the effective distance for the generation of additional Raman sidebands is comparable to the distance in which the relative phase of adjacent sidebands would slip by π radians. When this is the case, it is predicted that the medium will generate a collinearly propagating comb of Raman sidebands which, for vibrational scattering in H_2 , has a spectral width exceeding $60\,000\text{ cm}^{-1}$ [1].

By using spectral pulse modification techniques [2,3] it is, in principle, possible to independently adjust the relative amplitude and phase of each spectral component of such a Raman-generated spectral comb. One is, therefore, motivated to ask the question: Do there exist sets of Raman sidebands which will self-consistently establish the molecular coherence and propagate without change in amplitude or relative phase? Equivalently, do there exist periodic trains of femtosecond-time-scale optical pulses which establish the coherence and propagate without changing shape?

In this Letter we discuss the conditions for the existence of Raman eigenvectors as described in the previous paragraph. These conditions are as follows: (i) In analogy to electromagnetically induced transparency (EIT) in atoms, one must use the antiphased molecular state. (ii) There is a requirement on the magnitude of the Raman polarizability as compared to the background dispersive polarizability. (iii) The pulse train as a whole (for example, 20-ns long for typical Q -switched lasers) must have an energy which is sufficiently large that the total number of photons is large as compared to the number of molecules in the laser path. When these conditions are satisfied, also in analogy to EIT, the Raman coherence is self-consistently prepared by pulses in the front of the pulse train. Once prepared, assuming infinite dephasing time of the Raman transition, the medium will transmit all further pulses without change of pulse shape.

Before proceeding we note that there has been considerable theoretical and experimental work on multifrequency, on-resonance Raman generators [4,5]. The possibility of

Raman solitons has been noted [6]. Off-resonance excitation has been suggested as a means for obtaining a reduction in the refractive index [1] and also for generating a sinusoidally, frequency-modulated beam which, by using group velocity dispersion, can be temporally compressed into a train of subfemtosecond pulses [7]. Other pertinent work is listed in Refs. [8–10].

We begin by assuming monochromatic fields and showing the existence of self-consistent sets of Raman sidebands (eigenvectors) which propagate without change in amplitude or relative phase and, at the same time, produce the specified coherence. In the latter portion of this Letter, we numerically show how such eigenvectors and associated coherence may be established by applying smooth field envelopes to molecules which are initially in the ground state.

Proceeding as in Harris and Sokolov [1] and noting Fig. 1, the angular frequencies of the Raman sidebands are $\omega_q = \omega_0 + q(\omega_b - \omega_a - \delta\omega) = \omega_0 + q\omega_m$. The frequencies of the ground and excited vibrational-rotational states are ω_a and ω_b and the detuning from Raman resonance is $\delta\omega$. We assume the ideal case of zero linewidth for the Raman transition. We allow for an arbitrary number of states $|i\rangle$ with energies $\hbar\omega_i$ and matrix elements from states $|a\rangle$ and $|b\rangle$ to these states

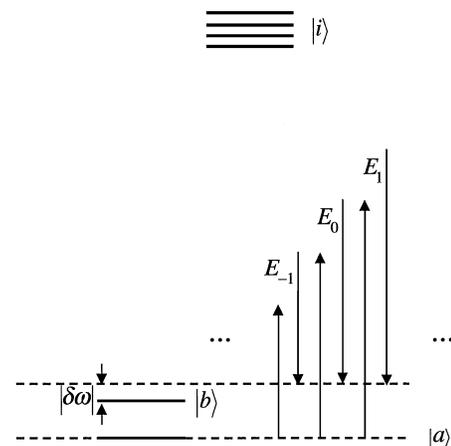


FIG. 1. Energy level schematic showing the interaction of Raman sidebands with the molecular states. In the configuration shown, the two-photon detuning $\delta\omega$ is negative.

μ_{ai} and μ_{bi} . By assuming that the derivatives of the probability amplitudes of the upper states $|i\rangle$ are small as compared to the detunings from these states, the problem may be written in terms of an effective Hamiltonian

$$H_{\text{eff}} = -\frac{\hbar}{2} \begin{bmatrix} A & B \\ B^* & D - 2\delta\omega \end{bmatrix}. \quad (1)$$

The quantities $A/2$ and $D/2$ are the Stark shifts of states $|a\rangle$ and $|b\rangle$, respectively, and B is the effective Rabi frequency. With the dispersion and coupling constants defined as

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

the matrix elements in Eq. (1) are $A = \sum_q a_q |E_q|^2$, $B = \sum_q b_q E_q E_{q-1}^*$, and $D = \sum_q d_q |E_q|^2$.

Working in local time, $\tau = t - (z/c)$, the one-dimensional propagation equation for the q th sideband is

$$\begin{aligned} \frac{\partial E_q}{\partial z} &= -j\eta\hbar\omega_q N (a_q \rho_{aa} E_q + d_q \rho_{bb} E_q \\ &\quad + b_q^* \rho_{ab} E_{q-1} + b_{q+1} \rho_{ab}^* E_{q+1}), \end{aligned} \quad (3)$$

where N is the number of molecules per volume and $\eta = (\mu/\epsilon_0)^{1/2}$.

Using the effective Hamiltonian of Eq. (1), the equations for the density matrix elements, also in local time, are

$$\begin{aligned} \frac{\partial \rho_{ab}}{\partial \tau} - j \left(\frac{A}{2} - \frac{D}{2} + \delta\omega \right) \rho_{ab} &= j \frac{B}{2} (\rho_{bb} - \rho_{aa}), \\ \frac{\partial \rho_{bb}}{\partial \tau} &= -\text{Im}(B^* \rho_{ab}), \end{aligned} \quad (4)$$

where $\rho_{aa} = 1 - \rho_{bb}$. We assume that at $\tau = 0$ all the molecules are in a single, nondegenerate ground state; therefore, $\rho_{aa}(z, 0) = 1$ and $\rho_{ab}(z, 0) = \rho_{bb}(z, 0) = 0$.

If the elements of the effective Hamiltonian vary slowly as compared to the separation of the eigenvalues and with $B = |B| \exp j\varphi$, the solution of the density matrix which evolves smoothly from the ground state is

$$\begin{aligned} \rho_{ab} &= \left(\frac{1}{2} \sin\theta \right) \exp j\varphi; & \rho_{bb} &= \sin^2 \left(\frac{\theta}{2} \right), \\ \tan\theta &= \frac{2|B|}{(2\delta\omega - D + A)}. \end{aligned} \quad (5)$$

In molecular systems with large one-photon detunings $A \cong D$. The sign of the detuning $\delta\omega$ from Raman resonance determines the sign of the coherence ρ_{ab} . If $\delta\omega$ is negative, then ρ_{ab} has the opposite sign than its driving

two-photon Rabi frequency B . We, therefore, refer to this coherence as antiphased. We will see that, for a normally dispersive medium, only an antiphased coherence will lead to the eigenvectors of this work.

We choose the time origin so that ρ_{ab} is real and define a vector \bar{E} with components $E_{-q}, \dots, E_0, \dots, E_q$. The propagation equations may then be written as

$$\frac{d\bar{E}}{dz} = \bar{M} \cdot \bar{E}. \quad (6)$$

From Eq. (3), the nonzero matrix elements of \bar{M} , each multiplied by $-j\eta\hbar\omega_q N$ are

$$\begin{aligned} M_{q,q-1} &= b_q^* \rho_{ab}, \\ M_{q,q} &= a_q \rho_{aa} + d_q \rho_{bb}, \\ M_{q,q+1} &= b_{q+1} \rho_{ab}^*. \end{aligned}$$

We seek self-consistent solutions of the steady-state density matrix [Eq. (5)] and the propagation equation [Eq. (6)]. If the density matrix elements are independent of distance, then a solution of Eq. (6) is

$$\bar{E}(z) = \xi_k \bar{v}_k \exp j\kappa_k z, \quad (7a)$$

where

$$\bar{M} \cdot \bar{v}_k = j\kappa_k \bar{v}_k. \quad (7b)$$

\bar{v}_k and $j\kappa_k$ are the normalized eigenvectors and eigenvalues of \bar{M} at an assumed value of coherence such that $\theta(z) = \theta(0) = \theta_0$ and $\varphi(z) = \varphi(0) = 0$. With $\delta\omega$ chosen, we define $A|_{\bar{v}_k} = A_0$, $B|_{\bar{v}_k} = B_0$, $D|_{\bar{v}_k} = D_0$. Consistency requires

$$\frac{2|\xi_k|^2 |B_0|}{2\delta\omega - |\xi_k|^2 D_0 + |\xi_k|^2 A_0} = \tan\theta_0, \quad (8a)$$

$$\text{Arg}(B_0) = \varphi(0) = 0. \quad (8b)$$

If a solution for $|\xi_k|^2$ exists, then the eigenvector \bar{v}_k propagates with a constant state of the molecular system [$\theta(z) = \theta_0$]. In the frequency domain, the components E_q of the eigenvector propagate with unchanged amplitude. Each sideband accumulates an absolute phase of $\kappa_k z$ so that the relative phase of the sidebands is unchanged. Equivalently, the Fourier transform of these sidebands is a periodic function of time which is independent of distance.

We find numerically that, for a normally dispersive medium, for Eq. (8b) to be satisfied, the quantity θ_0 must be negative. Equivalently, only the antiphased state (obtained by tuning above the Raman resonance) will yield self-consistent eigenvectors. In molecules, the dispersive parameters a_q and d_q are often of the same magnitude. When this is the case, the ratio of the coupling parameter b_q to either of these parameters determines the range of θ_0 (i.e., coherence) over which a solution exists. Because of the Franck-Condon factors, $0 < |b/a| < 1$; at the lower limit, the only self-consistent eigenvectors are those which approximate a single frequency when propagating alone. For intermediate values of $|b/a|$, solutions for $|\xi_k|^2$ exist

for some values of θ_0 but not for others; at sufficiently large $|b/a|$, solutions exist for all values of θ_0 .

The fundamental rotational transition of H_2 ($\omega_m = 354 \text{ cm}^{-1}$) is an example where the ratio $|b/a|$ is large and where the matrix elements (for Lyman and Werner bands) are known [7,11]. With states $|a\rangle$ and $|b\rangle$ as the $J'' = 0$ and $J' = 2$ rotational states of the H_2 ground vibrational level and an applied center frequency of $\omega_0 = 25\,500 \text{ cm}^{-1}$ (frequency-doubled Ti:sapphire), $|b/a| = 0.67$. Figure 2 shows the spectral components of an eigenvector which is generated at a coherence of $\rho_{ab} = -0.025$. For this eigenvector, about ten sidebands have substantial amplitudes. In time domain this eigenvector corresponds to a combination of amplitude and frequency modulation, with both modulation frequencies equal to the oscillation frequency and phased such that the peak in amplitude corresponds to the peak in frequency. For $N = 2.23 \times 10^{19} \text{ molecules/cm}^3$ the eigenvalue $\kappa_k = -10.7 \text{ rad/cm}$ is a measure of the phase retardation of the eigenvector as an entity. To this accuracy, the phase accumulation of the zeroth sideband, if propagating alone in an incoherent medium, is the same. If two sidebands propagate through the medium with $\rho_{ab} = -0.025$ and $N = 2.23 \times 10^{19} \text{ molecules/cm}^3$, approximately one additional sideband is generated after every 2 cm of propagation [7].

In general, we choose the basis set large as compared to the number of sidebands which have substantial amplitude. When this is the case truncation of the $\overline{\overline{M}}$ matrix does not affect the existence or shape of the eigenvectors. The reason for this is that $\overline{\overline{M}}$ is tridiagonal and each sideband is coupled only to its nearest neighbors.

We next consider the dynamic preparation of the medium. To do this we assume that at $\tau = 0$ all of the

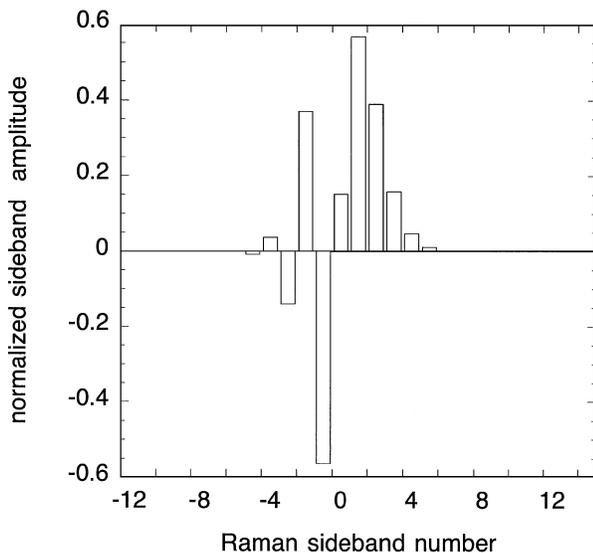


FIG. 2. An example of an eigenvector for the rotational transition in H_2 . The molecular coherence is $\rho_{ab} = -0.025$.

molecules are in the ground state. At $z = 0$ a smoothly rising pulse with a rise time of 10 ns is imposed on each component E_q of the selected eigenvector. The density matrix and propagation equations [Eqs. (3) and (4)] are solved numerically by the method of lines; at each point in the cell we solve the time domain density matrix equations for the matrix elements and use these elements to advance the propagation equations to the next step.

The results of this numerical simulation for the eigenvector of Fig. 2 are shown in Figs. 3 and 4. These figures assume H_2 at a density of $N = 2.23 \times 10^{19} \text{ molecules/cm}^3$ and a cell length of 35 cm. At $z = 0$ the power per area of center sideband is $1.15 \times 10^8 \text{ W/cm}^2$, corresponding to $|\xi_k| = 1.9 \times 10^6 \text{ V/cm}$.

Figure 3 shows, with the same normalization as Fig. 2, the amplitudes of several components of this eigenvector as a function of time, evaluated at $z = 0$ and $z = 35 \text{ cm}$. One notes that in the preparation period, i.e., for times less than about 24 ns, the amplitudes of the components differ from their steady-state values. After preparation is complete there are no further changes. Figure 4 shows the phase accumulation for these same sidebands. Following preparation, all sidebands accumulate a common phase $\kappa_k L$. In analogy with EIT [12], we have found that the total number of photons in the preparation time is within a factor of 10 (depending on the magnitude

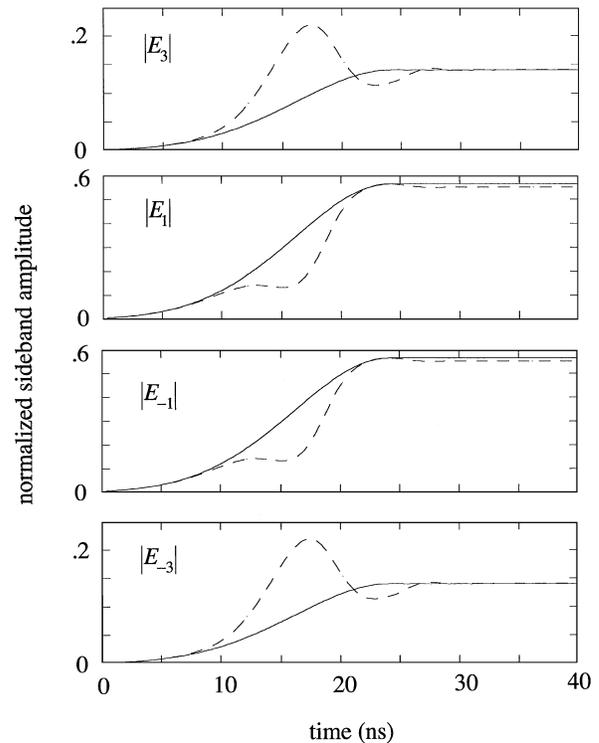


FIG. 3. The amplitudes of E_{-3} , E_{-1} , E_1 , and E_3 at $z = 0$ (solid line) and at $z = 35 \text{ cm}$ (dashed line) of the H_2 cell at a density of $N = 2.23 \times 10^{19} \text{ molecules/cm}^3$. After all sidebands have evolved to their steady-state values, the envelopes at $z = 0$ and at $z = 35$ coincide.

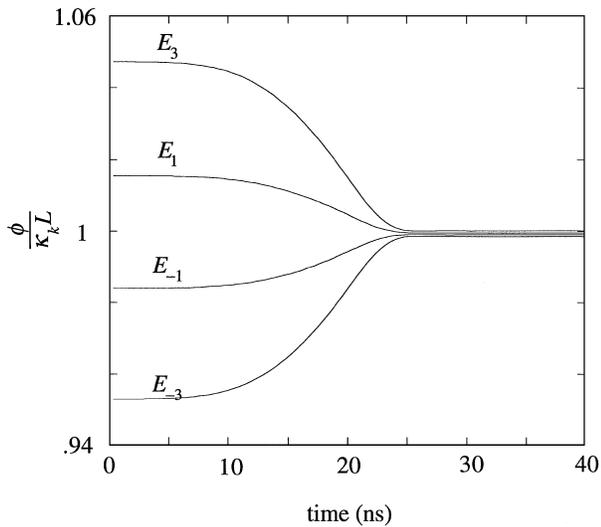


FIG. 4. Normalized phase shifts (ϕ) between $z = 0$ and $z = L = 35$ cm of the respective sidebands. After preparation, all sidebands accumulate a common phase $\kappa_k L$.

of the coherence) of the number of molecules in the laser path.

This work has shown that there are sets of Raman sidebands which self-consistently establish a molecular coherence and propagate without change in amplitude or relative phase. Equivalently, even in a dispersive medium, there exist periodic trains of femtosecond-time-scale optical pulses which propagate without changing shape. In common with EIT in three-state systems, these effects require sufficient pulse energy to establish an antiphased coherence.

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