

Subfemtosecond Pulse Generation by Molecular Modulation

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We suggest a technique for producing subfemtosecond pulses of radiation. The technique is based on using electromagnetically induced transparency to produce a strongly driven molecular coherence. This coherence results in a Raman spectrum with Bessel function amplitudes and phases corresponding to a frequency modulated signal, thereby allowing compression by the group velocity dispersion of the same medium. [S0031-9007(98)07282-2]

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This Letter suggests and analyzes a technique for producing subfemtosecond pulses of radiation. We will show that two laser beams whose frequency difference is slightly offset from a molecular transition will, for an appropriate choice of gas pressure and cell length, generate a spectrum of Raman sidebands whose Fourier transform is a periodic train of subfemtosecond pulses. The essence of the technique is the concurrent generation of a frequency modulated (FM) waveform and the use of group velocity dispersion to temporally compress this waveform. By numerically modeling this process in molecular deuterium (D_2), we calculate a generated train of pulses with a pulse spacing of 11.1 fs and a pulse length of 0.21 fs.

The coherence ρ_{ab} (Fig. 1) of the driven molecular transition is central to this work. This coherence is established by driving the molecular transition slightly off resonance with driving lasers of sufficient intensity that the product of their Rabi frequencies exceeds the product of the detuning from the molecular electronic states and the detuning from the Raman transition. With the linewidth of the applied laser pulses small as compared to the Raman detuning, the magnitude of the molecular coherence approaches 0.5, and its sign is determined by the sign of the Raman detuning.

For the conditions of the previous paragraph, the generation and phase-slip lengths are of the same order, and Raman sideband generation proceeds collinearly and very differently than in the conventional low coherence regime. In essence, the molecular motion now modulates the electronic refractive index much in the same way that a low frequency electric field modulates the refractive index of a polar crystal. Incident optical frequencies become frequency modulated with peak (sinusoidal) frequency deviations and spectral bandwidths that substantially exceed the width of the visible spectrum. This allows the extension of compression techniques, such as the use of group velocity dispersion as described here, to a new regime of short pulse compression.

In pertinent prior work, (1) the authors have noted how an electromagnetically induced transparency (EIT)-like excitation may be used to generate a broad comb of coherent sidebands, but the Bessel-function nature of the spectrum and the possibility of pulse compression were not

observed [1]. (2) Hakuta and colleagues have experimentally demonstrated the generation of collinear Raman sidebands in solid molecular hydrogen [2]. (3) Kawano *et al.* have demonstrated the use of stimulated rotational Raman scattering to generate a broad spectrum and discussed possibilities for mode locking this spectrum [3]. (4) There has been considerable work in the area of on-axis Raman sideband generation [4]. (5) There is also some relation to the work of Kaplan and Shkolnikov on Raman solitons [5]. (6) Workers in the field of high-order harmonic generation have noted the possibility of obtaining a short time structure [6]. (But, in high-order harmonic generation, less than one part in 10^{-4} of the incident pulse energy is converted into the phased spectrum; by the approach of this work, near-complete conversion is likely.) (7) There is now considerable work on EIT [7] and related subjects [8]. (8) It has been shown that, by using a large atomic coherence, nearly complete frequency conversion will occur in a single coherence length [9].

The body of this Letter divides into four parts: (a) a summary of the formalism describing the interaction of the multilevel system of Fig. 1 with a comb of Raman

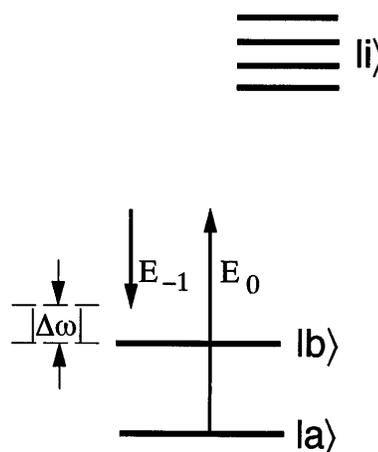


FIG. 1. Energy level schematic for establishing coherence ρ_{ab} in a molecular system. Laser fields are applied at the frequencies of the $q = 0$ and $q = -1$ sidebands. Here, the detuning $\Delta\omega$ from Raman resonance, as is required for establishing the antiphased state, is negative.

sidebands [1], this formalism includes all dispersive effects; (b) the FM solution for the ideal case of negligible dispersion; (c) a numerical simulation showing concurrent spectral generation and pulse compression in D_2 ; (d) a brief discussion of measurement possibilities.

We will work with the envelope quantities $E_q(z, t)$ and $\rho_{ab}(z, t)$ for electromagnetic fields and density matrix elements such that the full time and space quantities are $\hat{E}_q(z, t) = \text{Re}\{E_q(z, t) \exp[j(\omega_q t - k_q z)]\}$ and $\hat{\rho}_{ab}(z, t) = \text{Re}\{\rho_{ab}(z, t) \exp[j(\omega_m t - k_m z)]\}$, with $\omega_q = \omega_0 + q(\omega_b - \omega_a - \Delta\omega) = \omega_0 + q\omega_m$. The modulation frequency ω_m or, equivalently, the frequency difference between adjacent sidebands, is equal to the difference of the two applied frequencies ω_0 and ω_{-1} . The two-photon detuning $\Delta\omega$ is the difference of ω_m and the Raman transition frequency and, as shown in Fig. 1, is negative; $k_q = \omega_q/c$ and $k_m = \omega_m/c$.

We allow for an arbitrary number of virtual states $|i\rangle$ with energies $\hbar\omega_i$. The matrix elements from states $|a\rangle$ and $|b\rangle$ to these states are μ_{ai} and μ_{bi} , respectively. When the derivatives of the probability amplitudes of the upper states $|i\rangle$ are small as compared to the detunings from these states, the problem can be written in terms of an effective, distance-dependent, two-by-two Hamiltonian [1]:

$$H_{\text{eff}} = -\frac{1}{2} \begin{bmatrix} A & B \exp(-j\frac{\omega_m}{c} z) \\ C \exp(j\frac{\omega_m}{c} z) & D - 2\Delta\omega \end{bmatrix}, \quad (1)$$

where $A = \sum_q a_q |E_q|^2$; $B = \sum_q b_q E_q E_{q-1}^*$; $C = B^*$; and $D = \sum_q d_q |E_q|^2$. All rotating and nonrotating, as well as cross-transition, terms are retained within this Hamiltonian. We also assume the ideal case of zero linewidth for the $|a\rangle$ - $|b\rangle$ transition. The constants a_q , b_q , and d_q determine the dispersion and coupling and are

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

We assume that all of the molecular population is initially in the ground state and the envelopes of the exciting laser pulses vary slowly as compared to the separation of the eigenvalues of the Hamiltonian of Eq. (1). This allows adiabatic preparation of all of the molecules in one eigenstate. By choosing the sign of the detuning $\Delta\omega$, either the phased or antiphased eigenstate is selected at the input of the medium and is, as shown below, maintained throughout the medium. The molecular coherence ρ_{ab}

for the phased state has the same sign as the quantity B in Eq. (1) and, for the antiphased state, has the opposite sign. Both eigenstates lead to an FM signal but, as shown below, in a normally dispersive medium, with the boundary condition of two incident frequencies, only the antiphased state allows pulse compression.

Defining

$$B = |B| \exp(j\varphi); \quad \tan \theta = \frac{2|B|}{2\Delta\omega - D + A}, \quad (3)$$

the eigenstate, which adiabatically evolves from the ground state as field amplitudes are increased, and the coherence, which is obtained from it, are as follows:

$$\begin{aligned} |+\rangle &= \cos \frac{\theta}{2} \exp\left(j\frac{\varphi}{2}\right) |a\rangle \\ &+ \sin \frac{\theta}{2} \exp\left(-j\frac{\varphi}{2}\right) |b\rangle, \end{aligned} \quad (4)$$

$$\rho_{ab} = \frac{1}{2} \sin \theta \exp(j\varphi),$$

where positive θ corresponds to the phased state and negative θ to the antiphased state. By choosing the intensities of the applied fields and the detuning $\Delta\omega$, the quantity $\sin \theta$ can be set to any value between -1 and 1 .

The propagation equation for the q th sideband in local time is

$$\begin{aligned} \frac{\partial E_q}{\partial z} &= -j\eta\hbar\omega_q N (a_q \rho_{aa} E_q + d_q \rho_{bb} E_q + b_q \rho_{ab} E_{q-1} \\ &+ c_q \rho_{ab}^* E_{q+1}), \end{aligned} \quad (5)$$

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

In order to understand the numerical results which follow, we first exam the analytically solvable case of negligible dispersion and limited modulation bandwidth. We take all of the sidebands to be sufficiently far from the resonances that, in Eq. (2), $a_q = b_q = c_q = d_q \equiv a_0$. We define propagation constants $\beta_q = \eta\hbar\omega_q N a_0$ and change variables by $E_q = \tilde{E}_q \exp(-j\beta_q z)$, $\rho_{ab} = \tilde{\rho}_{ab} \exp(-j\beta_m z)$. With these approximations and definitions, Eq. (5) becomes

$$\frac{\partial \tilde{E}_q}{\partial z} = -j\beta_q (\tilde{\rho}_{ab} \tilde{E}_{q-1} + \tilde{\rho}_{ab}^* \tilde{E}_{q+1}). \quad (6)$$

The quantity $\tilde{\rho}_{ab} = \frac{1}{2} \sin \tilde{\theta} \exp(j\tilde{\varphi})$, where $\tilde{\theta}$ and $\tilde{\varphi}$ are given by Eq. (3) with E_q replaced by \tilde{E}_q . As the boundary conditions for the propagation equation, we assume that the fields $E_0(0)$ and $E_{-1}(0)$ are applied at the input of the medium and all other Raman sidebands are generated in the medium.

Although we will not make this approximation in the numerical work which follows, here we assume a restricted modulation bandwidth such that all ω_q are equal to ω_0 and $\beta_q = \beta_0$. By using the Bessel-function identities $2 \frac{\partial J_n(x)}{\partial x} = [J_{n-1}(x) - J_{n+1}(x)]$ and $\sum_n J_{n+q}(x) J_{n+p}(x) = \delta_{pq}$, it may be verified that, with $\gamma = \beta_0 \sin[\theta(0)]$, the solution $\tilde{\rho}_{ab}(z) = \rho_{ab}(0)$ and

$$\tilde{E}_q(z) = E_0(0) \exp\left[j\left(\varphi(0) - \frac{\pi}{2}\right)q\right] J_q(\gamma z) + E_{-1}(0) \exp\left[j\left(\varphi(0) - \frac{\pi}{2}\right)(q+1)\right] J_{q+1}(\gamma z) \quad (7)$$

satisfies both density matrix and propagation equations and also the boundary condition at $z = 0$. [$\varphi(0)$ is obtained from Eq. (3).] With $\sum_n J_n(x) \exp(jn\omega t) = \exp[jx \sin(\omega t)]$, the time domain signal is

$$E(t) = \text{Re}\{E_0(0) \exp j[\omega_0 t + \gamma z \cos(\omega_m t + \varphi(0))] + E_{-1}(0) \exp j[\omega_{-1} t + \gamma z \cos(\omega_m t + \varphi(0))]\}. \quad (8)$$

Equation (8) is a superposition of two FM signals with center frequencies corresponding to the frequencies which are applied at $z = 0$ and a peak phase deviation γz . It is important that the frequency modulation leaves the eigenvectors of the Hamiltonian unchanged.

The total number of sidebands generated in a cell of length L is approximately equal to $2\gamma L$. The assumption of limited modulation bandwidth ($\omega_q = \omega_0$) implies that $\gamma z \ll \omega_0/\omega_m$; i.e., the peak frequency excursion must be small as compared to the center frequency. We have checked that, when this assumption is satisfied, the solution of Eq. (7) coincides with the exact numerical solution of Eq. (6).

Equations (7) and (8) hold for both the antiphased ($\sin \theta < 0$) and the phased case ($\sin \theta > 0$). But, in the antiphased case, the higher frequencies (blue) are temporally advanced with regard to the lower frequencies (red), while, in the phased case, red is advanced as compared to blue. When $E_0(0) \sim E_{-1}(0)$, only the signal generated by the antiphased state allows pulse compression in a normally dispersive medium. Figure 2(a) shows the time domain signal produced by the antiphased state; i.e., an FM signal with an envelope at the beat frequency ω_m .

Pulse compression is obtained by setting the group velocity delay which corresponds to the frequency deviation

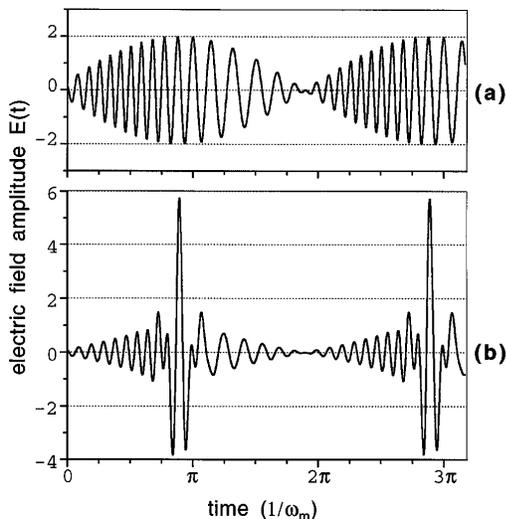


FIG. 2. Electric field amplitude vs time (a) as produced by an antiphased state without pulse compression. The parameters are $E_0(0) = E_{-1}(0) = 1$, $\omega_0/\omega_m = 18$, and $\gamma L = 7$. (b) Following pulse compression in a medium with a frequency-invariant group velocity dispersion and a length determined by Eq. (9).

$\omega_d = \omega_m \gamma L$ equal to $\pi/2\omega_m$. Here, but not in the numerical work which follows, we assume a frequency-invariant group velocity dispersion $\partial^2 k/\partial \omega^2$. The medium length l which is necessary for pulse compression is then determined by

$$\frac{\partial^2 k}{\partial \omega^2} \Big|_{\omega=\omega_0} l = \frac{\pi}{2\omega_m^2 \gamma L}. \quad (9)$$

Figure 2(b) shows the signal which is obtained when the signal of Fig. 2(a) is transmitted through a medium of this length. Pulse compression with a ratio of pulse width to pulse separation of about $1/2\gamma L$ is observed.

We turn next to the numerical solution for D_2 . We do not make any of the dispersive approximations described above. We find that both generation and pulse compression may be accomplished concurrently in the same medium. We assume E_0 and E_{-1} to be applied at $z = 0$ and retain sidebands between $q = -9$ and $q = 18$. The solution of Eq. (5) is obtained by forward stepping from $z = 0$ with the density matrix elements recalculated at each step.

We consider the fundamental vibrational transition in D_2 with $\omega_m = 2994 \text{ cm}^{-1}$ and take all molecules in the $J = 0$ ground state. The constants a_q , b_q , and d_q include the contributions of the 0 – 52nd vibrational transitions of the Lyman band and the 0 – 18th transitions of the Werner band. Oscillator strengths and Franck-Condon factors are obtained from Allison and Dalgarno and energies are obtained from Herzberg [10]. The D_2 density and the total cell length are chosen such that the zeroth sideband, if propagating alone, would accumulate a phase shift $\beta_0 L = 14\pi$ rad. At a pressure of 1 atm, this corresponds to $L = 3.6$ cm.

We envision using single-mode pulsed lasers with pulse lengths of ~ 5 ns and pulse energies of ~ 50 mJ. We take the applied laser frequencies to be $\omega = 28\,183 \text{ cm}^{-1}$ (frequency-tripled Nd:YAG) and $\omega_{-1} = 25\,189 \text{ cm}^{-1}$ (frequency-doubled Ti:sapphire). We choose the Raman detuning $\Delta\omega = -1$ GHz to be much larger than both the Raman linewidth (~ 0.3 GHz for D_2 at STP) and the laser linewidth (~ 0.1 GHz for a single-mode, 5-ns-pulse laser). We set the power densities of E_0 and E_{-1} at $z = 0$ to 10^{11} W/cm^2 . Evaluating Eqs. (1)–(4), $\sin \theta(0) \approx -0.9$. The coherence $|\rho_{ab}| \approx 0.45$ and the cell length are set such that about twenty sidebands are generated, and the concurrent group velocity delay is correct for pulse compression.

Figure 3 shows spectral and temporal evolution as a function of distance for the conditions of the previous

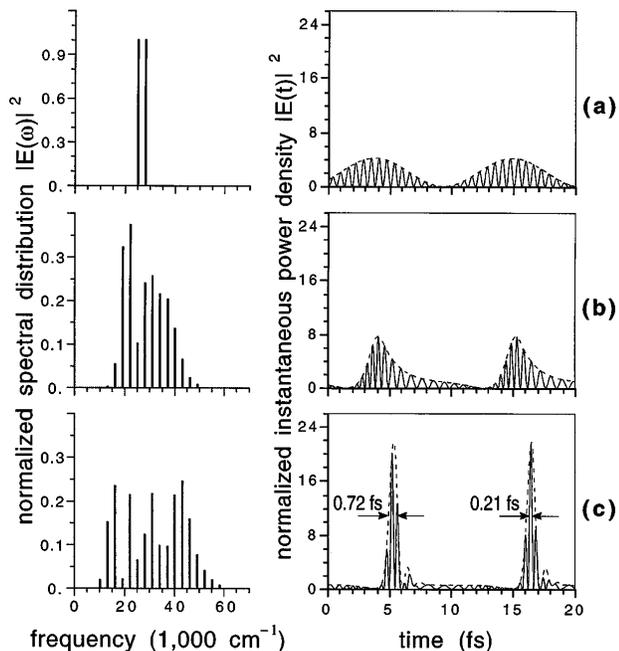


FIG. 3. Frequency spectrum (left) and instantaneous power density vs time (right) in D_2 at 1 atm pressure with applied power densities at $z = 0$ such that $|\rho_{ab}| \approx 0.45$. Parts (a)–(c) show the spectrum and temporal waveform for (a) $z = 0$ cm, (b) $z = 1.8$ cm, and (c) $z = 3.6$ cm. Sideband powers are normalized to those of the incident fields. The dashed lines on the time domain plots show the envelopes of possible single-cycle pulses.

paragraph. Sideband frequencies are predicted to range from about $1.4 \mu\text{m}$ in the infrared to 150 nm in the vacuum ultraviolet. At the appropriate cell length these frequencies synthesize a train of pulses with a pulse separation of 11.1 fs and a pulse length of 0.21 fs . We find sharp pulse compression at experimental conditions which are readily realizable. Because the ratio of the applied optical frequencies to the Raman transition frequency is, in general, noninteger, the peak of the optical cycle will drift from pulse to pulse within a narrow envelope. This envelope (width = 0.72 fs) is shown by the dashed line in each of the time domain plots of Fig. 3. For the numerical analysis we have treated the driving fields as monochromatic. For the pulsed excitation, as described above, we expect these results to apply for a time about equal to the dephasing time of the Raman transition, and therefore expect nearly all of the incident power to be converted into the periodic train of subfemtosecond pulses.

The predictions of this numerical analysis should be verifiable by two methods. The relative intensities of the sidebands could be measured; these differ in the phased and antiphased states, and are correlated with the phase of each sideband. In the time domain, the peak power density grows on a scale of 3 cm and varies in a predictable manner thereafter. In the region of the highest peak power ($\sim 5 \text{ mm}$ long), one might expect multiphoton processes in either D_2 itself or in a dopant to light up as an indication

of the presence of ultrashort pulses. The physics of such processes, as well as other nonlinear behavior in this regime is, in its own right, of considerable interest.

The principle approximations of the numerical results given here are the neglect of the dephasing of the Raman transition, the assumption that the driving fields are both quasimonochromatic and of an infinite extent in the transverse direction, and that all molecules are initially in the $J = 0$ ground state. Although the effects of self-focusing, temporal chirping, saturation of the Raman transition, and thermal population have not been discussed, we do not see contradictions.

In summary, we have described how the ideas of frequency modulation and pulse compression may be extended to the atomic time scale. The technique will allow the generation of pulses of high intensity radiation with a pulse length which is less than the revolution time of an electron in the first Bohr orbit of atomic hydrogen. The use of rotational Raman transitions, instead of vibrational transitions, is promising. For example, the $\nu' = 0, J' = 2 \rightarrow \nu'' = 0, J'' = 0$ rotational transition in H_2 will produce subfemtosecond pulses with a pulse spacing of 94 fs [11].

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