Subfemtosecond pulse generation by rotational molecular modulation

A. V. Sokolov, D. D. Yavuz, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305

Received November 13, 1998

We extend a recent suggestion for the generation of subfemtosecond pulses by molecular modulation [Phys. Rev. Lett. **81,** 2894 (1998)] to the rotational spectrum of molecular hydrogen (H_2). When a rotational transition $|a\rangle \rightarrow |b\rangle$ is strongly driven ($|\rho_{ab}| = 0.5$) the generation and phase-slip lengths are of the same order and the Raman spectrum has approximately Bessel function sideband amplitudes. Numerical simulation predicts that this spectrum (generated in a 14-cm-long cell at 1-atm pressure of H_2) will compress into a train of pulses with 94-fs pulse separation and a pulse length of 0.5 fs.

OCIS codes: 320.0320, 320.5520, 190.3100, 030.1640.

It was recently shown that the ideas of frequency modulation and pulse compression can be extended to molecular oscillation frequencies, thereby permitting generation of subfemtosecond pulses.[1] The essence of this technique is the use of a Raman transition with a sufficiently large coherence that the generation and phase-slip lengths are of the same order. When this is the case, the Raman spectrum is very broad (here, $\sim 70\,000\ \mathrm{cm}^{-1}$) and is generated collinearly. As initially generated, the spectrum has approximately Bessel function sideband amplitudes and in the time domain corresponds to a periodic beat of two frequency-modulated signals. As the waveform propagates, group-velocity dispersion causes temporal compression into the subfemtosecond domain.

The extension of this idea to the rotational, compared with the vibrational, spectrum of H_2 has three significant advantages: (1) the ratio of the molecular coupling constant to the molecular dispersion is three times larger; (2) the spacing of the periodic pulse train is 94 fs, compared with 7.8 fs; and (3) the use of rotational Raman scattering allows a much larger number of sidebands to be generated at the same driving laser intensities, with a total spectral width as large as in the vibrational case. Taken together, these advantages should permit the use of traditional autocorrelation and gating techniques and also, perhaps, spectral modification and recompression techniques.

The idea that a wide spectrum of Raman sidebands, if it is properly phase locked, would be Fourier transformed into a train of subfemtosecond pulses has been discussed by other authors: Kaplan and Shkolnikov have predicted the existence of 2π Raman solitons with a phase-locked spectrum[2]; Imasaka and colleagues have demonstrated the use of stimulated rotational Raman scattering to generate a broad spectrum and discussed possibilities for phase locking this spectrum.[3] In other research on subfemtosecond pulse generation Hänsch proposed a pulse synthesizer that uses separate phase-locked laser oscillators[4]; workers in the field of high-order harmonic generation have noted the possibility of obtaining an attosecond time structure[5]; and Corkum *et al.* proposed a method for a single subfemtosecond pulse generation.[6] In other related work Kocharovskaya and colleagues have suggested using a Raman medium inside a laser cavity to provide phase modulation to cause mode locking[7]; Hakuta *et al.* have experimentally demonstrated the generation of collinear Raman sidebands in solid molecular hydrogen.[8] There has also been considerable research in the area of on-axis Raman sideband generation.[9]

First we present the formalism and give numerical results for simultaneous spectrum generation and pulse compression by rotational Raman scattering in H_2. We then discuss the possibility of ideal phase compensation by spectral filtering with a phase mask.

We consider one-dimensional propagation of a set of quasi-monochromatic Raman sidebands with electric-field envelopes $E_q(z, t)$ such that the total field is $\hat{E}(z, t) = \sum_q \mathrm{Re}\{E_q(z, t)\exp[\, j(\omega_q t - k_q z)]\}$, with $\omega_q = \omega_0 + q(\omega_b - \omega_a - \Delta\omega) = \omega_0 + q\omega_m$. The total space and time quantity for the coherence (off-diagonal density matrix element) of the Raman transition is $\hat{\rho}_{ab}(z, t) = \mathrm{Re}\{\rho_{ab}(z, t)\exp[\, j(\omega_m t - k_m z)]\}$. Two-photon detuning $\Delta\omega$ is the difference of the Raman transition frequency and modulation frequency ω_m, set by the driving fields; $k_q = \omega_q/c$ and $k_m = \omega_m/c$.

We analyze a model molecular system with a Raman transition between states $|a\rangle$ and $|b\rangle$ and an arbitrary number of upper states $|i\rangle$ with energies $\hbar\omega_i$ (Fig. 1). The matrix elements from states $|a\rangle$ and $|b\rangle$ to states $|i\rangle$ are μ_{ai} and μ_{bi}, respectively. When the derivatives of the probability amplitudes of the upper states $|i\rangle$ are small compared with the detunings from these states, the problem can be written in terms of an effective, distance-dependent, two-by-two Hamiltonian[10]:

$$H_{\mathrm{eff}} = -\frac{\hbar}{2}\begin{bmatrix} A & B\exp\left(-j\,\dfrac{\omega_m}{c}\,z\right) \\ C\exp\left(j\,\dfrac{\omega_m}{c}\,z\right) & D - 2\Delta\omega \end{bmatrix}, \tag{1}$$

where $A = \sum_q a_q|E_q|^2$, $B = \sum_q b_q E_q E_{q-1}{}^*$, $C = B^*$, and $D = \sum_q d_q|E_q|^2$. We also assume the ideal case of zero

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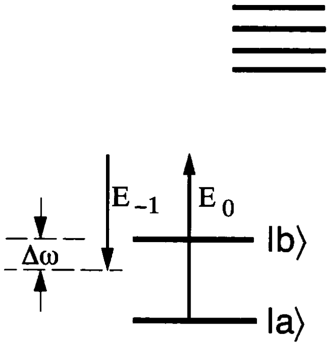

Fig. 1. Energy-level schematic for establishing coherence ρ_{ab} in a molecular system. Laser fields are applied at the frequencies of the $q = 0$ and $q = -1$ sidebands.

linewidth for the $|a\rangle - |b\rangle$ transition. The constants a_q, d_q, and b_q determine the dispersion and coupling and are

$$a_q = \frac{1}{2\hbar^2}\sum_i\left[\frac{|\mu_{ai}|^2}{(\omega_i - \omega_a) - \omega_q} + \frac{|\mu_{ai}|^2}{(\omega_i - \omega_a) + \omega_q}\right],$$

$$b_q = \frac{1}{2\hbar^2}\sum_i\left[\frac{\mu_{ai}\mu_{bi}^*}{(\omega_i - \omega_a) - \omega_q} + \frac{\mu_{ai}\mu_{bi}^*}{(\omega_i - \omega_b) + \omega_q}\right],$$

$$d_q = \frac{1}{2\hbar^2}\sum_i\left[\frac{|\mu_{bi}|^2}{(\omega_i - \omega_b) - \omega_q} + \frac{|\mu_{bi}|^2}{(\omega_i - \omega_b) + \omega_q}\right]. \tag{2}$$

We assume that all the molecular population is initially in the ground state and, as the laser fields are applied, is adiabatically prepared in one eigenstate. We define $B = |B|\exp(j\varphi)$ and $\tan\theta = |B|/[\Delta\omega - (D/2 - A/2)]$; this eigenstate and its coherence are

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

$$\rho_{ab} = 1/2 \sin\theta\exp(j\varphi), \tag{3}$$

where positive θ corresponds to the phased state and negative θ corresponds to the antiphased state of the molecular system. Adiabatic preparation of a highly coherent medium, with $|\rho_{ab}| \approx 0.5$, is central here. It ties this study to that of Jain *et al.* on nonlinear optical generation at maximal coherence[11] and also has a relation to electromagnetically induced transparency.[12]

The slowly varying envelope propagation equation for the qth sideband in local time is

$$\frac{\partial E_q}{\partial z} = -j\eta\hbar\omega_q N(a_q\rho_{aa}E_q + d_q\rho_{bb}E_q + b_q\rho_{ab}E_{q-1} + b_{q+1}^*\rho_{ab}^*E_{q+1}), \tag{4}$$

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

We now proceed to the numerical solution for H_2. We show concurrent Raman generation and pulse compression. We assume monochromatic fields E_0 and E_{-1} to be applied at $z = 0$, with power densities of 10^{10} W/cm^2, and retain sidebands from $q = -80$ to $q = 160$. We obtain the solution of Eq. (4) by forward stepping from $z = 0$ with the density matrix elements recalculated at each step.

We consider the fundamental rotational transition in H_2 with $\omega_b - \omega_a = 354\,\text{cm}^{-1}$ and take all molecules in the $J = 0$ ground state. The constants a_q, b_q, and d_q include the contributions of all allowed rotation–vibrational transitions in Lyman and Werner bands. Transition frequencies are obtained from Herzberg.[13] The applied laser frequencies are $\omega_{-1} = 25\,200\,\text{cm}^{-1}$ and $\omega_0 = 25\,554\,\text{cm}^{-1}$ (frequency-doubled Ti:sapphire); the two-photon detuning is $\Delta\omega = -0.5$ GHz.

Reduced matrix elements of the dipole moment operator P are related to vibrational band Einstein-A coefficients:

$$|\langle J'', v''|\,|P|\,|J', v'\rangle|^2 = \frac{3}{8\pi^2}\epsilon_0\hbar\lambda_i{}^3 H_{J''J'}A_{v'v''}, \tag{5}$$

where λ_i is the transition wavelength. The Honl–London factors $H_{J''J'}$ for P, Q, and R branches are J'', 0, and $J'' + 1$ for the Lyman band and $J'' - 1$, $2J'' + 1$, and $J'' + 2$ for the Werner band. The coefficients $A_{v'v''}$ for H_2 are obtained from Allison and Dalgarno.[14] We calculate the matrix elements μ_{ai} and μ_{bi} by multiplying $\langle J'', v''|\,|P|\,|J', v'\rangle$ by appropriate $3j$ symbols (we assume linearly polarized fields).

As shown in Eqs. (2), dispersion constants a_q and d_q consist of sums over positive terms proportional to $|\mu_{ai}|^2$ and $|\mu_{bi}|^2$. However, coupling constants b_q involve terms proportional to cross products $\mu_{ai}\mu_{bi}^*$,

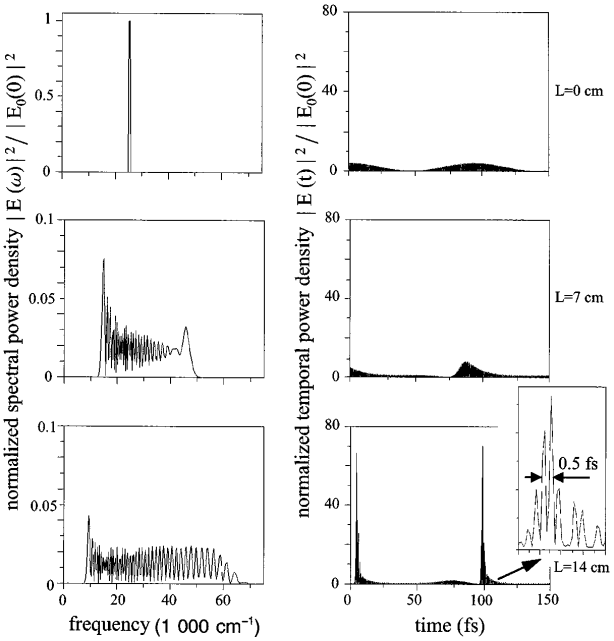

Fig. 2. Evolution of the ultrashort pulses in the time domain (right) and in the frequency domain (left) through the 14-cm-long H_2 cell at 1-atm pressure. The applied laser power densities are 10^{10} W/cm^2, such that $\rho_{ab} = -0.45$. Sidebands powers are normalized to those of the incident fields.

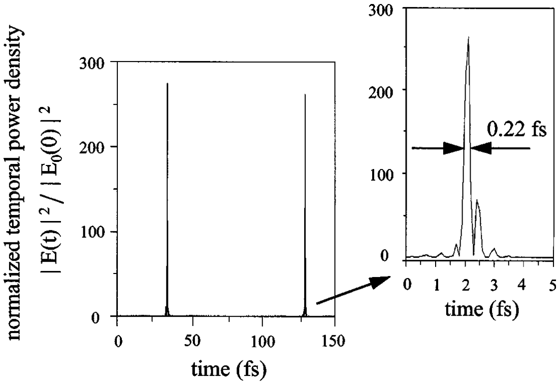

Fig. 3. Instantaneous power density of a signal generated in a 14-cm-long H_2 cell at 1 atm and $\rho_{ab} = -0.45$, with sideband phases corrected by an external spectral-phase filter.

which can be positive or negative and can partially cancel each other. This partial cancellation and reduction of Raman polarizabilities takes place if $|a\rangle$ and $|b\rangle$ belong to different vibrational levels[15] but does not occur in the case of rotational Raman scattering, allowing for higher coupling constants.

Figure 2 shows the calculated spectral and temporal profiles of the generated waveform at different distances in the H_2 cell at 1-atm pressure. At $z = 14$ cm there are nearly 180 sidebands generated that cover $70\,000\ \mathrm{cm}^{-1}$ of the spectrum and Fourier transform into a train of pulses with 0.5-fs pulse width and 94-fs pulse spacing and with the peak intensity exceeding the average intensity by a factor of 35. The coherence $\rho_{ab} = -0.45$ is maintained roughly the same through the medium.

The pulse compression of Fig. 2 is not perfect. We note that the phases of the sidebands are well determined, though not equal, and suggest that they can be compensated for almost perfectly by use, for instance, of spectral filtering with a phase mask.[16] Figure 3 shows the temporal waveform of a signal with the spectral power densities of Fig. 2 and phases perfectly corrected by an external spectral-phase filter. We predict half-cycle pulses (duration 0.22 fs) with a peak intensity exceeding the total input intensity by a factor of 140.

We have examined the sensitivity of this technique to fluctuations of applied intensities, cell pressure, and length. Varying the input intensities by a few percent, or varying the H_2 pressure by a few Torr, changes the output phases insignificantly and does not affect the temporal waveform.

The principal approximations of the numerical results given here are the neglect of the dephasing and the assumptions that the driving fields are both quasi-monochromatic and of infinite extent in the transverse direction and that all molecules are initially in the ground rotational state. For the pulsed excitation we expect these results to apply for a time approximately equal to the dephasing time and therefore expect nearly all the incident power to be converted into the train of subfemtosecond pulses. The thermal population of states other than $J = 0$ will effectively reduce the molecular coherence. For example, in parahydrogen at room temperature, 44% of the molecules will contribute to the coherence ρ_{ab}, and a cell of approximately twice the length compared with the ideal low-temperature case will be needed for broad spectrum generation.

In summary, the use of rotational versus vibrational H_2 states provides approximately ten times as many sidebands and a much larger pulse spacing/pulse width ratio and seems to be the preferred first experiment.

This research was supported by the U.S. Army Research Office, the U.S. Air Force Office of Scientific Research, and the U.S. Office of Naval Research. A. Sokolov's e-mail address is sokol@leland.stanford.edu.

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