PHYSICAL REVIEW A, VOLUME 65, 021801(R)

Raman technique for single-cycle pulses

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(Received 5 June 2001; published 10 January 2002)

We show that two Raman generators which have different Raman transition frequencies and are used in series result in a multiplicative increase in the total number of generated sidebands. When phase-corrected, these sidebands synthesize to a train of randomly timed single-cycle pulses. For series generators on the rotational transitions of molecular D_2 ($J=0\rightarrow2$) and H_2 ($J=1\rightarrow3$), each with 40 modes, the average peak power enhancement is 1600 and the average duration between random pulses is 1.74 ps.

DOI: 10.1103/PhysRevA.65.021801 PACS number(s): 42.50.Gy, 32.80.Qk, 42.60.Fc, 42.65.Dr

The recent experiments by Sokolov and colleagues have shown that coherent molecular oscillations may generate a comb of coherent collinear sidebands with 50 000 cm^{-1} of spectral bandwidth [1]. The essence of this technique is the use of a Raman transition with a sufficiently large coherence that the generation length and the phase-slip length are of the same order. The coherence is established by driving the molecular transition with two single-mode laser fields, slightly detuned from the Raman resonance, so as to excite a single molecular eigenstate [2]. Two recent experiments have shown the mutual coherence of the generated sidebands: In the first, three sidebands are selected and their phases adjusted to demonstrate AM or FM light with a modulation frequency of 2994 cm^{-1}. In the second, five sidebands are selected and their phases are adjusted to form a train of femtosecond time-scale pulses [3]. As in traditional mode locking, the synthesis of well-formed subfemtosecond and single-cycle optical pulses will require phase correction of as many modes as can be practically obtained. Present liquid-crystal and acoustic techniques [4] allow in excess of 1000 resolvable pixels and it is desirable to have a technique for generating a total number of sidebands which are on this order.

This paper suggests a practical method for accomplishing this generation. A schematic of the technique is shown in Fig. 1. Two Raman generators of the type described in the previous paragraph are placed in series. As an example, the first is an H_2 cell with a $J=1\rightarrow3$ rotational Raman transition $\omega_a=587\ cm^{-1}$, and the second is a D_2 cell with a $J=0\rightarrow2$ rotational transition of $\omega_b=179\ cm^{-1}$. As shown, the cells are driven by three linearly polarized, single-mode-pulsed Ti:sapphire lasers whose frequencies are ω_0, $(\omega_0-\omega_a)$, and $(\omega_0-\omega_b)$. Two of these lasers drive the coherence of the molecular hydrogen which, in turn, modulates each of the incident laser beams. At the output of the first cell the generated spectrum consists of two sets of sidebands which are offset from each other by ω_b (Fig. 1). These sidebands enter the D_2 cell and, acting in unison, drive the $\omega_b=179\ cm^{-1}$ coherence. This coherence imposes additional FM modulation on each of the entering sidebands. The net result is a Fourier series with angular frequencies

$$\omega_{q,r}=\omega_0+q\omega_a+r\omega_b. \tag{1}$$

For each pulse the driving lasers are randomly phased and the spectrum of generated sidebands carries these three random phases. We assume that the set of generated sidebands is incident on a liquid crystal or acoustic phase corrector and that the recombined beam is incident on a target cell (for example, a Xe ionization cell) where the temporal structure and peak power is to be studied. The phase correction of the liquid-crystal array or the acoustic modulator is set by assuming that the driving lasers have fixed (nonrandom) phases. It is also set to compensate for any dispersion between itself and the target cell.

We find and will show numerically that, in the limit of a large number of modes, irrespective of the initial random phases, the Fourier transform of the phase-corrected comb corresponds to a train of optical pulses with a peak power which is enhanced relative to the average driving power by a factor equal to the product of the modes of the independent cells. Individual pulses occur at random times with an average periodicity T. If the Raman transition frequency and number of modes of each (independent) cell are denoted by ω_a and N_a and ω_b and N_b, and if the ratio of ω_a to ω_b is irrational, then the peak power enhancement, average periodicity T, and pulse width of the random pulse train are

$$\text{Average peak power enhancement}=N_aN_b,$$

$$T=(2\pi)\frac{N_aN_b}{N_a\omega_a+N_b\omega_b}, \tag{2}$$

$$\delta T=(2\pi)\frac{1}{N_a\omega_a+N_b\omega_b}.$$

Before proceeding with a detailed discussion of this suggestion, we cite connections to other work. Of particular pertinence is the work of Korn and colleagues, who have impulsively excited a vibrational mode of SF_6 and used the resulting time-varying refractive index to produce a sequence of compressed fs pulses [5]. In 1993, Losev and Lutsenko [6] suggested and demonstrated on-resonance two-color pumping, and Yoshikawa and Imasaka [7] suggested the application of multicomponent Raman spectra to femtosecond time-scale pulses. Rotational Raman spectra extending from the infrared to the ultraviolet regions have been demonstrated [8]. Hakuta and colleagues have used solid hydrogen for collinear Raman sideband generation [9]. Possible application of Raman sidebands to the synthesis of solitons with self-

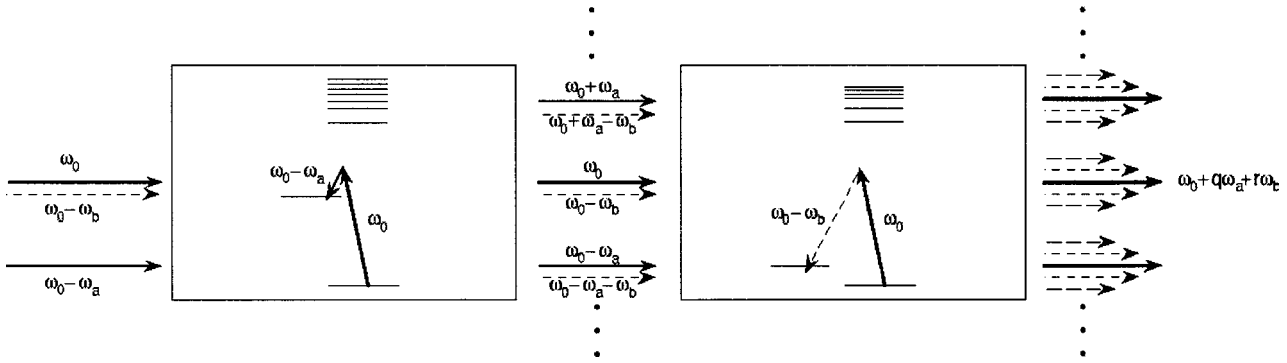

FIG. 1. Schematic of the suggested technique. Three randomly phased laser beams with frequencies denoted by ω_0, $(\omega_0-\omega_a)$, and $(\omega_0-\omega_b)$ drive series Raman generators with incommensurate transition frequencies ω_a and ω_b. The generated spectrum is a Fourier series with terms of the form $\omega_0+q\omega_a+r\omega_b$. With phase correction the series synthesizes to a random train of pulses with a peak power enhancement which is equal to the product of the number of sidebands of the series generators, if alone, and an average pulse periodicity given by Eq. (2).

induced transparency (SIT) or electromagnetically induced transparency (EIT) character has been suggested by Kaplan [10] and by Yavuz *et al.* [11].

We continue with the description and a numerical example. Following Harris and Sokolov [2], we make approximations which lead to an ideal Bessel function solution. These are the neglect of dispersion and the approximation in the coupled propagation equations that all of the sideband frequencies may be replaced by ω_0. Because the essence of the suggested technique is in the behavior of the incommensurate Raman frequencies and random phases, the departure of the numerical (density matrix and slowly varying envelope) solution from the ideal Bessel function solution is not of importance. We also note that it does not matter which cell is placed ahead of the other. By using the Bessel identity $\Sigma_n J_{n+q}(x)J_{n+p}(x)=\delta_{pq}$, it may be shown that the magnitude of each of the Raman coherences depends only on the intensity of the entering laser drivers, i.e., the molecular coherences do not vary with distance in either of the cells. In fact, by direct solution of the appropriate two-species propagation equation, it may be shown that the two molecular species may be mixed in any proportion in a single cell. For all of these cases, the generated spectrum is a Fourier series of the form $\Sigma E_{q,r}\exp[j(\omega_0+q\omega_a+r\omega_b)t]$. With $\varphi_0=\arg[E_{0,0}(0)E^*_{-1,0}(0)]$ and $\theta_0=\arg[E_{0,0}(0)E^*_{0,-1}(0)]$, the coefficients of the two-frequency series are

$$\begin{aligned}E_{q,r}(\gamma_a l_a,\gamma_b l_b)=&E_{0,0}(0)\exp[j(\varphi_0-\pi/2)q+j(\theta_0-\pi/2)r]J_q(\gamma_a l_a)J_r(\gamma_b l_b)\\&+E_{-1,0}(0)\exp[j(\varphi_0-\pi/2)(q+1)+j(\theta_0-\pi/2)r]J_{q+1}(\gamma_a l_a)J_r(\gamma_b l_b)\\&+E_{0,-1}(0)\exp[j(\theta_0-\pi/2)(r+1)+j(\varphi_0-\pi/2)q]J_{r+1}(\gamma_b l_b)J_q(\gamma_a l_a).\end{aligned}\tag{3}$$

With the subscripts a and b denoting the respective cells, the arguments of the Bessel function amplitudes $\gamma_a l_a$ and $\gamma_b l_b$ are

$$\begin{aligned}\gamma_a l_a&=\eta\hbar\omega_0\mathcal{N}_a b_a|B_a|l_a/\sqrt{\delta\omega_a^2+|B_a|^2},\\\gamma_b l_b&=\eta\hbar\omega_0\mathcal{N}_b b_b|B_b|l_b/\sqrt{\delta\omega_b^2+|B_b|^2}.\end{aligned}\tag{4}$$

The Raman Rabi frequencies in each cell are $B_a=b_aE_{0,0}(0)E^*_{-1,0}(0)$ and $B_b=b_bE_{0,0}(0)E^*_{0,-1}(0)$, the quantities $\mathcal{N}_a$ and $\mathcal{N}_b$ are the number of molecules per volume, $\delta\omega_a$ and $\delta\omega_b$ are the two-photon detunings from the respective Raman transitions, and $\eta=\sqrt{\mu/\epsilon_0}$. The Raman coupling coefficients b_a and b_b of the respective molecules with linearly polarized driving lasers and matrix elements μ_{ij} are

$$b=\frac{1}{2\hbar^2}\sum_i\left[\frac{\mu_{gi}\mu^*_{ei}}{(\omega_i-\omega_g)-\omega_0}+\frac{\mu_{gi}\mu^*_{ei}}{(\omega_i-\omega_e)+\omega_0}\right].\tag{5}$$

We assume that the generated frequency-modulated laser beams are incident on a liquid-crystal or acoustic phase corrector which is programmed to maximize the peak intensity in a later target cell. The phase correction for each of the Raman sidebands is set by ignoring the random phases and assuming that the initial phases of each of the three driving lasers are zero. This may be done because the ratio of the Raman transition frequencies is irrational, and as a function of time, there is a sweep-through of all possible phases. The phase correction $C_{q,r}$ at each frequency $\omega_{q,r}$ is then

$$C_{q,r}=\arg[E_{q,r}|_{\varphi_0=\theta_0=0}].\tag{6}$$

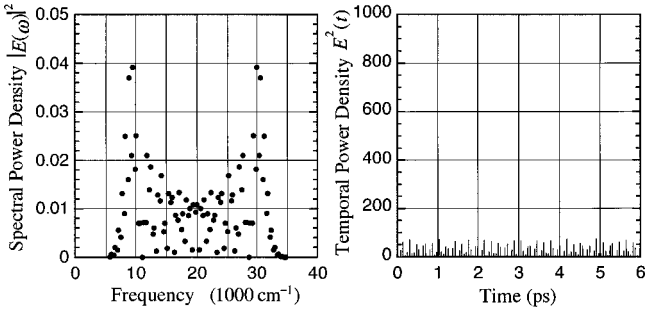

FIG. 2. Power spectral density and time-domain synthesis for an H_2 cell and phase compensator driven by three frequencies ω_0, $\omega_0-\omega_a$, and $\omega_0-\omega_b$. $\omega_0=20\,000\ \mathrm{cm}^{-1}$, $\omega_a=587\ \mathrm{cm}^{-1}$, $\omega_b=179\ \mathrm{cm}^{-1}$, and the modulation depth $\gamma L=20$. The total average driving power is one unit.

We proceed with a numerical example. As per the preceding discussion, the three incident frequencies are ω_0, $\omega_0-\omega_a$, and $\omega_0-\omega_b$. We take $\omega_0=20\,000\ \mathrm{cm}^{-1}$, $\omega_a=587\ \mathrm{cm}^{-1}$, and $\omega_b=179\ \mathrm{cm}^{-1}$ so as to correspond to the transitions of H_2 and D_2, respectively. We assume molecular densities and drive strengths so that the modulation depth $\gamma L=20$ in both cells. We thus expect two sets of about $2\gamma L=40$ sidebands of substantial intensity at the end of the first cell and 1600 sidebands at the end of the second.

Figures 2(a) and 2(b) show the power spectral density and time-domain synthesis if only the first cell and phase compensator are present. Figures 3(a) and 3(b) show the power spectral density and time-domain synthesis if only the second cell and phase compensator are present. Figures 4(a) and 4(b) show these same quantities with both cells in series.

The first cell, if alone, produces a spectrum with a width of about 23 480 cm^{-1} ($\approx 40\times 587\ \mathrm{cm}^{-1}$). In the time domain, following phase compensation, this spectrum synthesizes to a train of pulses with an average periodicity of 56.8 fs and an approximate peak to average power enhancement of 40. The second cell, if alone, produces a spectrum with a width of about 7160 cm^{-1} ($\approx 40\times 179\ \mathrm{cm}^{-1}$). In the time domain, following phase compensation, this spectrum synthesizes to a train of pulses with a periodicity of 186.2 fs and a peak to average power enhancement which is approximately equal to that of the first cell.

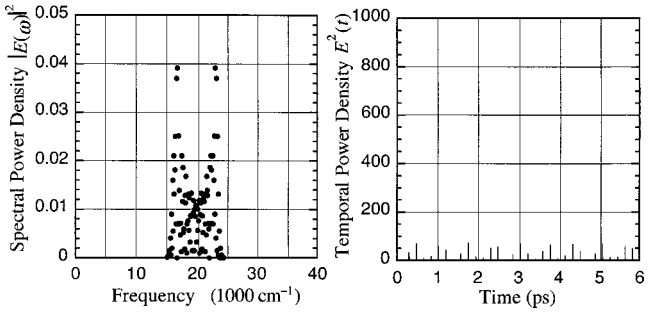

FIG. 3. Power spectral density and time-domain synthesis for a D_2 cell and phase compensator driven by three frequencies ω_0, $\omega_0-\omega_a$, and $\omega_0-\omega_b$. As in Fig. 1, $\omega_0=20\,000\ \mathrm{cm}^{-1}$, $\omega_a=587\ \mathrm{cm}^{-1}$, $\omega_b=179\ \mathrm{cm}^{-1}$, and the modulation depth $\gamma L=20$. The total average driving power is one unit.

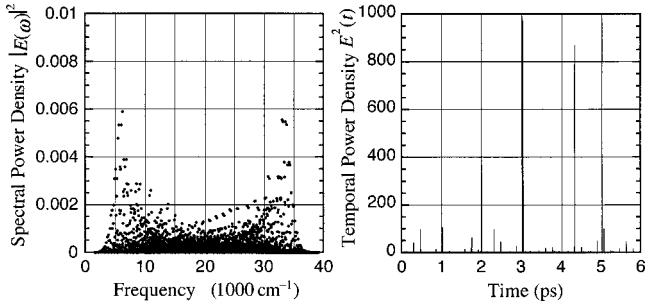

FIG. 4. Power spectral density and time-domain synthesis for the H_2 and D_2 cells in series. The parameters are the same as those of Figs. 2 and 3.

Both cells in series produce a spectral width of 30 640 cm^{-1} [$\approx 40\times(587+179)\ \mathrm{cm}^{-1}$]. In the time domain, the pulse structure is randomly timed with an approximate periodicity of 1.74 ps. Here, the peak to average power enhancement is approximately 1600 and is approximately equal to the product of the number of sidebands of the respective cells [Eq. (2)].

Figure 5 shows the instantaneous electric field for a cosine and a sine pulse for series cells as in Fig. 4. Because of the large number of sidebands which are generated with the cells in series, we typically find well-separated single-cycle pulses which have a peak power enhancement within 50% of N_aN_b.

In summary, we have described a technique for obtaining a multiplicative increase in the number of generated Raman sidebands and, therefore, in the peak power and structure of a generated single-cycle waveform. Some extensions seem likely: As noted earlier, the two Raman species, for example D_2 and H_2, may be mixed in the same cell. Also, the technique may employ an arbitrary (technology-limited) number of molecular species, with one additional laser needed for each species. For example, three mixed Raman media which are driven by four randomly phased lasers will produce the cube of the number of sidebands of each medium, if alone. We have recently experimentally demonstrated an off-resonance, linearly polarized, Raman process on the $\nu'=0$, $J'=3\rightarrow\nu''=0$, $J''=1$ rotational transition of H_2 at 587 cm^{-1} [12]. We generate 37 coherent sidebands with a spectral bandwidth of over 20 000 cm^{-1}. Experiments with parameters much like those described here should soon be possible.

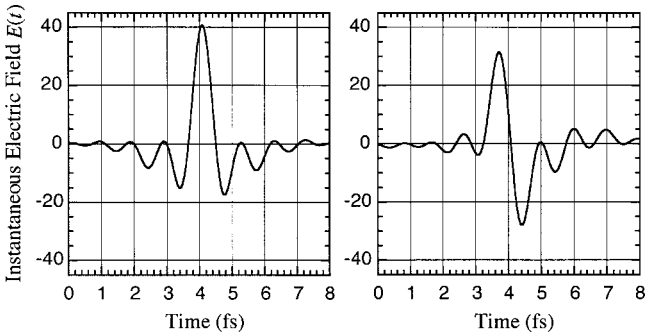

FIG. 5. Instantaneous electric field strength for cosine and sine pulses for the conditions of Fig. 4.

It also seems likely that this multiplicative generation technique should be applicable to the impulsive excitation method of Korn and colleagues [5].

One application of high-power ($>10^{15}$ W/cm^2) single-cycle optical pulses is to high-order harmonic generation. An electron moving in the field of a single-cycle pulse moves away from, and then returns to, the nucleus only once [13]. Christov *et al.* have shown that, as the driving pulse length is shortened, the high harmonic periodic spectrum merges to form a near-continuous spectrum which in the time domain corresponds to a pulse of radiation whose width is well into the attosecond range [14].

The authors gratefully acknowledge helpful discussions with Marty Fejer, Scott Sharpe, Alexei Sokolov, and Rick Trebino. This work was supported by the U.S. Office of Naval Research, the U.S. Air Force Office of Scientific Research, and the U.S. Army Research Office. D.R.W. acknowledges support from the Fannie and John Hertz Foundation.

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