Sub-Single-Cycle Optical Pulse Train with Constant Carrier Envelope Phase

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We describe the synthesis of periodic waveforms consisting of a train of pulses that are 0.83 cycles long and have an electric field pulse width of 0.44 fs using 7 Raman sidebands generated by molecular modulation in H_2. We verify by optical correlation that the carrier-envelope phase is constant in these waveforms when they are synthesized from commensurate sidebands. The estimated overall shift of the carrier-envelope phase is less than 0.18 cycles from the first to the last pulse of nearly 10^6 pulses in the pulse train.

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The overall route to the synthesis of a periodic train of arbitrarily shaped optical pulses, and, in particular, single-cycle optical pulses requires that the frequency ω_n of the nth term of the Fourier series of every pulse train be an integer multiple of the frequency difference ω_0 of every pair of adjacent terms in the Fourier series, $\omega_n = n\omega_0$, $n = 1, 2 \ldots$. When this is the case, the carrier-envelope phase (CEP) is fixed, where the CEP is the phase difference between the peak of the envelope of a pulse and the closest peak of the carrier wave on which it is superimposed. In recent years, the technique of molecular modulation pioneered by Harris and his co-workers has been used by several groups to generate a comb of optical frequencies with a bandwidth spanning as much as four octaves [1–7]. By taking a subset of the frequencies in this comb the synthesis of a train of several- to single-cycle pulses has been reported [6,8,9]. However, in these prior works the peak of the carrier wave in the pulses periodically sweeps through all values enclosed by the pulse envelope.

It is the intent of this Letter to advance the molecular modulation technique for optical waveform synthesis in two ways. First, by using optical correlation, we demonstrate the key role that commensurate frequencies play in achieving a constant CEP, and second, by increasing the bandwidth of the frequency comb we show periodic waveforms that consist of pulses that are 0.83 cycles long and have an electric field width of 0.44 fs. The estimated slip in the CEP is less than 0.18 cycles from the first to the last pulse of nearly 10^6 pulses in a pulse train. We believe that extension of this technique now under way in our laboratory will allow the synthesis of arbitrary optical waveforms much like those obtained with present day rf synthesizers.

In molecular modulation, a Raman transition is driven *adiabatically* by two high-intensity lasers so that the molecular coherence approaches its maximum value of 0.5. At this point the driving-laser frequencies are modulated to produce many sidebands that propagate collinearly with the driving lasers. The modulation frequency is equal to the difference of the frequencies of the two driving lasers. We have previously reported generation of sidebands by molecular modulation in room-temperature H_2. By appropriately detuning the incident lasers from the Raman resonance and applying sufficiently high intensities we overcame the large Doppler broadening in H_2 to create a large coherence. This then led to the production of sidebands with wavelengths that range from 2216 nm in the infrared to 133 nm in the vacuum ultraviolet. The frequencies of these sidebands span nearly 4 octaves and a total of more than 70 600 cm^{-1} [7]. Such a spread in frequency is more than sufficient for use to synthesize a train of single-cycle subfemtosecond pulses.

We describe here a procedure to generate sidebands that form a commensurate spectrum and the construction of ultrashort pulses with a constant CEP. The sideband-generation setup is similar to that employed in Ref. [7]. Our driving lasers are two near-transform-limited lasers, one is a dye laser system [10] tuned to 602 nm and the other is a Ti:sapphire laser system [11] tuned to 802 nm. The two laser frequencies are exact multiples of their difference in frequency. A calibrated wave meter (HighFinesse model WS-Ultimate-UV, absolute accuracy ~30 MHz) is used to determine the laser wavelengths. The energy and pulse width of the 602 nm beam are 6.7 mJ and 3 ns. For the 802 nm beam these quantities are 7.3 mJ and 3.2 ns. The two beams are combined by a dichroic mirror and focused into a 50 cm long H_2 cell filled to 500 Torr at 298 °C. The focal spot diameter of the 602 and 802 nm beams are 160 and 170 μm, respectively, corresponding to intensities of 11.1 and 10.1 GW/cm^2. The difference of the two laser frequencies is tuned to 1.5 GHz below the H_2 $Q(1)$ Raman

resonance of 4155.25 cm^{-1} and the 602 nm pulse is delayed by 0.5 ns relative to the 802 nm pulse.

Under these conditions, a set of more than ten commensurate sidebands is generated from the cell. The frequencies of the sidebands constitute the terms of a Fourier series and the temporal waveforms produced by them will be periodic. We use the approach of nonresonant four-wave difference-frequency mixing ($\omega_l = \omega_i + \omega_j - \omega_k$) described in Ref. [9] to verify this. The experimental setup is similar to that shown in Fig. 1 of that reference. A subset of 6 sidebands, $q = -2$ to 3 (q is the order of the Raman sideband) with their corresponding wavelengths of 1203 ($q = -2$), 802, 602, 481, 401, and 344 nm, respectively, are collimated by a uv achromat. To avoid damage to the optical components, the copropagating beams are attenuated by a mirror (15% T at 602 nm and 802 nm and >70% T at other wavelengths). The sidebands are dispersed by a prism pair and passed through a phase modulator. The phase modulator (Jenoptik SLM-S 640) is a panel consisting of a linear array of 640 liquid-crystal pixels, each 10 mm high and 97 μm wide. Its working wavelength range is 430–1500 nm. This range allows us to adjust the phase of four ($q = -2$ to 1) sidebands. All 6 sidebands are recombined and focused to an intensity of 16 GW/cm^2 inside a cell containing 100 Torr Xe. We monitor the difference-mixing signal at 301 nm (equivalent to $q = 4$) using a solar-blind photomultiplier.

Here we explain the phase adjustment procedure. With commensurate sidebands the electric field of the pulse train associated with the sidebands can be written in the form $E(t) = \sum_n E_n(t) = \sum_n A_n(t)\cos(\omega_n t + \phi_n)$, where $A_n(t)$ is an envelope function, $\omega_n = n\omega_m$, ω_m is the modulation frequency, and ϕ_n is the phase of the nth sideband. It follows from Fourier transform theory that the narrowest envelope is achieved when all the term are in phase, i.e., ϕ_n is a constant. In fact as long as ϕ_n satisfies the linear relation $\phi_n = \phi_{CEP} + n\phi_m$, where ϕ_{CEP} and ϕ_m are independent of n, the fields can be rewritten as $E(t) = \sum_n A_n(t)\cos[n\omega_m(t + \phi_m/\omega_m) + \phi_{CEP}]$ and the narrowest envelope will be preserved. Here it can be immediately recognized that $E(t)$ is periodic, ϕ_{CEP} is the CEP, and ϕ_m leads to a time shift of the electric waveform. For sidebands generated by molecular modulation the linear phase condition is automatically satisfied and ϕ_{CEP} and ϕ_m depend only on the initial phases of the two incident lasers. However, the dispersion of the intervening optics and the Gouy phase shift that results from propagating these sidebands from the H_2 cell to the Xe cell cause shifts to the phases of the sidebands. It is hence necessary to compensate these shifts. Once compensated and if we do a bookkeeping of ϕ_n, it follows that the quantum pathways that lead to the generation of four-wave mixing signals in Xe will add coherently to give the largest four-wave mixing signal when $\phi_n = \phi_{CEP} + n\phi_m$. The sideband phases can thus be conveniently adjusted by maximizing the four-wave mixing signal, at 301 nm in our case. With six sidebands, there are nine quantum pathways that can generate the four-wave mixing signal (e.g., 2×344 nm $-$ 401 nm and 401 nm + 481 nm $-$ 802 nm, etc.). Reference [9] describes a procedure that adjusts the phase of each individual sideband to maximize the mixing signal, optimizing each sideband's phase iteratively until there is no further increase in the signal. Here we show a procedure where the phase adjustment can be completed in just 4 steps for 6 sidebands.

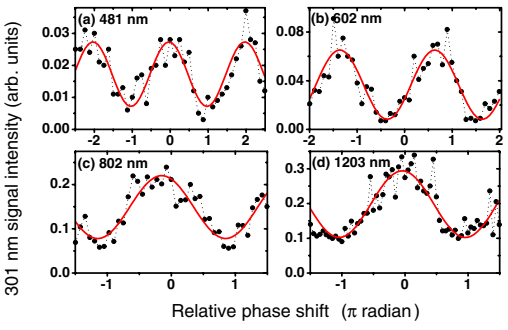

FIG. 1 (color online). Four-wave mixing signal at 301 nm in Xe as a function of the adjustment of the phase of a single sideband in progressive steps at the wavelengths of (a) 481 nm, 301 nm being generated with the sidebands $q = 1$, 2, 3; (b) 602 nm, after adding $q = 0$ to case (a); (c) 802 nm, after adding $q = -1$ to (b); and (d) 1203 nm, with all 6 sidebands included in the generation. Points: experimental data, Lines: simulation.

We start with the three highest order sidebands ($q = 1$, 2, 3 or $n = 5$, 6, 7) as input to the Xe cell. Since ϕ_m only needs to be independent of n, we let ϕ_m be equal to $\phi_7 - \phi_6$, which is kept unchanged throughout the entire experiment. With three sidebands there are two possible pathways to generate the next higher sideband at 301 nm ($2\omega_7 - \omega_6$ and $\omega_7 + \omega_6 - \omega_5$). Their relative phases are $2\phi_7 - \phi_6$ and $\phi_7 + \phi_6 - \phi_5$. Interference between these two paths results in a sinusoidal oscillation of the 301 nm signal when the phase ϕ_5 of the $q = 1$ (481 nm) sideband is varied, as shown in Fig. 1(a). This oscillation reaches its maximum whenever the phases $(2\phi_7 - \phi_6)$ and $(\phi_7 + \phi_6 - \phi_5)$ are equal (modulo 2π). This lets us set ϕ_5 to a value such that $(\phi_6 - \phi_5) = (\phi_7 - \phi_6) = \phi_m$. We next add the $q = 0$ sideband. A similar analysis shows that the 301 nm signal is maximized when ϕ_4 satisfies $(\phi_5 - \phi_4) = \phi_m$. The procedure is repeated in sequence until all 6 sidebands are included to generate the 301 nm signal as shown in Figs. 1(b)–1(d). For 6 sidebands, this phase adjustment is complete after 4 successive steps. In general, the phase adjustment takes $n - 2$ steps to complete for n sidebands. At this point, the 301 nm signal is at a global maximum. The phases of the 6 sidebands satisfy the condition $\phi_n = \phi_{CEP} + n\phi_m$. Fourier synthesis of these sidebands then results in a periodic pulse train that has a CEP of ϕ_{CEP} which is constant throughout the entire pulse train.

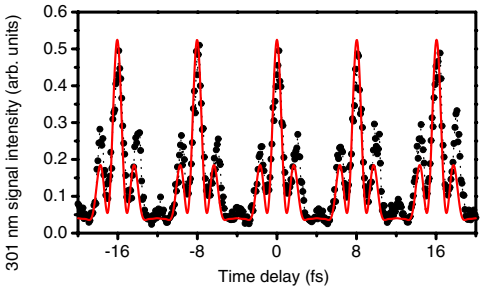

FIG. 2 (color online). Cross-correlation signal (points) at 301 nm as a function of the relative time delay between one pulse consisting of sidebands of wavelength and the corresponding pulse energy (μJ in brackets) of 1203 nm (34), 802 nm (117), 602 nm (207) with respect to a second pulse made of 481 nm (160), 401 nm (23), and 344 nm (7), respectively. The predicted signal (line) is calculated with the pulse energies shown here in brackets and normalized to the experimental value at zero time delay.

Physically ϕ_{CEP} is related to the phase difference between the two driving-laser pulses. In this experiment the driving lasers operate independently so that ϕ_{CEP} can fluctuate from zero to 2π from one ns pulse to another. Note that this phase adjustment procedure works also for incommensurate sidebands that have a finite carrier-envelope offset frequency ω_{CEO} by letting $\omega_n = \omega_{\mathrm{CEO}} + n\omega_m$ in the above analysis. However the CEP now has a time varying slip of $\omega_{\mathrm{CEO}}t$.

We next perform a cross-correlation experiment to confirm that the waveform is periodic. This is done by taking the waveform created by the three lowest-frequency sidebands ($q = -2, -1$, and 0) and electronically delaying it with respect to the waveform created by the three highest-frequency sidebands ($q = 1$–3) while recording the mixing signal at 301 nm. We employ the model described in Ref. [9] to calculate the cross-correlation signal, using the measured sideband amplitudes as input to predict the signal amplitude vs the delay time. Figure 2 shows that good agreement is obtained between prediction and measurements. This good agreement verifies that a periodic train of single-cycle pulses spaced 8 fs apart (equaling the time inverse of the sideband frequency spacing) and with a constant CEP has been successfully synthesized.

For comparison, we measure the cross-correlation signal from a set of sidebands consisting of incommensurate frequencies. For this measurement we tune the driving lasers to 589 and 780 nm, respectively. There is a new four-wave mixing signal wavelength of 298 nm. Now the carrier-offset frequency ω_{CEO} is changed from zero to 349.27 cm^{-1}. This is about 1/12 of 4155.2 cm^{-1}. Theory predicts then that the waveform will only repeat itself every $\sim$96 fs ($\sim 12 \times 8$ fs). Indeed the data (Fig. 3) agrees with this prediction. Theory further predicts a mirror image centered at zero time delay. We used this to establish the zero-time reference of the data in Fig. 3.

The waveform synthesized from 6 sidebands consists of periodic pulses whose envelope width is 1.6 fs. We proceed next to obtain shorter pulses by involving more sidebands. This is accomplished by using a homemade phase modulator composed of a row of five 14 mm high by 4 mm wide by 0.022 mm thick liquid-crystal panels. The size and location of each panel is designed to match the sideband beam size and to allow unimpeded passage of five sidebands. The liquid-crystal material is E7 from Merck & Co., Inc., and has a uv cutoff at 330 nm. With this new modulator one additional sideband, at 401 nm, can be controlled. Hence a total of 7 sidebands, the 6 used above ($q = -2$ to $q = 3$) plus $q = 4$ at 301 nm, can now be employed in the waveform synthesis. The four-wave mixing signal is at 267 nm. The total bandwidth is now 24 931.2 cm^{-1} which is equal to 2 octaves. This will give rise to an electric field cycle width of 0.83 cycles [12]. The phases of sidebands $q = -2$ to 2 are adjusted sequentially as described earlier to maximize the 267 nm signal. For cross correlation we

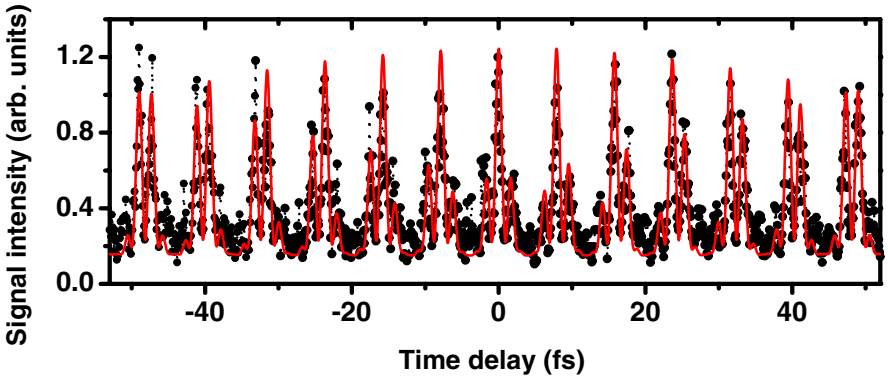

FIG. 3 (color online). Cross-correlation signal (points) produced by two subsets of incommensurate sidebands at 298 nm obtained and plotted as a function of relative pulse delay in a manner similar to that in Fig. 2. Here one pulse has sidebands with wavelength and corresponding pulse energy (μJ in brackets) of 1155 nm (16), 780 nm (188), 589 nm (240), and the second pulse has 473 nm (197), 396 nm (34), and 340 nm (3.7), respectively. Solid line is from calculation. Zero time delay is set to the center of the symmetric signal waveform.

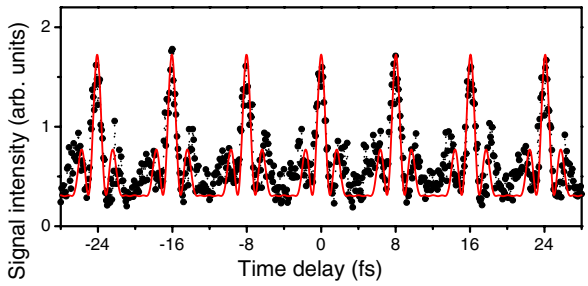

FIG. 4 (color online). Cross-correlation signal (points) at 267 nm as a function of relative time delay between one pulse consisting of the sidebands of wavelengths and corresponding pulse energies (μJ in brackets) of 1203 nm (26), 802 nm (148), 602 nm (144), with respect to another consisting of 481 nm (113), 401 nm (33), 344 nm (11), and 301 nm (2.3), respectively. The solid line is the calculated trace using pulse energies shown in brackets.

delay the waveform consisting of the three lowest-frequency sidebands with respect to the four highest-frequency sidebands. The resulting cross-correlation signal is shown in Fig. 4. The good match between theory and cross-correlation data in the figure serves as verification that with 7 sidebands we have produced a periodic train of 0.83-cycle pulses. From the measured sideband amplitudes we calculate that the pulse envelope FWHM is 1.4 fs. When $\phi_{\mathrm{CEP}} = 0$, this constitutes a subcycle cosine pulse that has an E field width of 0.44 fs. This width can increase by up to $\sim$10% for other values of ϕ_{CEP}. The maximum pulse peak power is approximately 0.9 MW.

The absolute frequency of the driving lasers is accurate to $\sim$30 MHz. This uncertainty leads to a maximum carrier-envelope offset frequency ω_{CEO} of 60 MHz. From this we estimate that the CEP slip between adjacent pulses in the waveform is at most 2.4×10^{-7} cycles. For a pulse train that lasts for 6 ns (covering pulses with more than 10% of the full power) there are about 7.5×10^5 pulses. Consequently the maximum change in the CEP from the first single-cycle pulse to the last pulse in the train is less than 0.18 cycles. This value can be improved to the sub-mrads level by stabilizing the laser frequencies to the Hz level with the help of a frequency comb [13]. Fluctuations in the signal caused by beam pointing jitter and thermal drifts give an estimated 0.1π phase jitter over a period of 30 min. Such fluctuations can be reduced by improving the mechanical and thermal stability of the optical components.

In these experiments we did not control or measure the CEP. However, the CEP of each ns pulse *relative* to the next can be determined by measuring the interference signal between the second harmonic of one of the driving lasers and the sideband that has the same wavelength [14]. It is then possible to apply these pulses to study phase-dependent processes by simultaneously measuring the interference signal and then assigning the relative phases to the corresponding data in the manner described in Ref. [15].

By incorporating 11 sidebands, the present scheme will provide a periodic train of subcycle pulses with <1 fs envelope width and a field width of about 0.3 fs. Furthermore, with the flexibility to adjust the phase of every frequency component, the sidebands can readily be used to synthesize arbitrary optical waveforms [16]. A basic component to forming nonsinusoidal waveforms is the first term in the Fourier series. With molecular modulation using vibrational H_2 this term is at 2406.6 nm. For this reason we are constructing a powerful source at 2406.6 nm to serve as the building block for this application.

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