Measurement of Fourier-synthesized optical waveforms

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Following the experiments of Shverdin and colleagues [Phys. Rev. Lett. **94**, 033904 (2005)], we describe a technique for determining the temporal envelope of an optical beam whose spectrum consists of n discrete, equally spaced frequency components. Four-wave mixing is employed to generate $n-1$ higher-frequency sidebands. The relative intensities of these sidebands, together with the intensities of the incident sidebands, determine the unknown relative phases of the incident beam. © 2005 Optical Society of America

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Over the past several years it has been shown that molecular modulation can produce a collinear beam of mutually coherent sidebands that extend in frequency from the IR to the far UV.[1–4] Adjusting the phases of these sidebands allows periodic optical waveforms with a prescribed temporal shape to be synthesized. In early experiments the temporal shape of the synthesized waveforms was measured by using multiphoton ionization.[5]

Recently, Shverdin *et al.*[6] used a new technique for waveform characterization based on four-wave mixing of the type $\omega_i+\omega_j-\omega_k$. They described how n equally spaced Raman sidebands incident on a Xe cell will generate $n-1$ higher-frequency (UV) sidebands that extend the incident comb of frequencies. Adjusting the relative phases of the incident sidebands and thereby shaping the incident temporal waveform dramatically altered the relative intensities of the UV sidebands. A train of single-cycle pulses maximized the UV intensity, FM-like waveforms minimized the intensity, and other waveforms produced particular relative intensity distributions.[6]

In this Letter we describe the inverse process: the use of the generated $n-1$ component UV spectrum to determine the unknown relative phases of the primary spectrum. With the primary sidebands' amplitudes known, the incident temporal waveform is determined to within the carrier-envelope phase. This method is suited for multioctave waveforms with discrete sidebands, a regime not easily accessible with conventional methods such as frequency-resolved optical gating.[7]

Figure 1 depicts the experimental setup for the technique. The generated Raman sidebands are dispersed by a prism, individually phase corrected, and refocused into the target Xe cell. A photomultiplier tube measures the power of each of the generated UV sidebands. Because it allows focusing to the cell center, we use the nonlinear difference-frequency process $\omega_i+\omega_j-\omega_k$. (The alternative, a sum-frequency process of the type $\omega_i+\omega_j+\omega_k$, will have a much lower generation efficiency because of Gouy phase shift.[8])

To start, we assume that the incident Raman sideband frequencies extend to zero so that the frequency of the qth sideband is $\omega_q=q\omega_v$. We also assume that the relative phases of the sidebands are controlled. Figure 2 shows three temporal waveforms, their spectral amplitudes (square root of their powers), and the spectral amplitudes of the generated UV sidebands. As expected, the single-cycle (mode-locked) waveform generates the highest-amplitude UV spectrum [Fig. 2(g)], while the FM-like spectrum generates the lowest amplitude [Fig. 2(h)]. The generated spectrum of the sawtooth waveform [Fig. 2(i)] is similar to the single-cycle generated spectrum but of lower amplitude and slower dropoff. The technique of this work determines the temporal waveform (left-hand panels) to within a carrier-envelope phase from the power spectra (middle and right-hand panels).

Experiments that use the Raman technique to produce a comb of sidebands have, to date, used two independent driving lasers whose absolute frequencies are not a multiple of their frequency difference.[1,2,4] As a result, the spectrum of n sidebands has two independent phases. Also, because the lowest frequency of the comb is offset from zero, the sidebands do not constitute a strict Fourier series, and the waveform is not periodic. In this situation we control $n-2$ relative phases and synthesize the envelope of the temporal waveform. The carrier-envelope phase now varies linearly with time, causing the waveform to drift slowly under the envelope.[9]

For a wide-bandwidth waveform, changes in carrier-envelope phase may drastically alter its shape. Illustrating this, Fig. 3(a) depicts a Fourier-synthesized sawtooth waveform and its envelope;

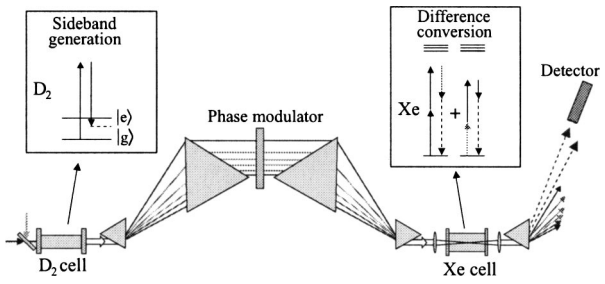

Fig. 1. Experimental setup for synthesis and characterization of a set of sidebands. Through molecular modulation, optical sidebands are produced in a D_2 cell. The sidebands are dispersed through a prism, individually phase modulated, recombined, and focused into a Xe cell. A detector measures the power spectral density of the sidebands generated through difference-frequency conversion.

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Figs. 3(b)–3(d) depict qualitatively dissimilar waveforms that differ only in carrier-envelope phase.

We describe two techniques that allow the determination of the temporal envelope, given the known powers of the n Raman sidebands and the $n-1$ generated UV sidebands. The first technique is suggested by noting that we may generate $n-2$ polynomial equations for the $n-2$ unknown relative phases. We consider the difference-frequency process $\omega_q=\omega_i+\omega_j-\omega_k$ and assume that all the generated sidebands are sufficiently far from resonance that the third-order susceptibility is independent of frequency. We also ignore all propagation effects including dispersion, which can be made negligible by either reducing the gas pressure or switching to a less dispersive medium. With the electric field of the jth sideband written as $(1/2)E_j\exp[\imath(\omega_j t+\phi_j)]+\text{c.c.}$, the power $P(\omega_q)$ generated in sideband q by sidebands 1 through n is

$$P(\omega_q)=K\left|\sum_{i=q-n+1}^{n}\sum_{j=q-i+1}^{n}E_iE_jE_k^*\exp[i(\phi_i+\phi_j-\phi_k)]\right|^2, \tag{1}$$

where $k=i+j-q$, $q>n$, and K is an unknown proportionality constant. The summation in Eq. (1) results in only one term for the highest sideband, $q=2n-1$. The power of this sideband is therefore independent of the input phases, and we may use it to solve for the unknown proportionality constant K.

We next set the two lowest phases to zero with the following linear transformations: adding a common phase to each sideband (a carrier-envelope phase shift) and adding a phase proportional to each sideband frequency (a time origin shift). Both of these transformations leave $P(\omega_q)$ unchanged; thus neither the time origin nor the carrier-envelope phase can be determined by our technique.

We let $E_q\exp[\imath\phi_q]=z_q$, transforming all equations to polynomials in the unknown complex variables z_q. For example, for four input sidebands, the powers of the two phase-dependent, generated sidebands are

$$P(\omega_5)=K|z_3^2z_1^*+2z_4z_2z_1^*+2z_4z_3z_2^*+z_4^2z_3^*|^2,$$

$$P(\omega_6)=K|2z_4z_3z_1^*+z_4^2z_2^*|^2,$$

where $z_q^*=E_q^2/z_q$. We numerically solve these polynomials for all sets of solutions and retain only those solutions corresponding to measured magnitudes, i.e., $|z_q|=|E_q|$. We are left with $2^{(n-2)}$ sets of solutions.

A second experiment is necessary to select the correct solution from the previous set of solutions. This is easily done: if all the Raman sidebands except for the lowest three are blocked, then the difference-frequency process generates two UV sidebands. Solving the equations associated with these sidebands gives two solutions for z_3. By cross referencing these two values for z_3 with the original $2^{(n-2)}$ solutions for z_3, we pick the pair of values for $\{z_3,\ldots,z_n\}$ that solves all equations. The waveforms represented by the two solutions are time-reversed versions of each other. The analytic procedure described here is incapable of differentiating between time-reversed solutions.

The procedure as described above may turn out to be impractical. By using Mathematica's NSolve function we are unable to obtain solutions for $n>6$ sidebands. In this case, it may be possible to use adaptive numerical techniques similar to those used in frequency-resolved optical gating to solve the polynomial equations.

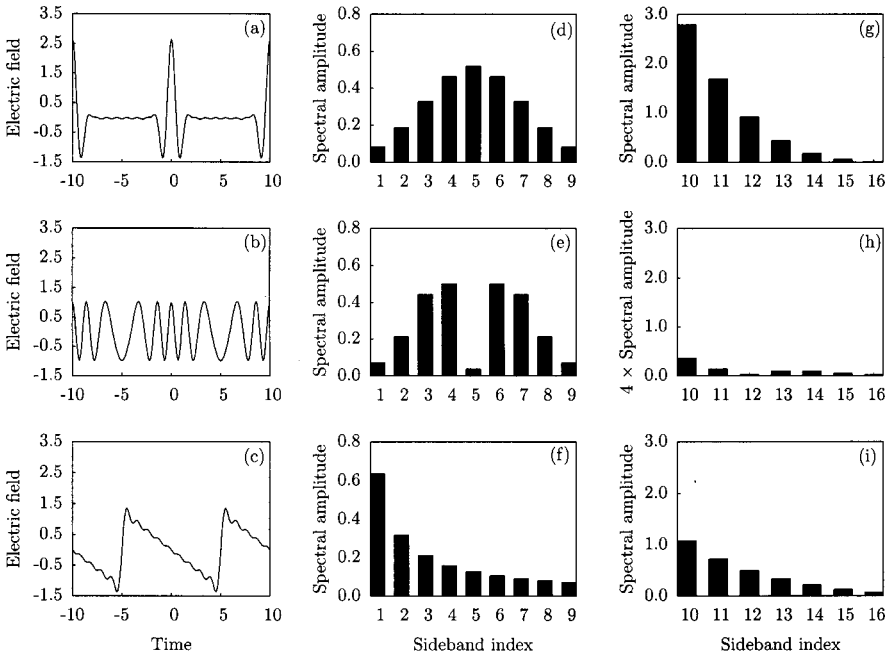

Fig. 2. Three waveforms of equal power are depicted (left-hand panels) along with the spectral amplitudes (middle panels) and the spectral amplitudes generated by difference frequency conversion (right-hand panels). From top to bottom, the waveforms are (a) single-cycle, (b) FM, and (c) sawtooth waveforms. The waveforms consist of nine Fourier sidebands spanning 3.2 octaves. Amplitude comparisons between rows are meaningful, but comparisons between columns are not.

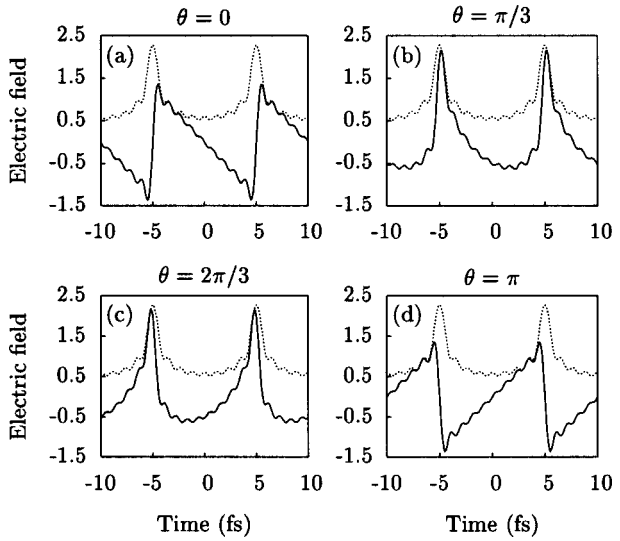

Fig. 3. (a) Nine-Fourier-component sawtooth waveform (solid) and its envelope amplitude (dotted). The waveform is significantly altered as the carrier-envelope phase is changed by (b) $\pi/3$ rad, (c) $2\pi/3$ rad, and (d) π rad while the envelope amplitude remains unchanged.

We next describe a related iterative technique that works for an arbitrary number of sidebands. We set ϕ_1 and ϕ_2 to zero as before and block all but the lowest three sidebands. The equations for generated sidebands $q=4$ and $q=5$ uniquely determine the single unknown, ϕ_3 (and its time-reversed value). Next, the fourth Raman sideband is uncovered. Plugging one of the values for ϕ_3 into the equations for generated sidebands $q=5$ and $q=6$ uniquely determines the single unknown, ϕ_4. In this manner, by unblocking successive Raman sidebands, we may iteratively solve for the phases of an arbitrary number of sidebands.

A third characterization technique substitutes a sum-frequency generation process for the difference-frequency process. Experimental implementation would require enhanced conversion efficiency (with, for example, the use of gas jets). This process generates more sidebands, resulting in an overdetermined set of polynomial equations. Therefore a single measurement is sufficient to uniquely identify the phases. Using this method, we have obtained solutions for as many as $n=8$ sidebands.

In summary, this Letter describes several techniques for characterizing the complex envelope of multioctave temporal waveforms consisting of discrete, equally spaced sidebands.

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