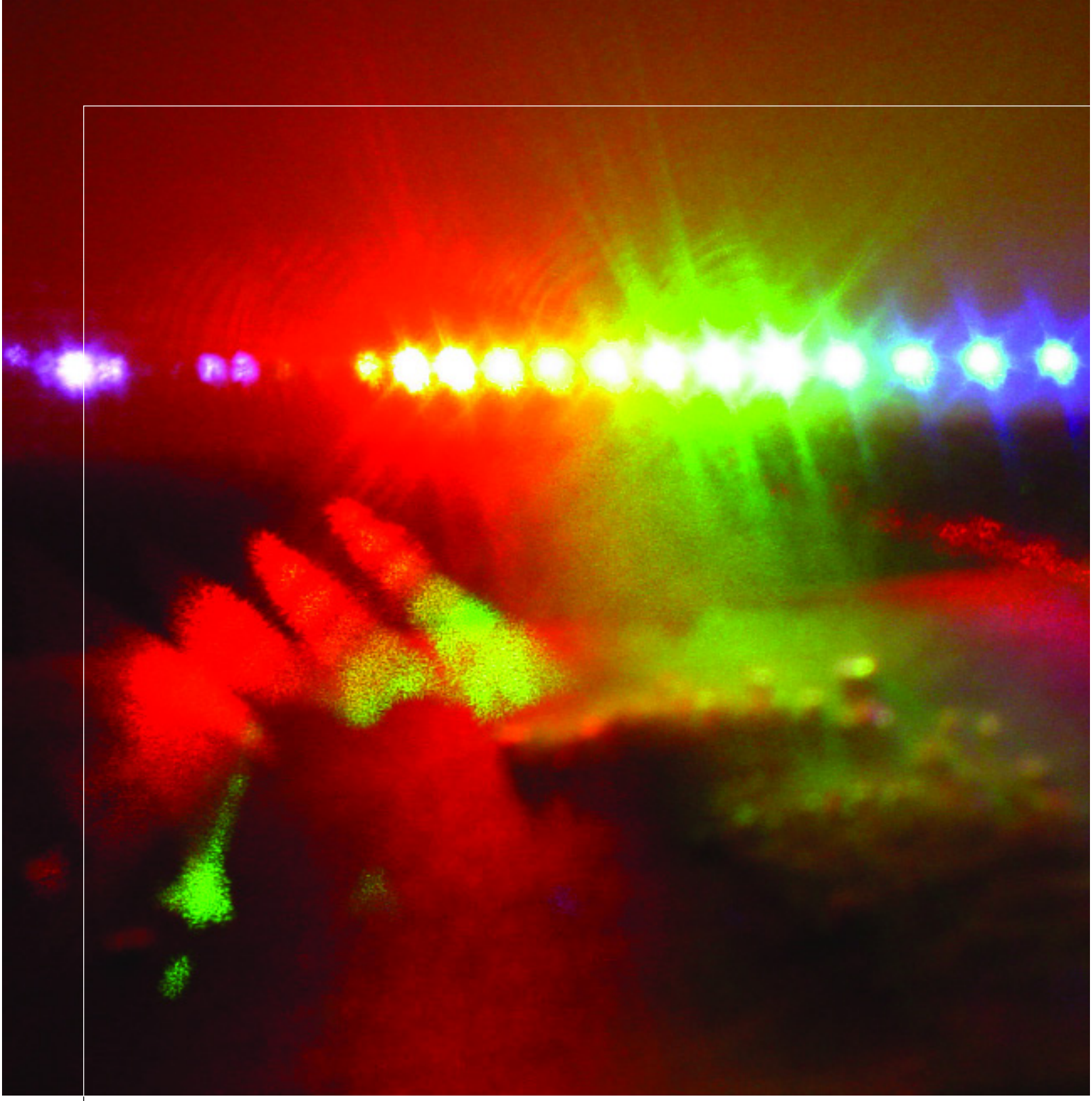

A Quasiperiodic Approach to Ultrashort Pulses

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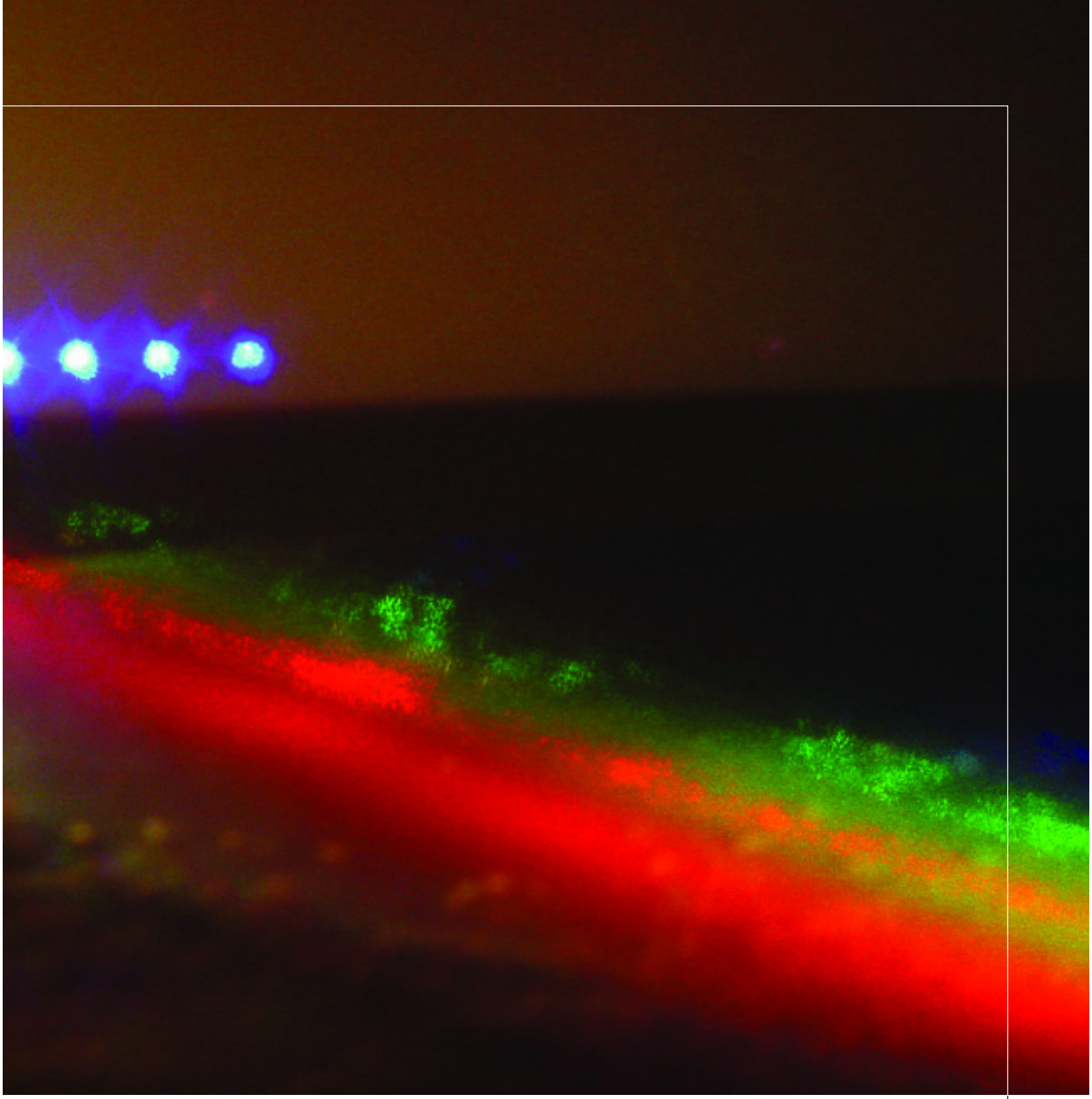
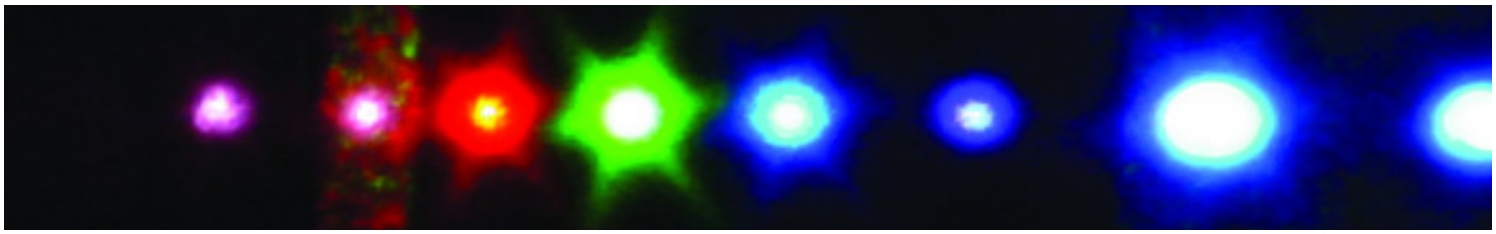


To optically control atomic and molecular processes, the optical waveform must be shaped with as much precision as possible. Precise waveform shaping requires maximization of the number of phase-adjustable frequencies. In this article the authors describe a general technique to multiplicatively increase the number of generated frequencies by use of modulators in series.

A quasiperiodic spectrum of collinear Raman sidebands is dispersed and projected on a screen. The beams are scattered as they pass through liquid nitrogen vapor.



Many methods of generating broad spectra and short pulses rely on modulation of a beam of laser light. In communications, modulation is a means of imprinting information on a transmission medium. In a certain sense, our objective is the same because we ultimately hope to transmit as much "information" as possible—in the form of an optical waveform—to receivers like atoms and molecules. In this article we attempt to further understanding of how multiple modulations can best be used to generate arbitrary waveforms with subcycle, subfemtosecond features in the optical part of the spectrum.

In optical communications, information is routinely encoded onto a carrier wave—a monochromatic beam of light—by use of lithium niobate electro-optic modulators. A modulator is simply a nonlinear device that mixes a signal with a carrier wave, producing additional frequency components which are called sidebands. To obtain larger modulation frequencies, it is necessary to start with something that can oscillate at a higher rate. The largest vibrational frequencies are found in the lightest molecules: Diatomic molecules of hydrogen can be driven roughly ten thousand times faster than conventional electro-optic modulators. Two single-mode Q-switched lasers with a frequency difference slightly detuned from a vibrational resonance can drive molecules into a coherent superposition of the ground state and an excited vibrational state such that all the molecules vibrate in unison. As the molecules stretch, each H_2 molecule looks more like two separate hydrogen atoms, so the refractive index increases. As the molecules contract, they look more like single helium atoms and the refractive index decreases. Consequently, the refractive index of the medium is modulated at the drive frequency. Light at frequency f_0 traveling through the medium experiences a time-varying refractive index and becomes frequency modulated, developing sidebands with frequencies $f_q = f_0 + qf_a$, where f_a is the modulation frequency and q is a positive or negative integer. The two lasers that drive the vibration of the molecules are also modulated to produce many sidebands. Application of this method to the fundamental vibrational transition in molecular deuterium yields 17 sidebands that range in wavelength from 3 μm in the near-infrared to 195 nm in the ultraviolet, as shown in Fig. 1.[1] If one considers that this generation can take place at pressures on the order of a tenth of an atmosphere, it is clear that molecules can be remarkably efficient modulators. Under some circumstances, nearly 100% conversion from the driving lasers into the generated sidebands is observed.[2]

Is there a practical means to increase spectral density while maintaining bandwidth?

We have found experimentally that a slight detuning to one side of resonance results in self-focusing and broader sideband generation. A related technique employs a pulse that is shorter than the molecular oscillation to impulsively excite molecules exactly on resonance. Both impulsive excitation and molecular modulation are viable in a variety of media. Experiments have been performed in SF_6, CO_2, hydrogen, deuterium and solid hydrogen.[3-7]

Use of the term "modulation" implicitly suggests that the optical beams generated at different frequencies will be mutually coherent and collinear. It is not at all clear, however, that these properties will carry over to a process in which the bandwidth is so wide: Traditional stimulated Raman scattering processes produce beams that emerge in cones, with each frequency at a different angle. Mode-locking of Raman modes was first suggested by Yoshikawa and Imasaka[3] and by Kaplan[8] ten years ago. It has recently been experimentally confirmed that molecular modulators generate beams that are collinear and mutually coherent over their spatial and temporal profiles.[9] This suggests that arbitrary waveform synthesis with subfemtosecond features can be achieved. Such arbitrary waveform synthesis requires two essential elements: bandwidth and a large number of spectral components.

We use octaves (or in other words the number of doublings of the lowest frequency necessary to get to the highest frequency) as a scale-invariant measure of bandwidth. The importance of bandwidth is illustrated in Fig. 2, which shows phase-locked waveforms synthesized from a source with a bandwidth of a) 0.1 octave, b) 1 octave, c) 3 octaves. The need for a large number of sidebands is immediately clear when one considers the limited capabilities of an octave of bandwidth in which the only frequency components present are those at the far ends. As the number of intermediate frequencies is increased, the time resolution will be fixed by the bandwidth but the period will lengthen, thereby expanding the range of synthesizable waveforms. The more sidebands there are, the higher the peak intensity can be: Modulation of a single frequency to split the same energy over N frequencies and then individually phase shifting them to produce the shortest possible pulse will result in an N-fold increase in peak intensity.

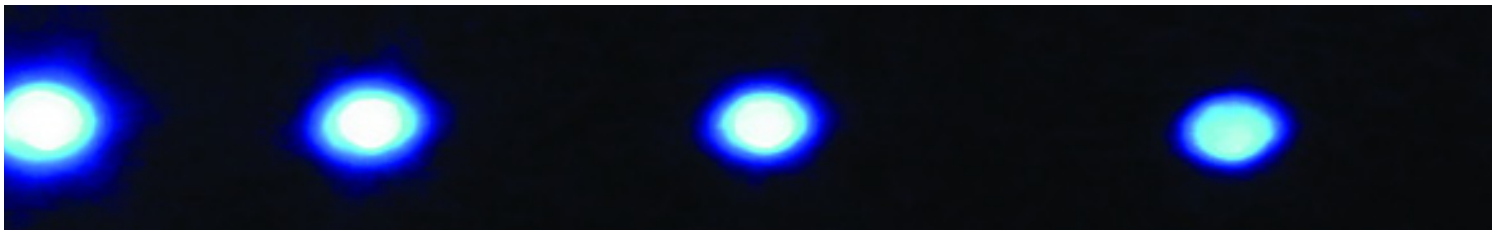


Figure 1. Photograph of the spectrum generated from vibrational modulation of deuterium and imaged by dispersion through a prism. The ultraviolet beams are visible because of the fluorescence of the paper on which they are projected.

While molecular modulation produces several octaves of bandwidth (Fig. 1), the spectral density inherent in this approach is limited. The logical question is then: Is there a practical means to increase spectral density while maintaining bandwidth? A schematic of such a technique[10] is shown in Fig. 3. Two molecular modulators are placed in series. As an example, the first is a cell of deuterium gas with a vibrational Raman transition f_a, and the second is a cell of hydrogen gas with a rotational transition of f_b. As shown, the cells are driven by three single-mode pulsed lasers whose frequencies are f_0, (f_0-f_a) and (f_0-f_b). Two of these lasers drive the vibrational coherence of the molecular deuterium 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 f_b (Fig. 3). These sidebands enter the H_2 cell and, acting together, drive the rotational coherence. This coherence imposes additional frequency modulation on each of the entering sidebands. The net result is a set of sidebands with frequencies

$$f_{q,r}=f_0+qf_a+rf_b. \qquad (1)$$

In the limit of a large number of modes, regardless of the phases of the driving lasers, 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 power of the driving lasers by a factor equal to the product of the modes generated in each cell. If the number of modes of each cell alone is N_a and N_b, and if the ratio f_a to f_b is irrational, then the peak power enhancement is N_aN_b and the pulse width is

$$\delta T=\frac{1}{N_af_a+N_bf_b}.$$

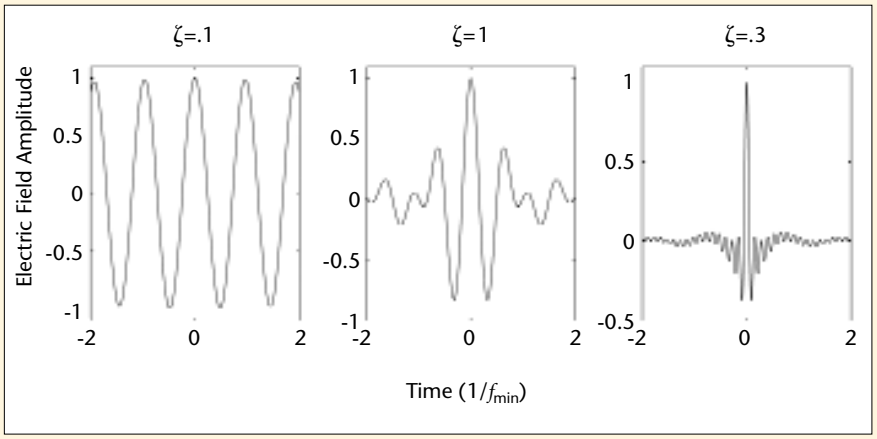


Figure 2. Phase-locked waveforms synthesized from 100 components spanning ζ octaves where $2^{\zeta}=\frac{f_{max}}{f_{min}}$.

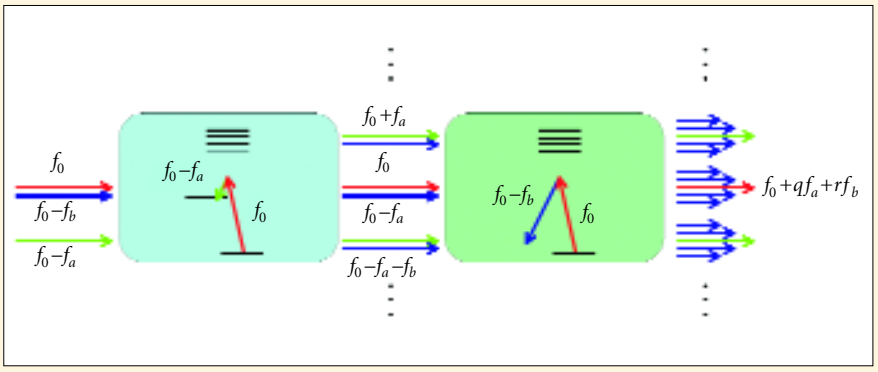


Figure 3. Schematic of the suggested technique. Three randomly phased laser beams with frequencies denoted by f_0, (f_0-f_a) and (f_0-f_b) drive series molecular modulators with incommensurate transition frequencies f_a and f_b. The generated spectrum consists of frequencies of the form $f_0+qf_a+rf_b$. 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 periodicity which also scales as the product of the number of modes.

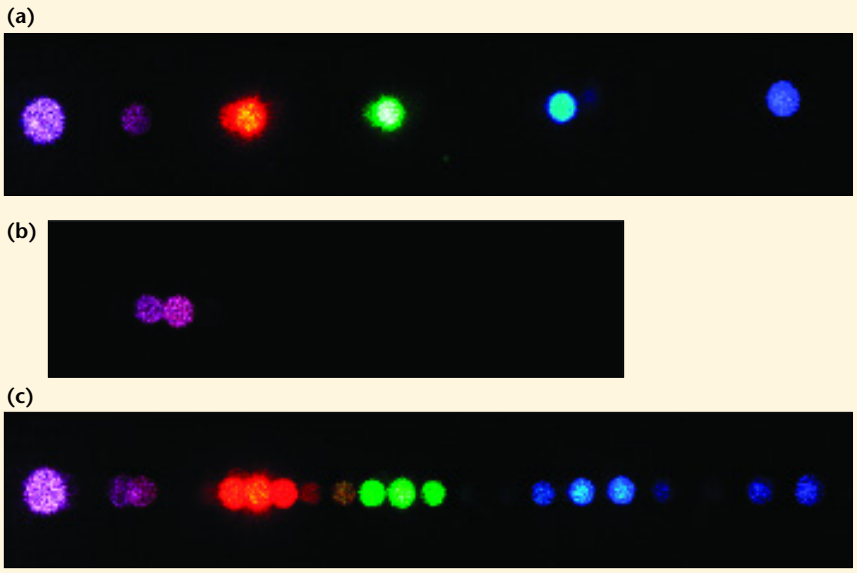


Figure 4. Photographs of (a) part of a vibrational spectrum alone, (b) an intentionally narrowed rotational spectrum and (c) the spectrum produced by driving both oscillations in the same cell.

By allowing the beams to propagate further after the prism, we can zoom in on one of the sidebands to resolve the predicted neighboring rotational sidebands, clearly illustrating the quasiperiodicity of the spectrum.

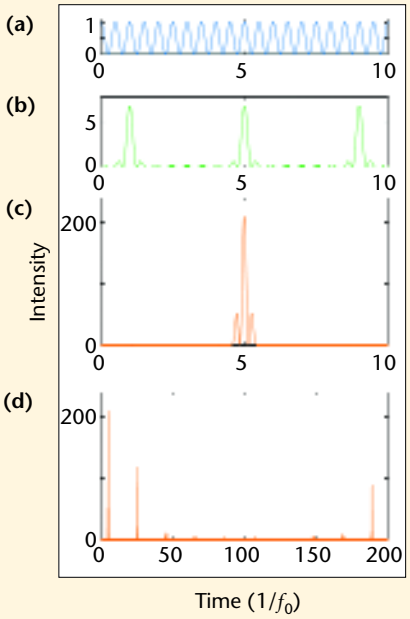


Figure 5. Intensity waveforms from (a) an unmodulated carrier frequency (b) phase locking several equally spaced frequencies, (c) phase locking frequencies generated from two modulations, where the modulation frequencies are incommensurate. Part (d) shows this waveform on a longer time scale, in a way which displays its quasiperiodicity. The time scale is in units of the carrier frequency period.

Individual pulses occur at random times with an average periodicity, which may be shown to be

$$T = \frac{N_a N_b}{N_a f_a + N_b f_b} .$$

These results hold for any modulators that can be used in series.

We note that it does not matter in which order the cells are placed. The magnitude of each of the oscillations depends only on the intensity of the incident driving lasers; i.e., the molecular coherences do not vary with distance in either of the cells. For this reason, the two molecular species may be mixed in any proportion in a single cell. This makes implementing the multiplicative technique particularly easy.

We have demonstrated this technique by exciting oscillations in two gases with two Ti:sapphire lasers and one Nd:YAG laser. The spectrum generated via vibrational modulation of deuterium is shown in Fig. 4(a). When the rotational drive is intentionally weakened by means of detuning from resonance, the hydrogen molecules produce a narrow rotational spectrum consisting of only five sidebands [Fig. 4(b)]. Combination of both excitations yields a spectrum where the rotational modulation of each vibrational sideband can clearly be seen [Fig. 4(c)].

We now examine the importance of the ratio of the two modulation frequencies. Two oscillators at frequencies f and $2f$ have a common period of $1/f$. There is, therefore, a well-defined phase between them. If instead we make the second frequency $2.1f$, the oscillators will no longer match after a time $1/f$. Before they rephase, their phases will "slip" with respect to each other, only rephasing after a time $10/f$. Mutually irrational frequencies have no well-defined phase and can be said to sweep through all possible phases. A waveform composed of such incommensurate frequencies will be quasiperiodic. The frequencies will partially rephase at multiples of the old periodicities and, after an arbitrarily long delay, the waveform will come arbitrarily close to repeating itself (although it will never exactly repeat).

What this means is that the phases of the driving lasers need not be fixed. They may instead be random from shot to shot, and the generation process will be unaffected because the driving pulses are nanoseconds long, so long that millions of laser cycles occur during each one. Any phase-sensitivity that would be seen for very short driving pulses disappears when the pulses are long, eliminating the need to phase-lock the driving lasers.

Figure 5 shows simple numerical illustrations of the basic results of the multiplicative technique. Part a) shows

the intensity waveform of an unmodulated carrier wave with unity amplitude. After modulation of the carrier frequency and phase adjustment of the resulting frequencies to form the shortest possible pulses, the intensity waveform exhibits more intense peaks which appear less frequently [Fig. 5(b)]. A second modulation, with a frequency incommensurate with the first, heightens the effect [Fig. 5(c)]. The time delay was chosen to select a peak in the quasiperiodic waveform. Viewing the same waveform on a time scale twenty times longer [Fig. 5(d)], the quasiperiodic enhancement of a few peaks and the suppression of the rest become evident.

Observe also the role of quasiperiodicity in the frequency domain: If one modulation frequency is an exact multiple of the other, once the width of the second set of sidebands reaches the spacing of the first, sidebands begin to overlap with and interfere with each other, limiting the total number of components. With incommensurate frequencies this is not the case, and production of more sidebands yields ever more independently adjustable frequencies which independently contribute to the waveform.

By restoring the rotational drive to full strength, we obtain a rotational spectrum with a width that far exceeds the vibrational spacing. Figure 6 shows the full power multiplicative spectrum. By allowing the beams to propagate further after the prism, we can zoom in on one of the sidebands to resolve the predicted neighboring rotational sidebands, clearly illustrating the quasiperiodicity of the spectrum.

One advantage to successive modulations is increased stability. Producing 100 sidebands through a single modulation requires 100 steps in the modulation process, each dependent on the previous one, while producing the same number through two different modulations can be done with 10 steps of one and 10 steps of the other. Fewer steps mean greater stability against perturbations. Similarly, multiple modulations can result in a more even energy distribution, with the bigger modulation frequency spreading energy over a large bandwidth and the smaller one locally redistributing it.

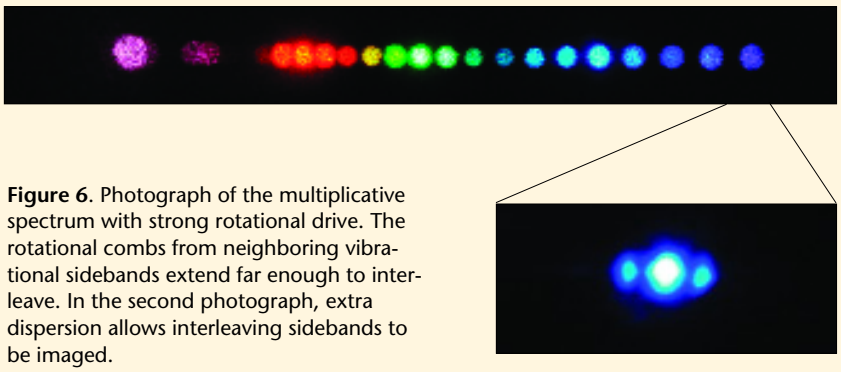


Figure 6. Photograph of the multiplicative spectrum with strong rotational drive. The rotational combs from neighboring vibrational sidebands extend far enough to interleave. In the second photograph, extra dispersion allows interleaving sidebands to be imaged.

Thanks to this improved ability to synthesize arbitrary waveforms, we are approaching the goal of true control over atomic and molecular motion.

Once the frequencies are generated, the laser beams can be sent through a liquid crystal or acoustic phase corrector for pulse shaping. Mode-locking the entire available bandwidth would produce a quasiperiodic train of subcycle subfemtosecond pulses. Thanks to this improved ability to synthesize arbitrary waveforms, we are approaching the goal of true control over atomic and molecular motion. It becomes possible to perform experiments in which the long end of the spectrum drives the nuclei and the short end drives the electrons.

Conclusions

In summary, we have described a technique for obtaining a multiplicative increase in the number of generated sidebands, and therefore, in the peak power 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, in principle, employ an arbitrary 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 driven separately. At present there are several methods of obtaining an optical spectrum with one or more octaves of bandwidth. The multiplicative generation technique should be applicable to several of these techniques, as well as to any set of cascadable modulators.

Acknowledgments

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