Single-Cycle Optical Pulses

Synchronized with Molecular Oscillations

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- This work will show that coherent molecular motion can be used to modulate light, and as a result produce sub-femtosecond and sub-cycle pulses,

- show that this light source can be applied for multiphoton ionization, and the ionization, in turn, can be utilized to characterize the source,

- describe how this new light source can be used to study new physics.

Ultrashort Pulses

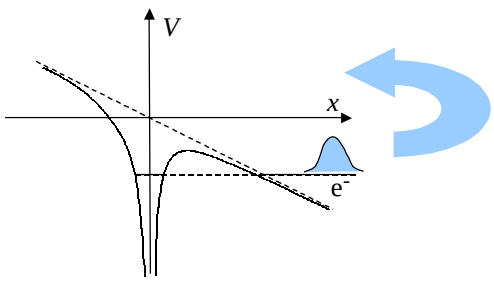
Why?

- observation and control of molecular and atomic dynamics;
- time-resolved photoelectron and photoionization spectroscopy;
- timed Coulomb explosion imaging;
- UV and X-ray amplification and lasing.

Techniques for attosecond pulse generation:

- HHG,
- Raman.

Atom in a strong laser field

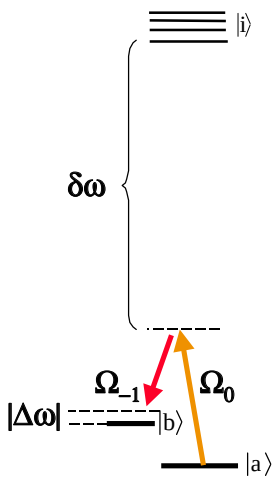


High-order Harmonic Generation
=
tunneling ionization + recollision

Outline:

1. Broadband collinear Raman generator:
   - high coherence, low pressure regime;
   - key idea: adiabatic preparation of a single molecular superposition-state;
   - two mechanisms of molecular modulation.
2. Multiphoton ionization with single-cycle pulses, and waveform characterization by pulse-shape dependent photoionization.
3. Sub-cycle pulse shaping.
4. Synchronization of the pulse train with respect to the molecular motion.

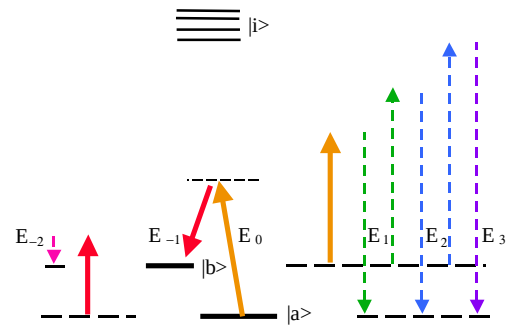
Strongly driven molecular systems



$$\frac{\Omega_0 \Omega_{-1}}{\delta\omega} > |\Delta\omega|$$

$$|\rho_{ab}| = \rho_{aa} = \rho_{bb} = \frac{1}{2}$$

Sideband generation and propagation



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

1) Refractive index control in an EIT-like effect.

2) Broadband spectral generation.

3) Ultrashort pulse generation.

PULSE COMPRESSION BY PARAMETRIC BEATING . . .

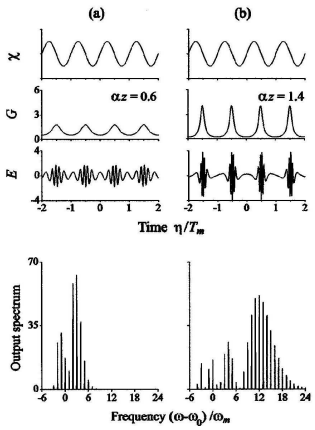


FIG. 2. Compression of a probe pulse, calculated from Eq. (42) for $\alpha z = 0.6$ (a) and 1.4 (b). The carrier frequency, pulse width, and peak time of the input probe field are $\omega_0 = 5.2\omega_m$, $T = 10T_m$, and $\eta_p = 0$, respectively. The plots for the susceptibility χ as a function of time show only the shape but not the exact magnitude of this function. The plots for the output field E and for the spectral intensity $|E_\omega|^2$ are normalized to the peak value of the input field.

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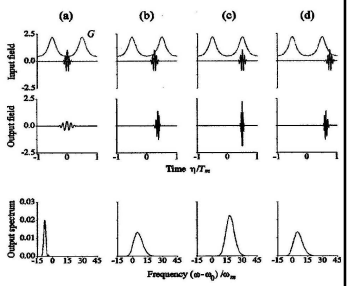
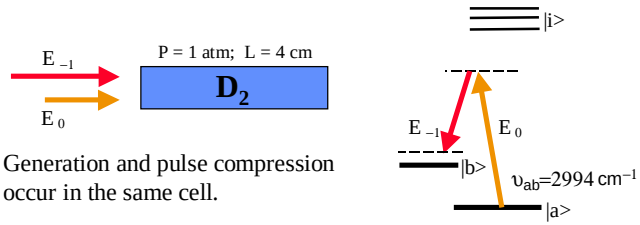
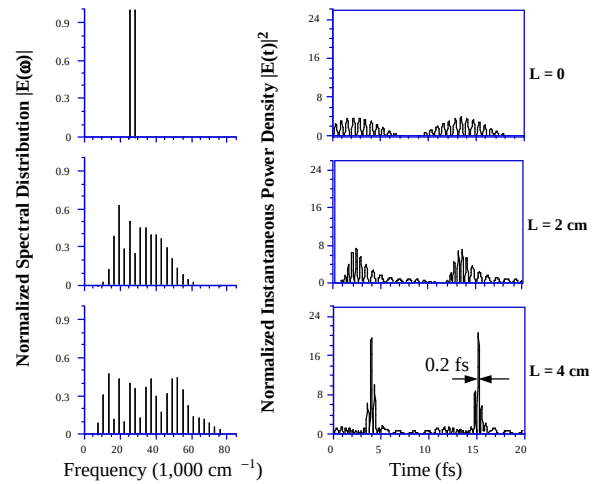


FIG. 3. Compression or stretching of a probe pulse that is short compared to the Raman period. The plots are calculated from Eq. (42) for the parameter $\alpha z = 0.8$. The carrier frequency and pulse width of the input probe field are $\omega_0 = 15.2\omega_m$ and $T = 0.1T_m$, respectively. The injection time is $\eta_p / T_m = 0$ (a), 0.25 (b), 0.5 (c), and 0.75 (d). The plots for the input field, output field, and output spectrum are normalized to the peak value of the input field. The dotted lines show G as a function of time. Note that the number of optical oscillations is the same for all time-domain plots.

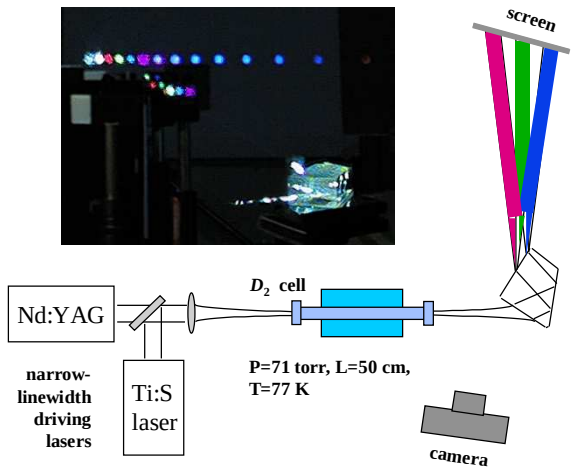
Subfemtosecond pulse generation by molecular vibration in deuterium



Generation and pulse compression occur in the same cell.

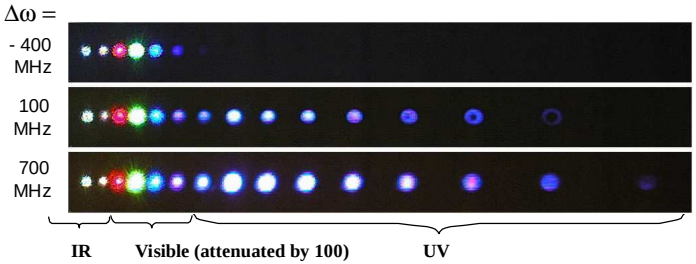


Collinear generation of a wide vibrational Raman spectrum in low-density molecular deuterium (D_2)



We observe:

13 anti-Stokes + 2 Stokes + 2 pump frequencies.



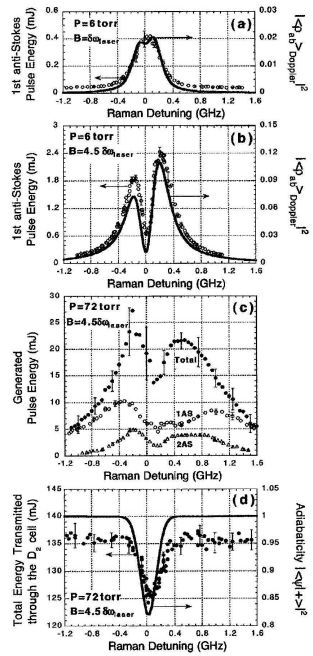
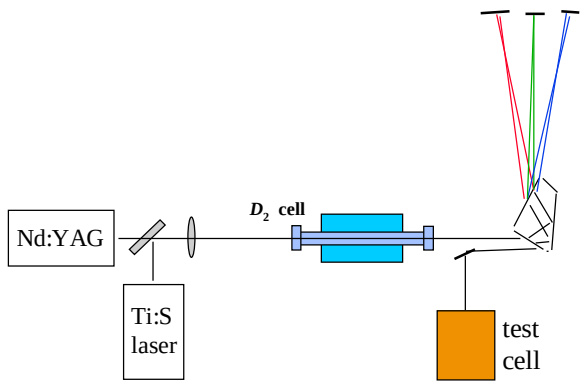


FIG. 4. Raman generation by adiabatic eigenstates in D_2. (a) First anti-Stokes generation by weakly driven vibrations ($B \approx \delta\omega_{\rm laser}$) at $P = 6$ torr. (b) First anti-Stokes generation by strongly driven vibrations ($B \gg \delta\omega_{\rm laser}$) at $P = 6$ torr. In both (a) and (b) the solid lines show the calculated square of the Doppler-averaged coherence ρ_{ab} at the pulse peak. (c) Sum over pulse energies of all generated Raman sidebands (filled circles), first anti-Stokes generation (open circles), and second anti-Stokes generation (triangles) for $B \gg \delta\omega_{\rm laser}$ and $P = 72$ torr. (d) Total energy in all applied and generated sidebands at the output of the D_2 cell. The solid line represents the calculated fraction of the molecular population in the eigenstate which evolves adiabatically from the ground state [Eq. (3)].

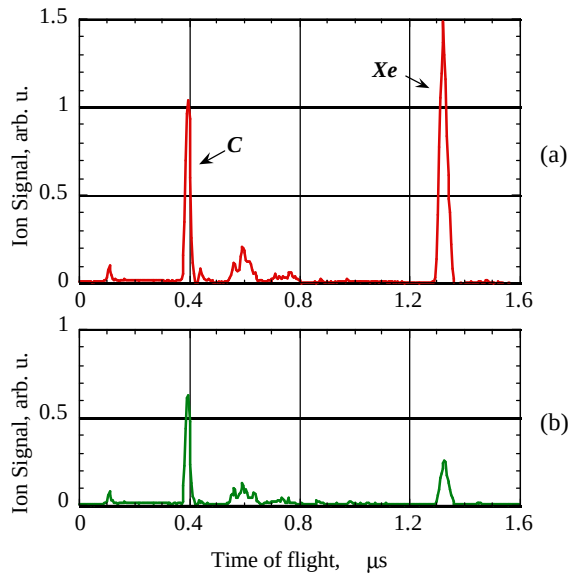
Spectral modification



Coherent Control of Multiphoton Ionization on a Few-Femtosecond Time Scale

- pulse shaping by spectral modification;
- selective ion production;
- number of photons, required for ionization: 11 or 5 ?
- waveform characterization by pulse-shape dependent photoionization.

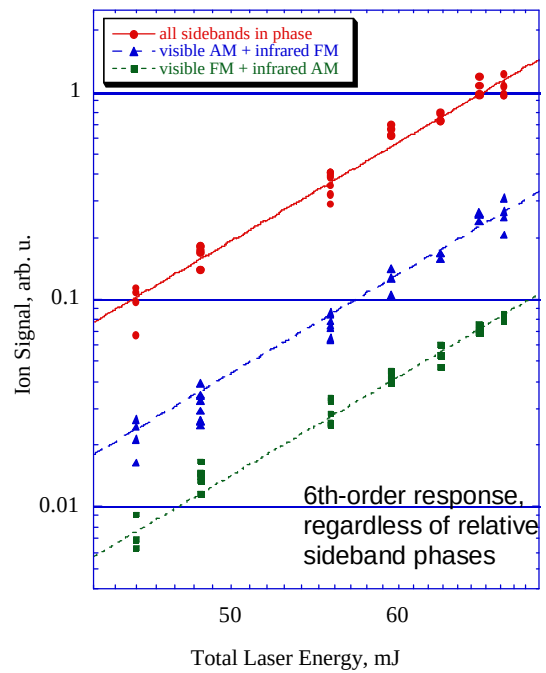
Selective ion production



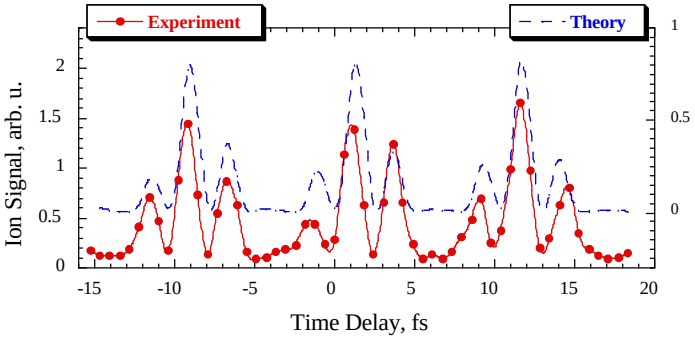
Mass spectrum produced by 5 sidebands phased to
(a) maximize photoionization of Xenon, and
(b) minimize photoionization of Xenon.

Multiphoton ionization of Xenon.

How many photons are required: 11 or 5 ?
Does that number depend on pulse shape?



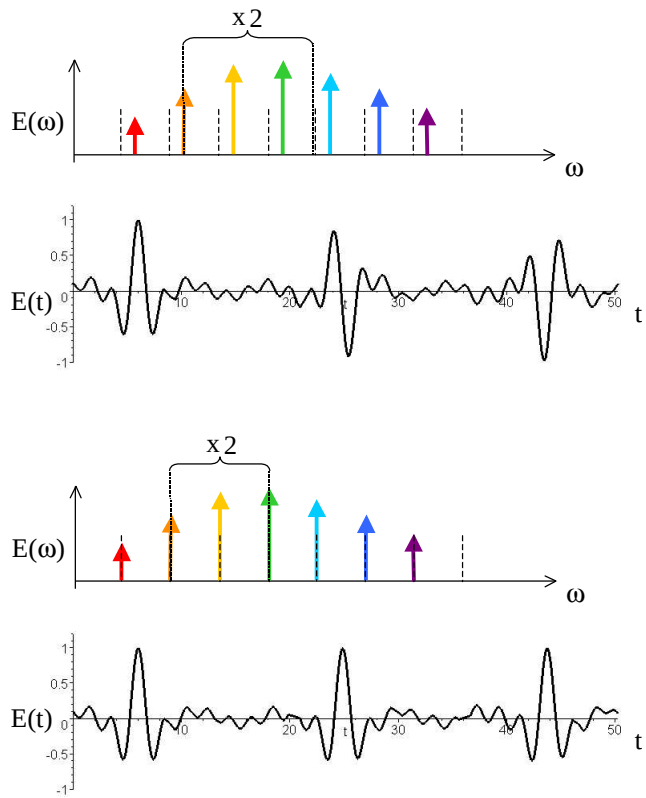
Correlation of waveforms, synthesized
by subsets of Raman sidebands



Simple calculation, assuming
equal sideband amplitudes and a 6th-order detector:

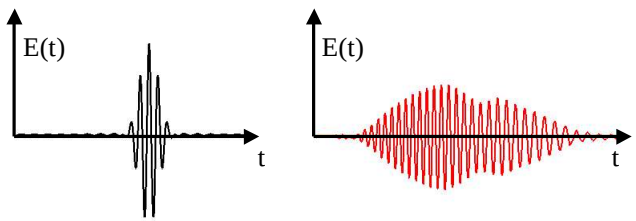
$$\{ \mathrm{Sin}[w_1 t] + \mathrm{Sin}[w_2 t] + \mathrm{Sin}[w_3 t] + \mathrm{Sin}[w_4(t+t)] + \mathrm{Sin}[w_5(t+t)] \}^{12}$$

Sub-cycle phase selection

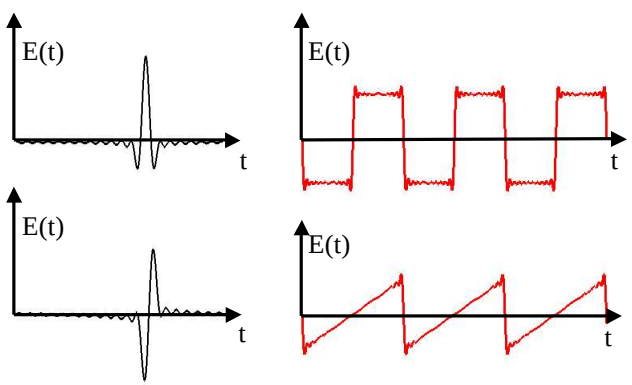


Pulse shaping: “old” and “new”

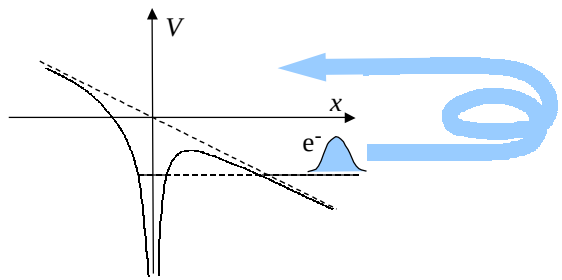
“Traditional” pulse shaping:



Sub-cycle pulse shaping:

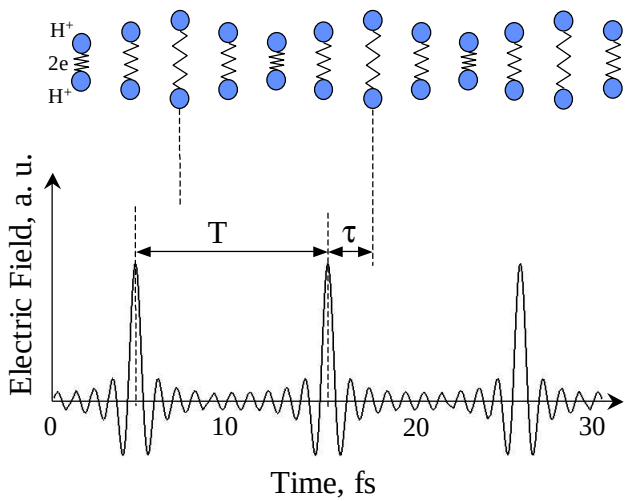


Atom (or molecule) in a strong sub-cycle-shaped laser field.

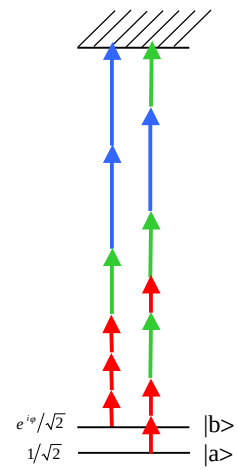


The electron trajectory, the recollision time, and the recollision energy can be controlled precisely.

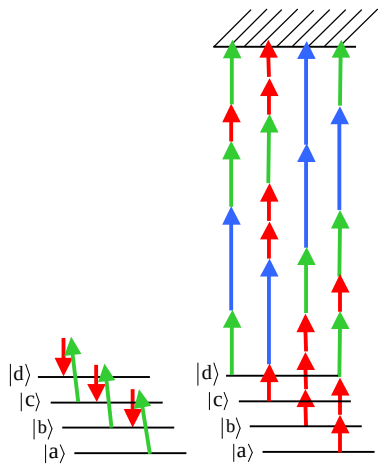
Molecular oscillations modulate light and produce trains of pulses, which are (by the very nature of the modulation process) perfectly synchronized with the molecular motion.



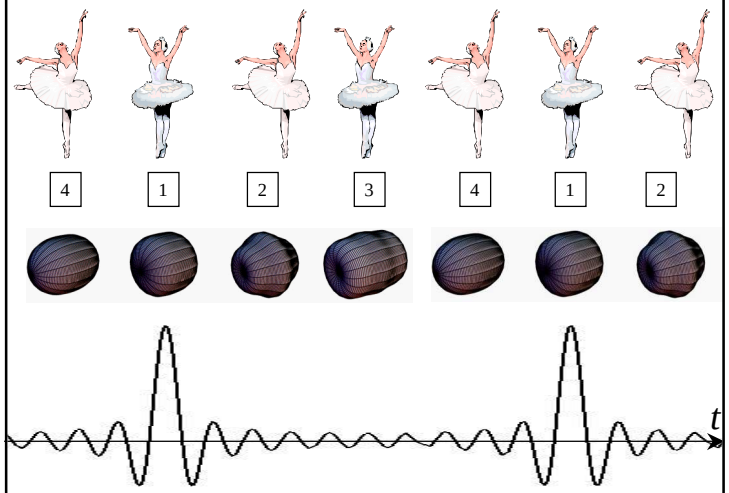
EIT-like control of multiphoton ionization



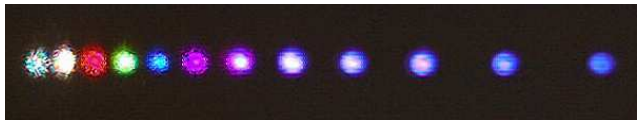
EIT-like control of
multiphoton ionization
with a possibility of a
molecular wavepacket excitation



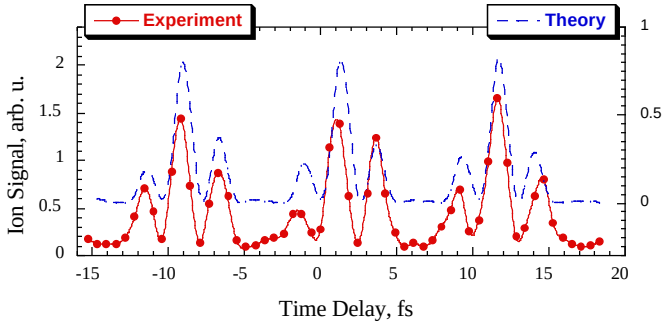
Possible extensions of our technique range from studying complicated multi-mode motion of complex molecules, to probing ultrafast electronic dynamics in atoms.



Extension of EIT ideas to molecular systems allowed us to create a new light source: a collinear Raman generator.



This new light source allows us to study new physics:



We demonstrate photoionization with single-cycle pulses.

Theory: S. E. Harris and A. V. Sokolov, Phys. Rev. A **55**, R4019 (1997); S. E. Harris and A. V. Sokolov, Phys. Rev. Lett. **81**, 2894 (1998); A. V. Sokolov, Opt. Lett. **24**, 1248 (1999).

Experiment: A. V. Sokolov, D. R. Walker, D. D. Yavuz, G. Y. Yin, and S. E. Harris, Phys. Rev. Lett. **85**, 562 (2000); A. V. Sokolov, D. R. Walker, D. D. Yavuz, G.Y. Yin, and S. E. Harris, Phys. Rev. Lett. **87**, 33402 (2001).

Conclusion:

We have demonstrated that coherent molecular motion can modulate light, producing ultrashort pulses which are perfectly synchronized with the molecular oscillation.

This light source opens new horizons for sub-cycle pulse shaping, and for subfemto-second studies of atomic and molecular dynamics.