Generation and Application of Carrier Envelope Phase Controlled Single-Cycle Optical Pulse Train

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Generation of sub-single-cycle pulses in the optical region

- Introduction
- Review of basic concepts
- Modeling molecular modulation
- Present status of experiments
- How to determine pulse duration
- Advance concepts
<table>
<thead>
<tr>
<th>Prefix for small and large numbers:</th>
<th>micro  $10^{-6}$</th>
<th>mega  $10^6$</th>
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<td>giga  $10^9$</td>
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<td>tera  $10^{12}$</td>
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<tr>
<td>yotta  $10^{-24}$</td>
<td>yocto  $10^{24}$</td>
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</tbody>
</table>

**Man made today:**

- **shortest time** -- about 100 as
- **most intense light** -- $10^{23}$ $\text{W/cm}^2$

*Courtesy C. D. Lin, Kansa State U*
Attoworld

Attosecond: $10^{-18}$ second

$C \times 10^{-18}$ sec = 0.3 nm
Time scales

300 nm optical cycle

vibration

dissociation

rotation

fs laser systems

electron dynamics

as soft x-ray pulses

Short pulses can be used to monitor and control molecular and electronic motion
Characteristic length and time scales for structure and dynamics in the microcosm

F. Krausz & M. Ivanov, Rev. Mod. Phys. 81 163 (2009)
Laser pulses got shorter over the years.

Peak intensity increased.

- **Focused intensity, W/cm²**
  - 1970: $10^{10}$
  - 1990: $10^{15}$
  - 2010: $10^{20}$
  - Chirped Pulse Amplification

- **Shortest optical pulse measured, fs**
  - 1960: $10^4$
  - 1970: $10^3$
  - 1980: $10^2$
  - 1990: $10^1$
  - 2000: $10^0$
  - 2010: $10^{-1}$

- **Mode-locked Dye and Ti:Sapphire lasers**
- **HHG pulses**
- **Molecular modulation**

Ultrafast science

High field physics
Correlation between time and frequency

\[ x(t - t_0) \overset{FT}{\leftrightarrow} e^{-j\omega t_0} X(\omega) \]

Fourier transform:

\[ X(\omega) = \int_{-\infty}^{\infty} x(t)e^{-i\omega t} \, d\omega \]
Effect of random phase
Principle of optical interference of coherent light fields

(a) In phase  (b) Random phase
What is a single cycle optical pulse

(a) Monochromatic light: sinusoidal wave propagation
(b) Beating of two waves, $\omega_1$ and $\omega_2$
What is a single cycle optical pulse

(c) Many waves propagating to form a wave packet (left)
(d) Ultimate wavepacket is a single-cycle and sub-cycle pulse pulse (right)
Optical cycle

\[ E(t) = \tilde{E}(t) + c.c. \]
\[ \tilde{E}(t) = A(t)e^{i(\omega_0 t + \phi)} \]

\[ \omega_0 = \frac{\int_0^{\infty} \omega |E(\omega)|^2 d\omega}{\int_0^{\infty} |E(\omega)|^2 d\omega} \]

Carrier frequency

\[ E(\omega) : \text{Fourier transform of } E(t) \]

Single cycle waveforms

Inverted cosine
\[ \phi_n = \pi \]

Cosine pulse
\[ \phi_n = 0 \]

Sine pulse
\[ \phi_n = \pi/2 \]

- Inverted cosine: 780 nm, 12,820 cm\(^{-1}\)
- Cosine pulse: 200 nm, 50,000 cm\(^{-1}\)
- Sine pulse: 684 as, 2.6 fs
Carrier envelope phase

\[ E(t) = E_0(t) \cos(\omega_0 t + \phi) \]
**Constant carrier envelope phase**

\[ E(t) = \sum_{n} E_n(t) \cos(\omega_n t + \phi_n) \quad \Phi_n = \text{carrier envelope phase} \]

incommensurate \quad \omega_n = n \omega_m + \omega_{ceo} \quad \phi_n = \omega_{ceo} t + \phi'_n

commensurate \quad \omega_q = n \omega_m \quad \phi_n = \phi_{CEP} + n \phi_m

\[ E(t) = \sum_{n} A_n(t) \cos(n \omega_m (t + \phi_m / \omega_m) + \phi_{CEP}) \]

Constant CEP requires that the frequencies are commensurate and the relative phases form an arithmetic series.
Ingredients of an attosecond single-cycle optical pulse:

1. Broad spectrum – 2 or more octaves
2. In phase condition
3. Constant carrier envelope phase:
   • Commensurate frequencies
   • Constant phase difference between adjacent spectral components
4. Stable and controllable carrier envelope phase
Methods of generating attosecond pulses

A High-order harmonic generation of phase-stabilized femtosecond pulse

Advantages: single pulse
100 attosecond
Disadvantages: 30-100 eV photons
very low power
constitutes a few cycles

Methods of generating attosecond pulses

High-order stimulated Raman scattering using molecular modulation

Advantages: IR-UV region, good power, single-cycle
Disadvantages: complex setup, 8-50 fs pulse spacing, limited to ~300-500 as

M.Y. Shverdin et al., PRL 94, 033904 (2005)
Three-step model

Isolated Attosecond-pulse production

Attosecond Bursts Trace the Electric Field of Optical Laser Pulses

The familiar textbook sketch of light's oscillating electric field can now be drawn directly from measurements.

Figure 3 | Evidence of delayed photoemission. a, The 4f and conduction-band spectrograms, following cubic-spline interpolation of the measured data.
Common theme
Photon energies 30 eV to 100 eV (EUV to soft x-ray region)

Traditional pump-probe measurements
Photoelectron or photoion detection

Weak pulses - long signal acquisition time
Molecular modulation is analogous to electro-optic modulation

Refractive Index: \( n = n_0 + \delta \cos \omega_m t \)

\( \omega_q = \omega_0 + q \omega_m \quad q = -2, -1, 0, 1, 2, 3, \ldots \)

Alexei Sokolov
Steve Harris
Two strong laser fields adiabatically drive the molecules into a maximally coherent state.

Maximal coherence, $\rho_{ab} = 0.5$
Coherent Molecular Excitation

\[ H_{\text{eff}} = -\hbar \begin{bmatrix} \Omega_{aa} & \Omega_{ab} \\ \Omega_{ba} & \Omega_{bb} - \delta \end{bmatrix} \]

\[ \Omega_{aa} = \frac{1}{2} \sum_q a_q |E_q|^2 \]
\[ \Omega_{bb} = \frac{1}{2} \sum_q b_q |E_q|^2 \]
\[ \Omega_{ab} = \Omega_{ba}^* = \frac{1}{2} \sum_q d_q E_q E_{q+1}^* \]

\[ a_q = \frac{1}{2\hbar^2} \sum_j \left( \frac{|\mu_{ja}|^2}{\omega_j - \omega_a - \omega_q} + \frac{|\mu_{ja}|^2}{\omega_j - \omega_a + \omega_q} \right) \]
\[ b_q = \frac{1}{2\hbar^2} \sum_j \left( \frac{|\mu_{jb}|^2}{\omega_j - \omega_b - \omega_q} + \frac{|\mu_{jb}|^2}{\omega_j - \omega_b + \omega_q} \right) \]
\[ d_q = \frac{1}{2\hbar^2} \sum_j \left( \frac{\mu_{aj}\mu_{jb}}{\omega_j - \omega_b - \omega_q} + \frac{\mu_{aj}\mu_{jb}}{\omega_j - \omega_a + \omega_q} \right) \]
Solution to the Hamiltonion:

- **Eigenfunctions**
  \[ |\pm\rangle = \cos \theta^{(\pm)} |a\rangle + \sin \theta^{(\pm)} e^{-i\phi} |b\rangle \quad \Omega_{ab} = |\Omega_{ab}| e^{i\phi} \]

  \[
  \tan \theta^{(\pm)} = \frac{2|\Omega_{ab}|}{\Omega_{aa} - \Omega_{bb} + \delta \pm \sqrt{\left(\Omega_{aa} - \Omega_{bb} + \delta\right)^2 + 4|\Omega_{ab}|^2}}
  \]

- **Eigenvalues**
  \[
  E_{\text{eff}} = -\hbar \lambda^{(\pm)} = \frac{\hbar}{2} \left(\Omega_{aa} + \Omega_{bb} - \delta\right) \pm \frac{\hbar}{2} \sqrt{\left(\Omega_{aa} - \Omega_{bb} + \delta\right)^2 + 4|\Omega_{ab}|^2}
  \]

- **Coherence**
  \[
  \rho_{ab}^{(\pm)} = \frac{1}{2} e^{i\phi} \sin 2\theta^{(\pm)} = \pm \frac{|\Omega_{ab}|}{\sqrt{\left(\Omega_{aa} - \Omega_{bb} + \delta\right)^2 + 4|\Omega_{ab}|^2}}
  \]

  \[
  |\rho_{ab}| = 0.5 \quad |\Omega_{ab}| \gg |\Omega_{aa} - \Omega_{bb} + \delta|
  \]
Adiabatically prepared molecules modulate the driving fields producing a wide comb of sidebands.

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

At maximum coherence, \( \rho_{ab} = 0.5 \) the dispersion and coupling terms become comparable. Phase-matching is then not important, and generation is collinear.

Courtesy M. Shverdin
Stimulated Raman Scattering

Traditional SRS:

- Generation occurs at **high gas pressure**
- Molecular excitation occurs **on-resonance**
- Anti-Stokes generation occurs **off-axis**
- Few Stokes and anti-Stokes orders are observed.

Courtesy M. Shverdin
Why low temperature

1. Put all molecules into one ro-vibrational state
   make all molecules contribute to the process

\[
\frac{\partial E_q}{\partial \xi} = \frac{iN\hbar \omega_q}{\varepsilon_0 c} \left( a_q \rho_{aa} E_q + b_q \rho_{bb} E_q + d_{q-1} \rho_{ba} E_{q-1} + d_q^* \rho_{ab} E_{q+1} \right)
\]

2. Reduce Doppler width to increase the coherence
   molecules with equal detuning but opposite in sign will
   off-set their contribution to the coherence build up

\[
\Delta \nu_D = \frac{v_0}{c} \sqrt{\frac{2kT}{m}} = \frac{1}{\lambda_0} \sqrt{\frac{2kT}{m}} = 1.285 \times 10^{11} \sqrt{\frac{T(K)}{\mu}}
\]

\[
a_q = \frac{1}{2\hbar^2} \sum_j \left( \frac{\left| \mu_{ja}^2 \right|}{\omega_j - \omega_a - \omega_q} + \frac{\left| \mu_{ja}^2 \right|}{\omega_j - \omega_a + \omega_q} \right)
\]

\[
b_q = \frac{1}{2\hbar^2} \sum_j \left( \frac{\left| \mu_{jb}^2 \right|}{\omega_j - \omega_b - \omega_q} + \frac{\left| \mu_{jb}^2 \right|}{\omega_j - \omega_b + \omega_q} \right)
\]

\[
d_q = \frac{1}{2\hbar^2} \sum_j \left( \frac{\mu_{aj} \mu_{jb}}{\omega_j - \omega_b - \omega_q} + \frac{\mu_{aj} \mu_{jb}}{\omega_j - \omega_a + \omega_q} \right)
\]

\[
T = 300K \quad \Delta \nu_D, D_2 = 778 \text{ MHz}
\]

\[
T = 77K \quad \Delta \nu_D, D_2 = 389 \text{ MHz}
\]

\[
T = 300K \quad \Delta \nu_D, H_2 = 1100 \text{ MHz}
\]
D₂ Vibration Spectra: 16 sidebands, spaced by 2994 cm⁻¹

H₂ Rotation Spectra: 29 sidebands, spaced by 587 cm⁻¹

Multiplicative Spectra: ~ 200 sidebands, spaced by < 587 cm⁻¹
Motivation

• form subfemtosecond pulses
• produce THz pulse train
• generate tunable high power vacuum uv pulses
• arbitrary waveform synthesis

Use gas phase hydrogen at room temperature

Pros:
- large Raman transition of 4155 cm\(^{-1}\) for Q(1)
- 2/3 population in single quantum state (v=0, j=1) at room temperature
- many parameters are known
- nondestructible
- room temperature is easy to operate

Cons:
- large Doppler width
- requires two tunable high-power high-resolution lasers spaced 4155 cm\(^{-1}\) apart
- smaller pulse train pulse-to-pulse spacing
\[ \Omega_{aa} = \frac{1}{2} \sum_q a_q |E_q|^2 \]
\[ \Omega_{bb} = \frac{1}{2} \sum_q b_q |E_q|^2 \]
\[ \Omega_{ab} = \Omega_{ba}^* = \frac{1}{2} \sum_q d_q E_q E^*_q+1 \]
\[ H_{eff} = -\hbar \begin{bmatrix} \Omega_{aa} & \Omega_{ab} \\ \Omega_{ba} & \Omega_{bb} - \delta \end{bmatrix} \]
\[ d_q = \frac{1}{2\hbar^2} \sum_j \left( \frac{\left| \mu_{ja} \right|^2}{\omega_j - \omega_a - \omega_q} + \frac{\left| \mu_{ja} \right|^2}{\omega_j - \omega_a + \omega_q} \right) \]
\[ b_q = \frac{1}{2\hbar^2} \sum_j \left( \frac{\left| \mu_{jb} \right|^2}{\omega_j - \omega_b - \omega_q} + \frac{\left| \mu_{jb} \right|^2}{\omega_j - \omega_b + \omega_q} \right) \]
\[ d_q = \frac{1}{2\hbar^2} \sum_j \left( \frac{\mu_{ja} \mu_{jb}}{\omega_j - \omega_b - \omega_q} + \frac{\mu_{ja} \mu_{jb}}{\omega_j - \omega_a + \omega_q} \right) \]
\[ \frac{\partial \rho_{aa}}{\partial \tau} = i(\Omega_{ab} \rho_{ba} - \Omega_{ba} \rho_{ab}) + \gamma_\rho \rho_{bb} \]
\[ \frac{\partial \rho_{bb}}{\partial \tau} = -i(\Omega_{ab} \rho_{ba} - \Omega_{ba} \rho_{ab}) - \gamma_\rho \rho_{bb} \]
\[ \frac{\partial \rho_{ab}}{\partial \tau} = i(\Omega_{aa} - \Omega_{bb} + \delta + i\gamma_\rho) \rho_{ab} + i\Omega_{ab} (\rho_{bb} - \rho_{aa}) \]
\[ \frac{\partial E_q}{\partial z} = -j \eta \hbar \omega_q \left[ N(a_q \rho_{aa} E_q + d_q \rho_{bb} E_q) + 0.666N(b^*_q \rho_{ab} E_{q-1} + b^*_{q+1} \rho_{ab} E_{q+1}) \right] \]

Simulation

Thanks to Prof. Fam Le Kien for the full set of matrix element data.
Experimental Arrangement

- Laser frequency: $\delta \omega_{\text{laser}} \sim 100$ MHz
- Laser pulse duration: $\Delta t_{\text{laser}} \sim 5$ ns
- Wavelengths: $\lambda_0 = 589$ nm, $\lambda_{-1} = 780$ nm
- Laser intensity: $\sim 12$ GW/cm$^2$
- Angular frequency: $\Omega_{ab,z=0} \sim 2$ GHz
- Absorption coefficient: $\rho_{ab} \sim 0.4$
- Delay time: $t_{\text{delay}} \sim 0-1$ ns
- H$_2$ pressure: $\sim 1$ atm.
- H$_2$ Doppler width: 250-750 MHz

Diagram:
- Beam combiner
- Telescope
- MgF$_2$ lens
- q=-2 to 8
- q=8 and higher
Raman sidebands generated

q = -2, -1, 0, 1, 2, 3, 4, Etc.

\( q = 3, \lambda = 339.6 \text{ nm} \)
6 \( 238.6 \text{ nm} \)
9 \( 183.9 \text{ nm} \)
12 \( 149.6 \text{ nm} \)
14 \( 133.0 \text{ nm} \)

Total spectral span >70,000 cm\(^{-1}\)
(\(~500\) as)

15\(^{th}\) order at 126 nm observed
Note:

589 nm ↔ 16978 cm\(^{-1}\) (4 × 4155.2 = 16621 cm\(^{-1}\))

780 nm ↔ 12822.8 cm\(^{-1}\) (3 × 4155.2 cm\(^{-1}\) = 12465.6 cm\(^{-1}\))

The sidebands are not commensurate.

New input wavelengths:

\(\omega_0 = 16621\) cm\(^{-1}\) (602 nm)
\(\omega_{-1} = 12465.6\) cm\(^{-1}\) (802 nm)

These wavelengths produce a commensurate set of sidebands, as shown on the right:

\[ \omega_q = n \omega_m \]

<table>
<thead>
<tr>
<th>Raman Order</th>
<th>nm</th>
<th>cm(^{-1})</th>
<th>4 wave-mixing order</th>
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</tr>
<tr>
<td>9</td>
<td>185</td>
<td>54015</td>
<td></td>
</tr>
</tbody>
</table>
Phase adjustment Setup

Dye Laser
602nm

Ti:Sapphier Laser
802nm

Xe
602nm

802nm

PMT

Phase Modulator

Raman Sidebands Generation

Four Wave Mixing

(achromatic lens f=25cm for IR to UV)

(achromatic lens f=8cm for IR to UV)

LC phase modulator

\[ \lambda_{\text{cutoff}} \]
430 nm --> 380 nm

6 sidebands

7 sidebands

![Graph 1: Phase Shift vs. Applied Voltage](image1)

![Graph 2: Phase Shift vs. Applied Voltage](image2)
UV sidebands are generated at efficiencies $10^{-8}$ to $10^{-12}$ by a four-wave mixing process in a xenon cell at ~100 torr. Phasematching allows only the two photons up one photon down type of conversion. A total of $n-1$ different UV sidebands, beginning at the next short wavelength sideband, are generated.
Multiple quantum paths interference

Four wave mixing:

\[ \omega_5 + \omega_7 - \omega_4 = \omega_8 \]

\[ \omega_6 + \omega_5 - \omega_3 = \omega_8 \]

\[ \omega_6 + \omega_7 - \omega_5 = \omega_8 \]
Four Wave Mixing in Xe

\[ E_{\alpha} = \chi E_i E_j E_k^* \quad \phi_{\alpha} = \phi_i + \phi_j - \phi_k \]

\[ 7 : \begin{align*}
5 + 5 - 3 &= \quad \phi_5 = \phi_1 + 4s \\
5 + 4 - 2 &= \phi_4 = \phi_2 + 2s \\
5 + 3 - 1 &= \phi_3 = \phi_4 - \phi_2 = 2s \\
4 + 4 - 1 &= \phi_1 = \phi_3 - \phi_1 = 3t
\end{align*} \]

\[ \Rightarrow \quad \begin{align*}
\phi_5 &= \phi_2 + 3t \\
\phi_4 &= \phi_1 + 3t \\
3t - 4s &= 2s - 3t
\end{align*} \]

\[ t = s \quad \Rightarrow \quad \begin{align*}
\varphi_2 &= \varphi_1 + \Delta \\
\varphi_3 &= \varphi_1 + 2\Delta \\
\varphi_4 &= \varphi_1 + 3\Delta \\
\varphi_5 &= \varphi_1 + 4\Delta
\end{align*} \]
In phase condition

\[7 = 6+6-5, 6+5-4, 6+4-3, 6+3-2, 6+2-1.\]

\[5+5-3, 5+4-2, 5+3-1.\]

\[4+4-1\]

\[6, 5, 4 : 6+6-5, 6+5-4 \quad \Phi_{65} = \Phi_{54}\]

\[+3 : 6+4-3, 5+5-3 \quad \Phi_{65} = \Phi_{54} = \Phi_{43}\]

\[+2 : 6+3-2, 5+4-2 \quad \Phi_{65} = \Phi_{54} = \Phi_{43} = \Phi_{32}\]

\[+1 : 6+2-1, 5+3-1, 4+4-1 \quad \Phi_{65} = \Phi_{54} = \Phi_{43} = \Phi_{32} = \Phi_{21}\]
Searching in phase condition

- (a) 481 nm
- (b) 602 nm
- (c) 802 nm
- (d) 1203 nm

301 nm signal intensity (a. u.)

Relative phase shift (π radian)
How to measure the pulse width?

Autocorrelation is standard way to measure ultrafast pulsewidth. However it could not be done here because of the wide bandwidth.

**Solution:** Correlation using pulses formed by the sidebands themselves.

Synthesize two pulses from the subsets of sidebands and electronically delay one pulse with respect to the other. Measure the resulting four-wave signal with a photomultiplier.
Cross Correlation of Single Cycle Pulse Train

Sideband Orders

Simulation

Experiment
Cross correlation signal of incommensurate pulses

CEO frequency ~ 349 cm⁻¹
Waveform repeats every 96 fs
Pulse train

(a) Commensurate

(b) Incommensurate

E field amplitude (a. u.)

Time (fs)
Carrier envelope phase is constant to ~2.5 part in $10^6$

Total phase slip of <0.18 cycles over 1 million pulses
Status of sub-cycle optical pulse generation by molecular modulation

IAMS sub-cycle source

0.833 cycle per pulse
1.4 fs envelope
440 as cycle width
constant carrier envelope phase
2 ns pulse train duration
8.0 fs pulse spacing
~1 MW peak power

Total spectral span >70,000 cm⁻¹

Ingredients of an attosecond single-cycle optical pulse:

1. Broad spectrum – 2 or more octaves
2. In phase condition
3. Constant carrier envelope phase:
   • Commensurate frequencies
   • Constant phase difference between adjacent spectral components
4. Stable and controllable carrier envelope phase
CEP control
Phys. Rev. Lett. 102, 213902 (2009)
Summary and Outlook

- Generated commensurate pulse train
- Single pulse duration 1.4fs
- Sub-single-cycle pulse: 0.8 cycles
- CEP (carrier-envelope phase) control
- Sub-femtosecond pulse generation
- Arbitrary waveform
- Application for ultra-fast dynamics
Harmonics

- **0.833 cycle** per pulse
- **1.4 fs** envelope
- **440 as** cycle width
- constant carrier envelope phase
- 2 ns pulse train duration
- **8.0 fs** pulse spacing
- **~1 MW** peak power

- **~25,000 cm\(^{-1}\)**
- **~37,600 cm\(^{-1}\)**

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Attosecond pulse train

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<th>Nd:YAG Harmonics</th>
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<th>cm(^{-1})</th>
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<td>5</td>
<td>213</td>
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</table>

We can control the CEP and shape the pulse waveform. We can synthesize a waveform which like the XUV-IR combination.
Advance Concepts

Technology

- Generate subfemtosecond pulses: add more sidebands and improve sideband power
- Increase pulse-to-pulse spacing
- Develop control of carrier envelope phase
- Modulate in photonic crystal fiber
- Arbitrary waveform synthesis

Science

- Optical-deep uv attosecond pump-probe
- Tracing molecular vibrational wavepacket
- Low energy electron dynamics in atoms
- Electron dynamics in semiconductors: direct bandgap vs indirect bandgap
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