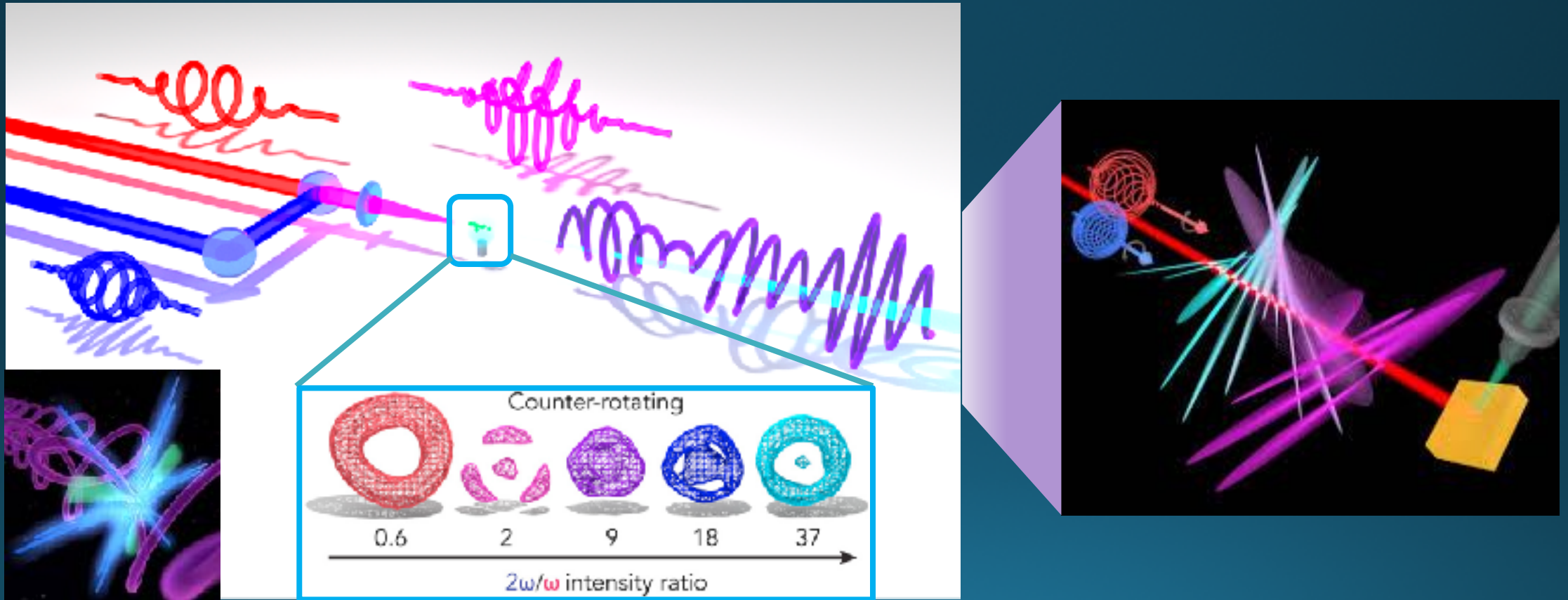


# Polarization Control of Attosecond High-Harmonic Waveforms via Helicity-Selective Circularly Polarized High Harmonic Generation

Kevin M. Dorney

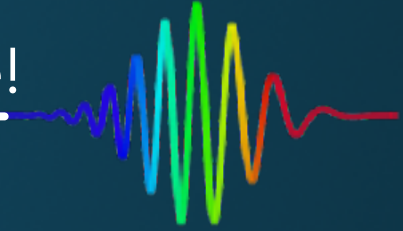
Kapteyn-Murnane Group, JILA and University of Colorado Boulder, USA





But First...

A Very Special Thanks to UFO XI Organizers and Many More!

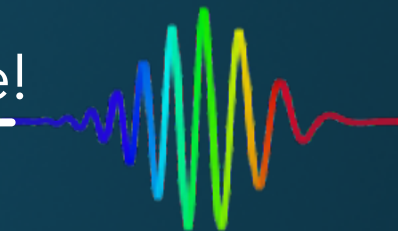






But First...

A Very Special Thanks to UFO XI Organizers and Many More!



Charles Durfee

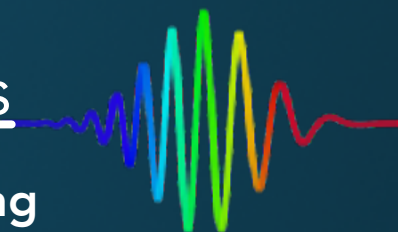


Alan Fry

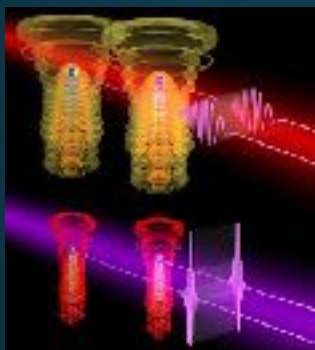




# Light and Materials Science in the KM Group: AMO Dynamics at Extreme Spatial and Temporal Scales

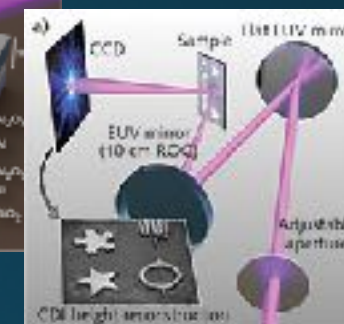
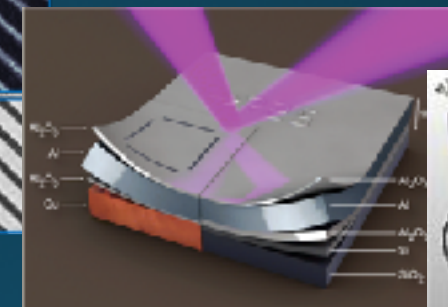


## Attosecond Extreme Nonlinear Optics



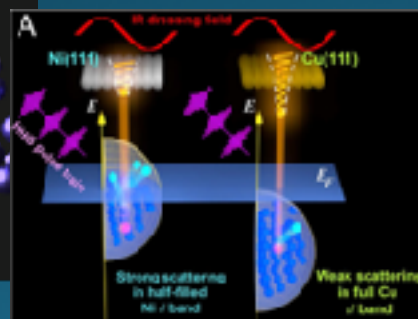
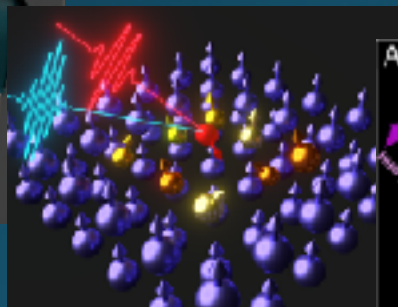
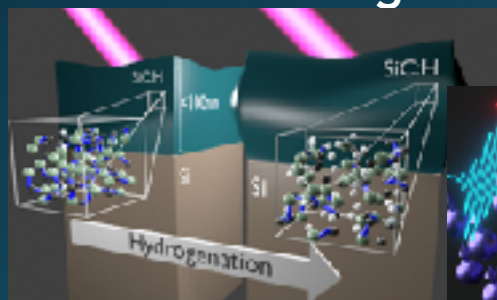
Popmintchev, Science 6265, 2015  
Dorney, PRL 118, 2017

## Coherent X-ray Imaging



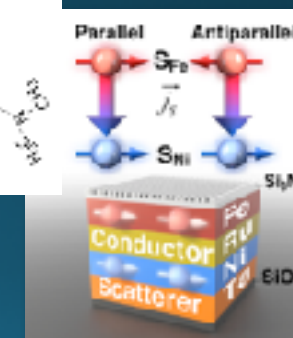
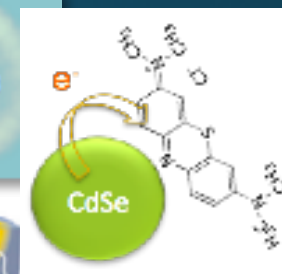
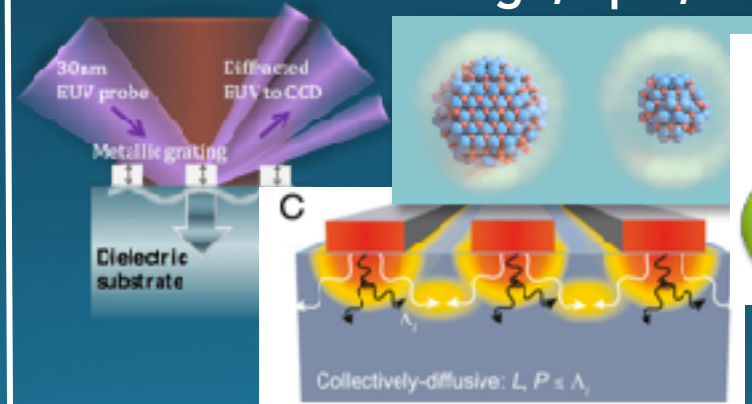
Gardener, Nat. Photon. 11, 2017  
Shanblatt, Nano Lett. 16, 2016

## Uncovering New Ultrafast Materials Science



Hernandez-Charapak, Nano Lett. 2017  
Tao, Science 353, 2016  
Chen, PNAS 114, 2017

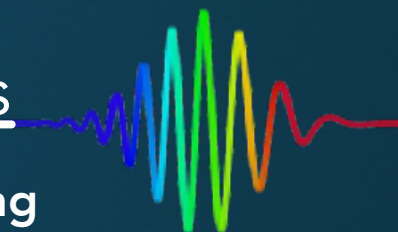
## Nanoscale Charge, Spin, Energy Transport



Hoogeboom-Pot, PNAS 112, 2015  
Ellis, JACS, 11 2015



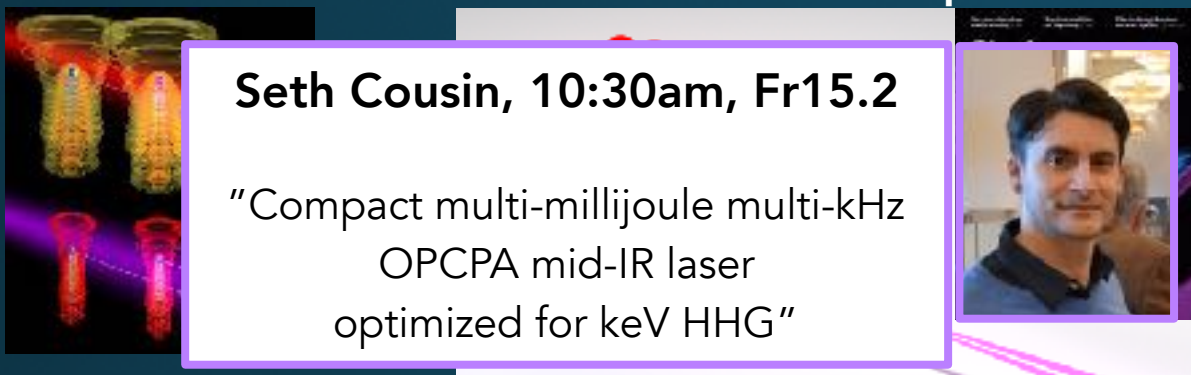
# Light and Materials Science in the KM Group: AMO Dynamics at Extreme Spatial and Temporal Scales



## Attosecond Extreme Nonlinear Optics

**Seth Cousin, 10:30am, Fr15.2**

"Compact multi-millijoule multi-kHz  
OPCPA mid-IR laser  
optimized for keV HHG"



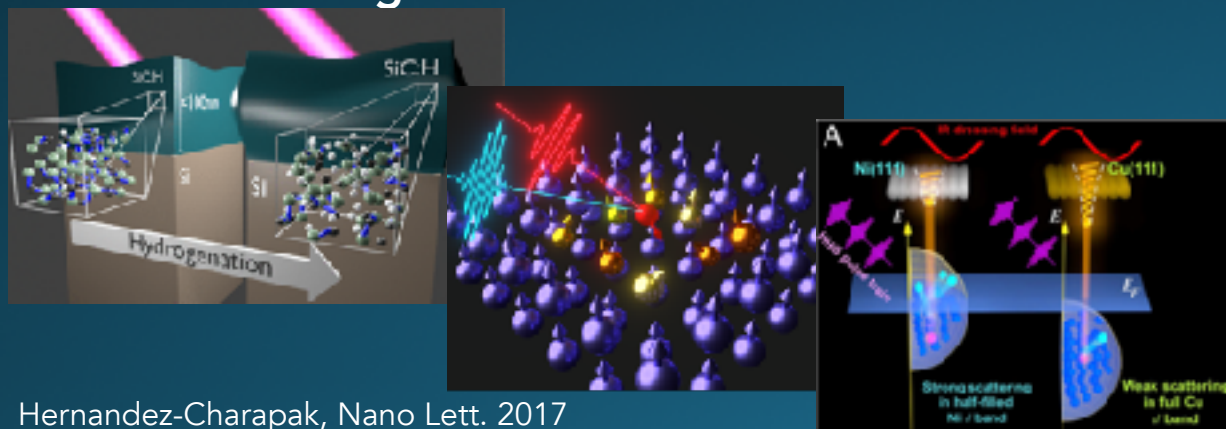
Popmintchev, Science 6265, 2015  
Dorney, PRL 118, 2017

## Coherent X-ray Imaging



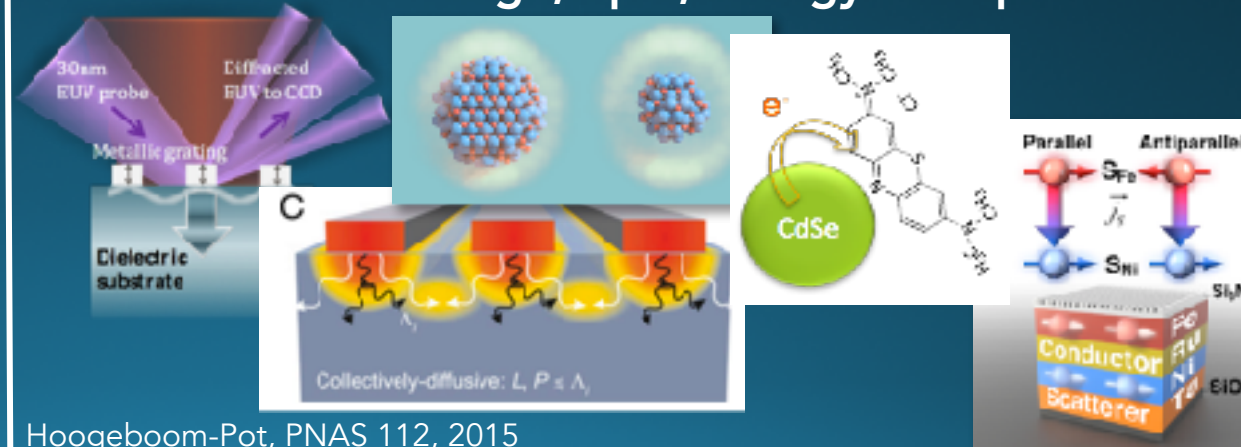
Gardener, Nat. Photon. 11, 2017  
Shanblatt, Nano Lett. 16, 2016

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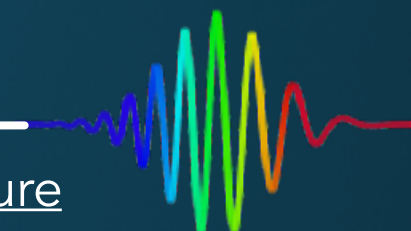
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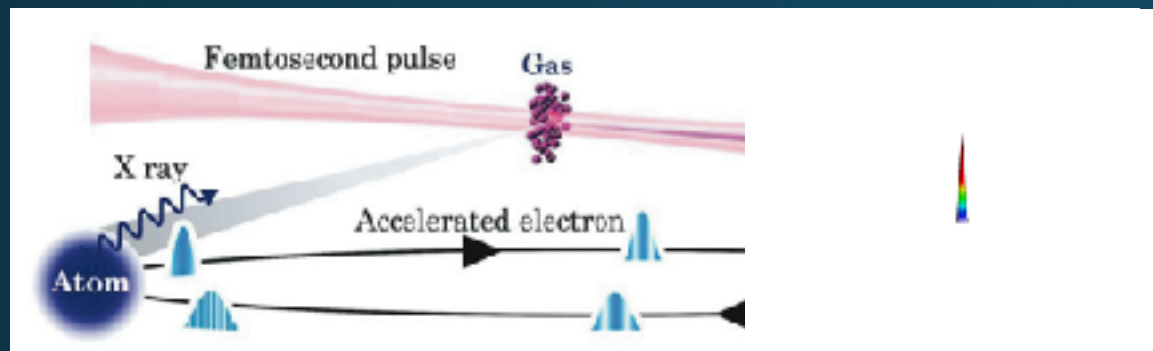
Hoogeboom-Pot, PNAS 112, 2015  
Ellis, JACS, 11 2015



# High Harmonic Generation (HHG): A Quantum Technology Tailored for Ultrafast Science



## Microscopic Picture



## Macroscopic Picture

**Kuchiev, JETP, 45. 404 (1987)**

Classical: Ferray, J. Phys. B., 1988; Corkum. PRL 1993

QM: Krause, Shafer, Kulander, PRL 1992; Lewenstein, PRA, 1994

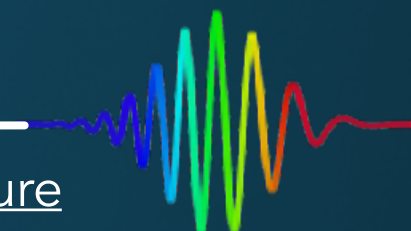
**Video: Courtesy of Carlos Hernandez-Garcia**

Chen, PNAS, 111, 2014

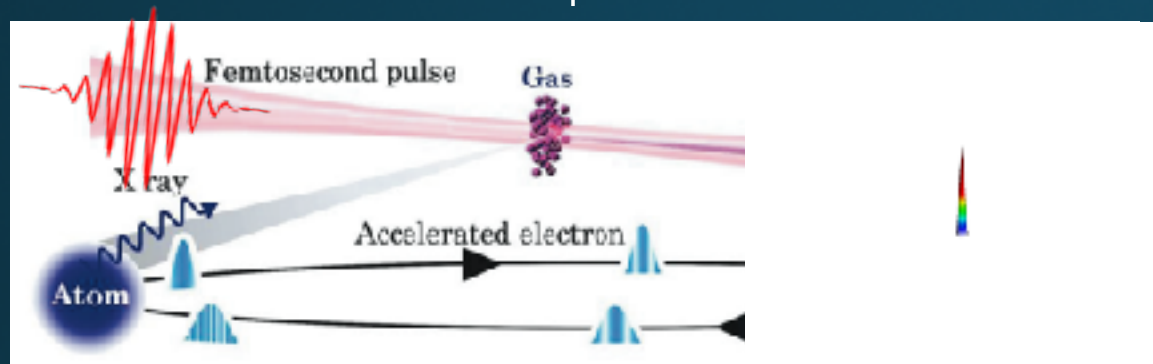
Hernandez-Garcia, Opt Exp, 106, 2017

Popmintchev, Science, 6265, 2015

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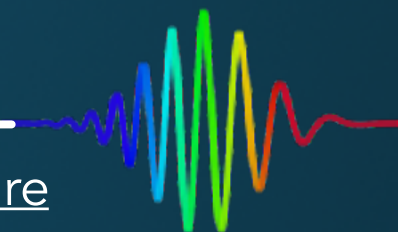
**Video: Courtesy of Carlos Hernandez-Garcia**

Chen, PNAS, 111, 2014

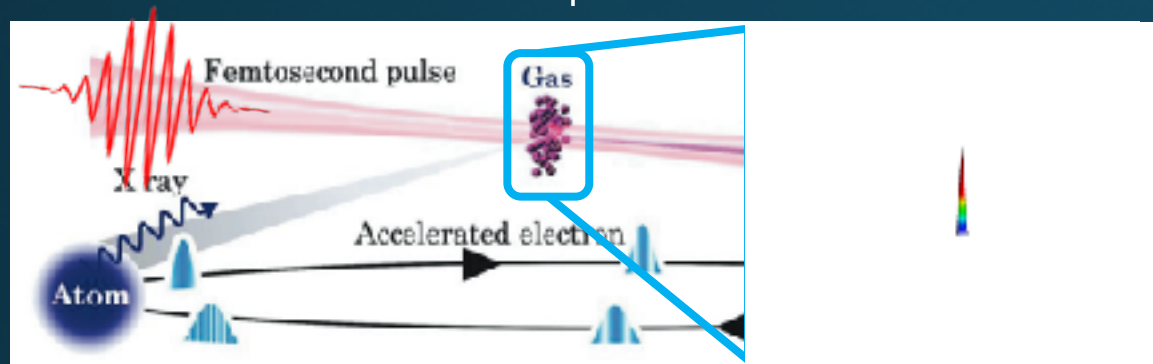
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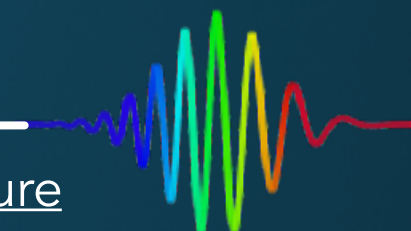
Chen, PNAS, 111, 2014

Hernandez-Garcia, Opt Exp, 106, 2017

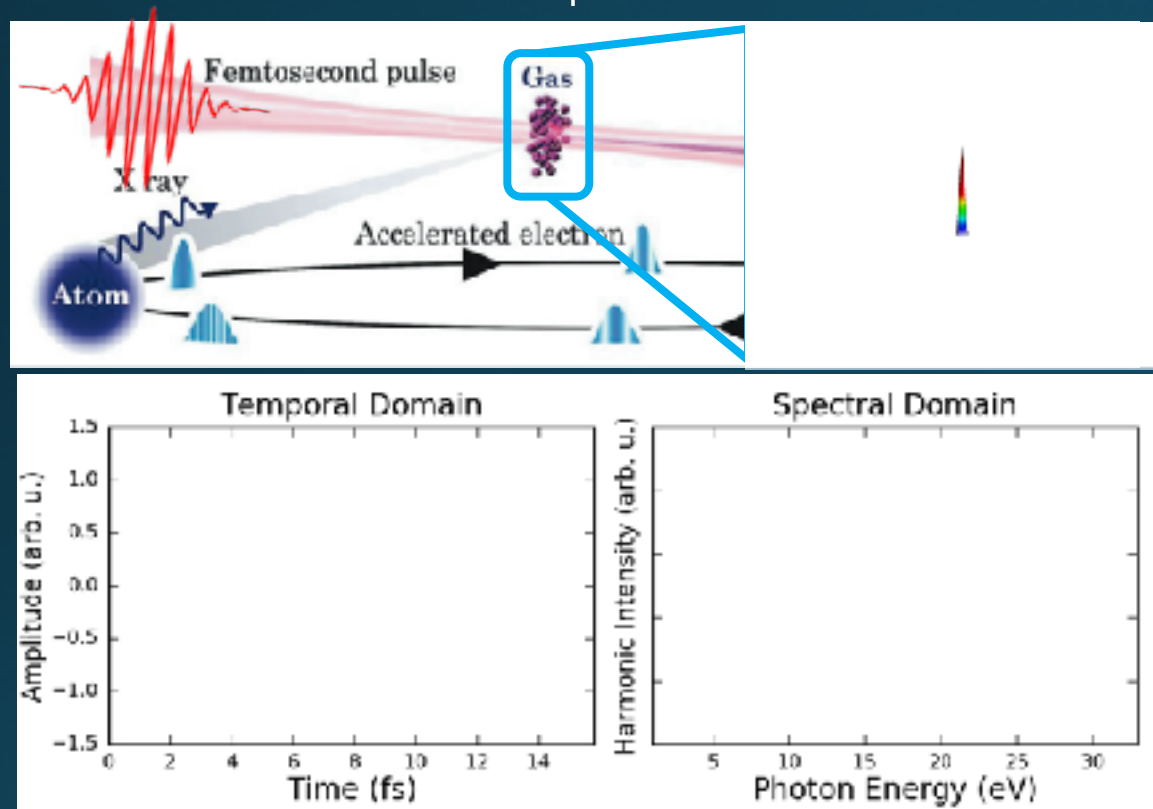
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## Macroscopic Picture

Kuchiev, JETP, 45. 404 (1987)

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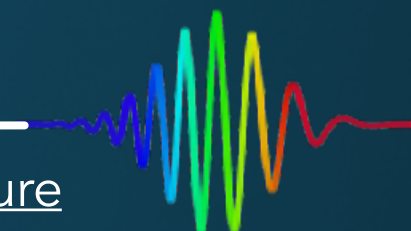
Video: Courtesy of Carlos Hernandez-Garcia

Chen, PNAS, 111, 2014

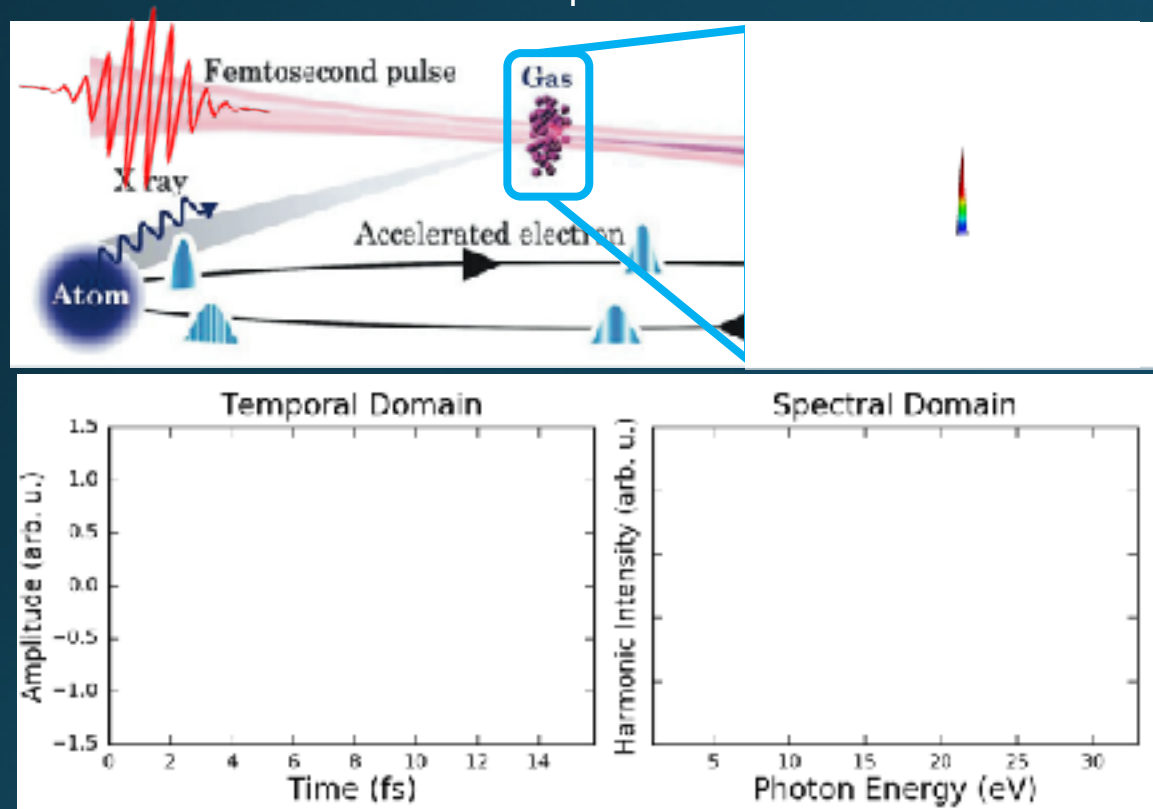
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Chen, PNAS, 111, 2014

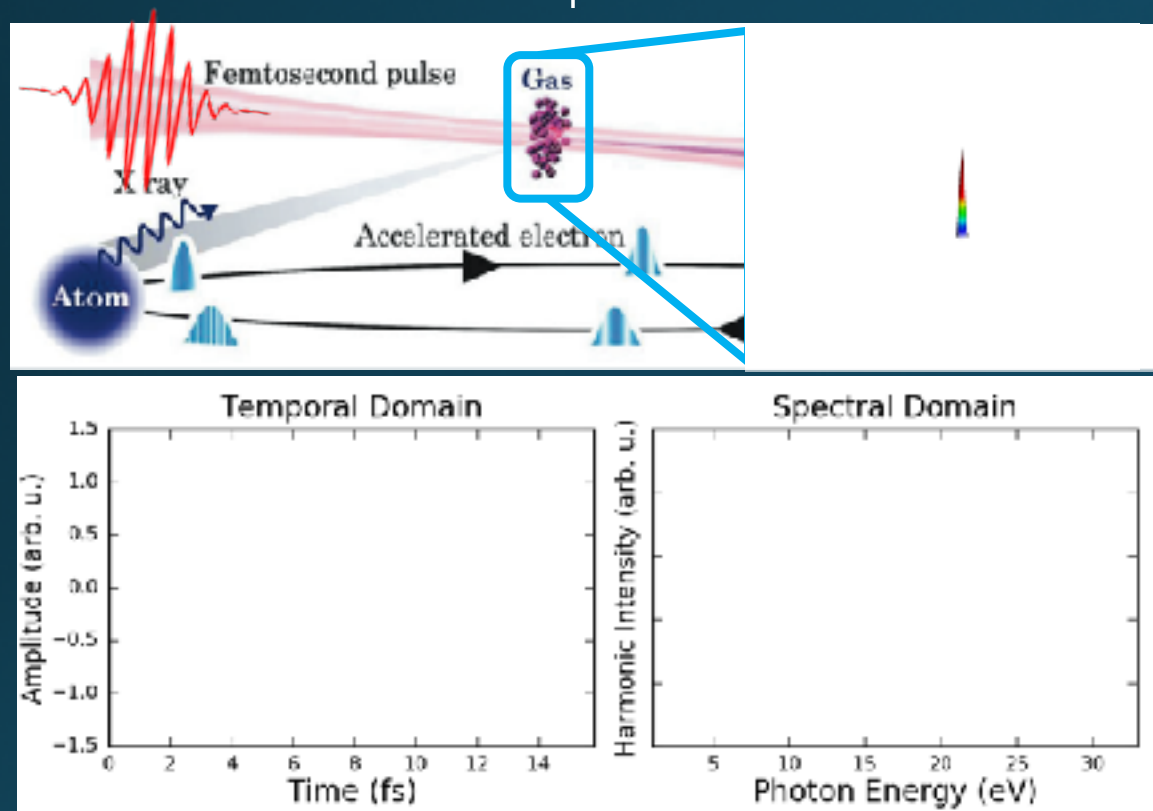
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## Microscopic Picture



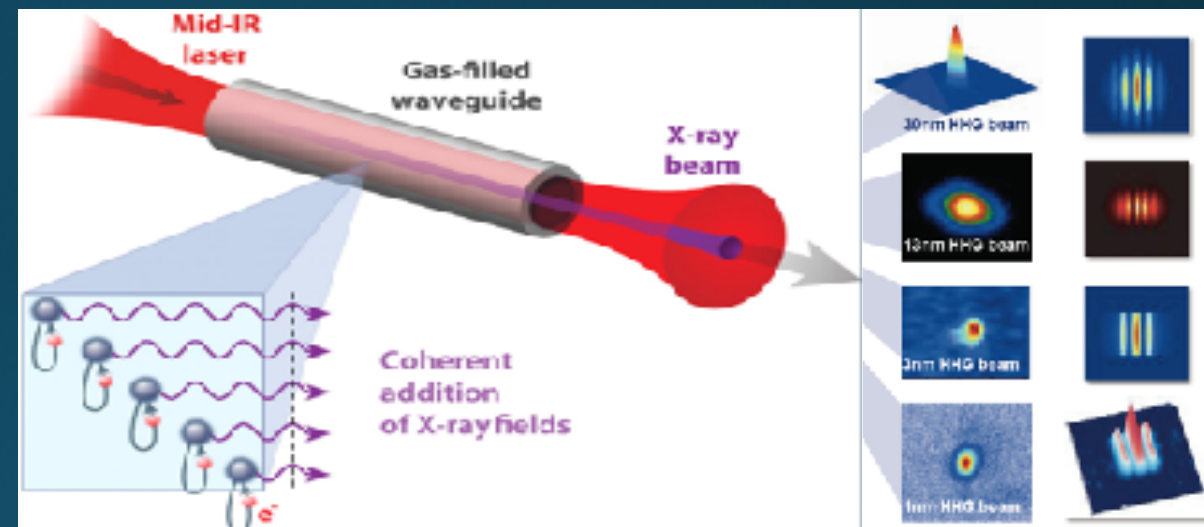
Kuchiev, JETP, 45. 404 (1987)

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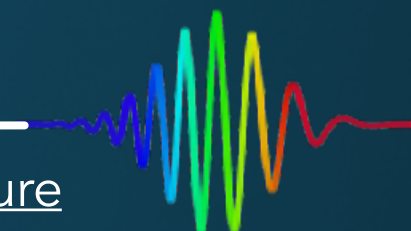


Chen, PNAS, 111, 2014

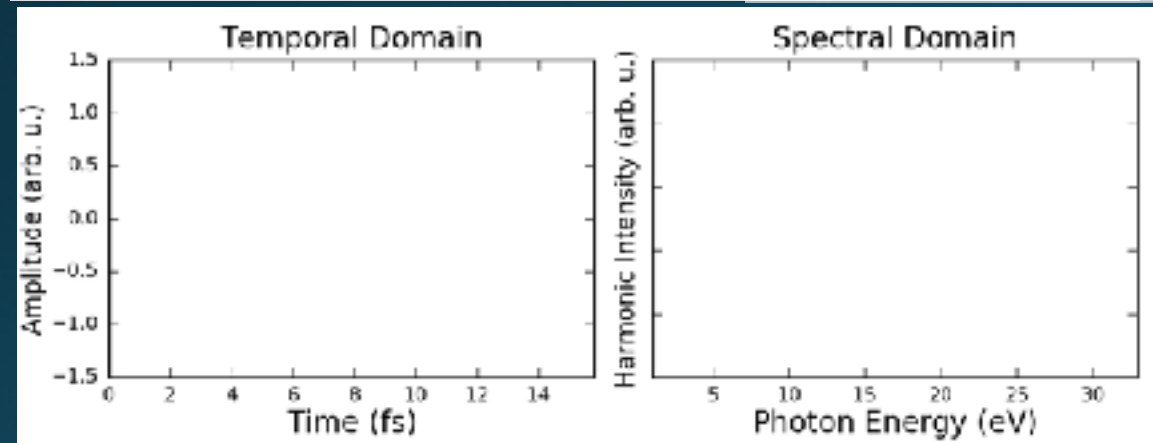
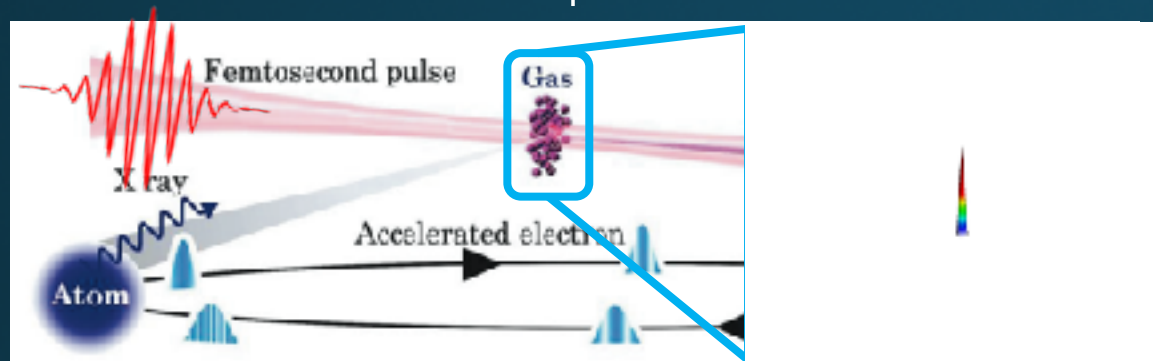
Hernandez-Garcia, Opt Exp, 106, 2017

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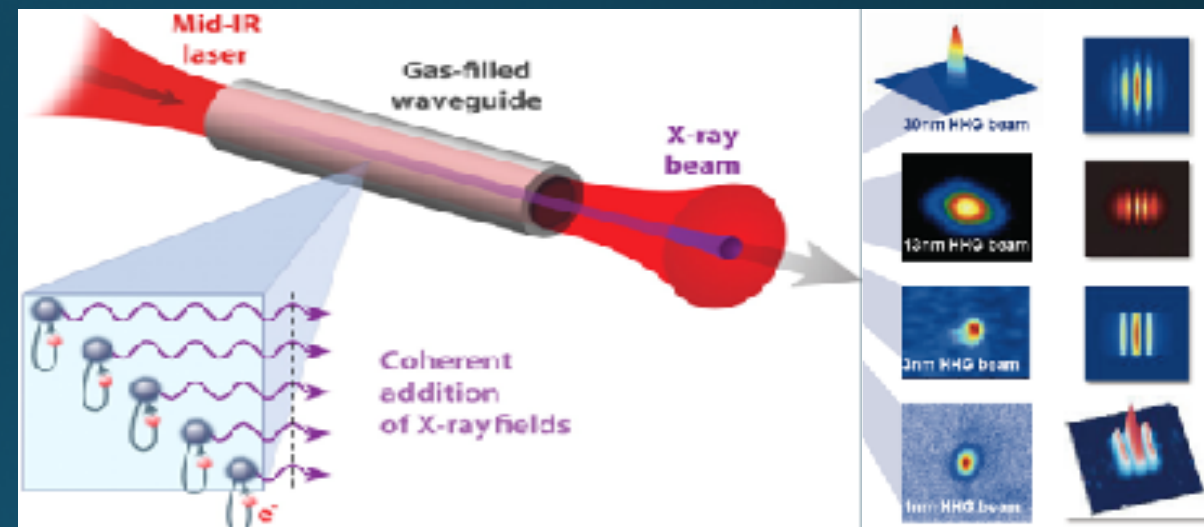
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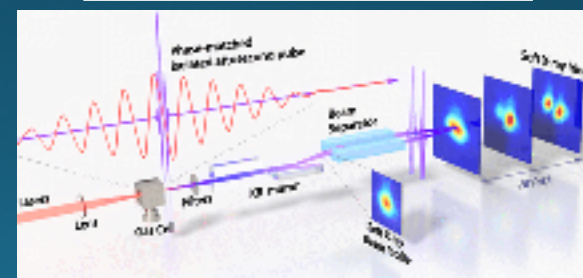
QM: Krause, Shafer, Kulander, PRL 1992; Lewenstein, PRA, 1994

Video: Courtesy of Carlos Hernandez-Garcia

## Macroscopic Picture



## Isolated Attosecond Pulses



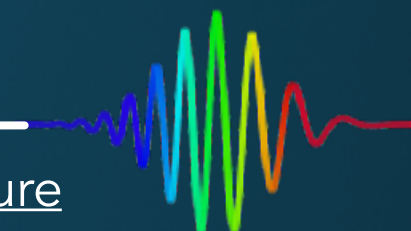
Chen, PNAS, 111, 2014

Hernandez-Garcia, Opt Exp, 106, 2017

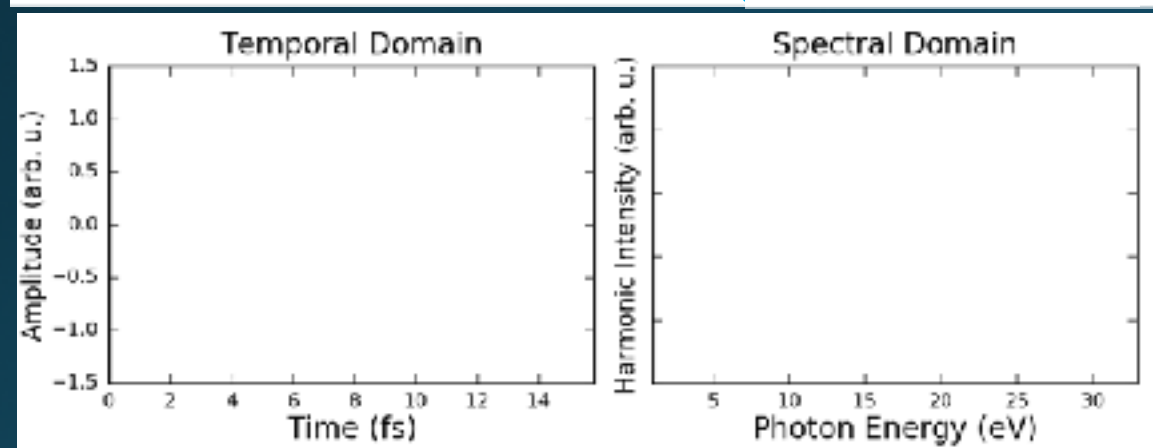
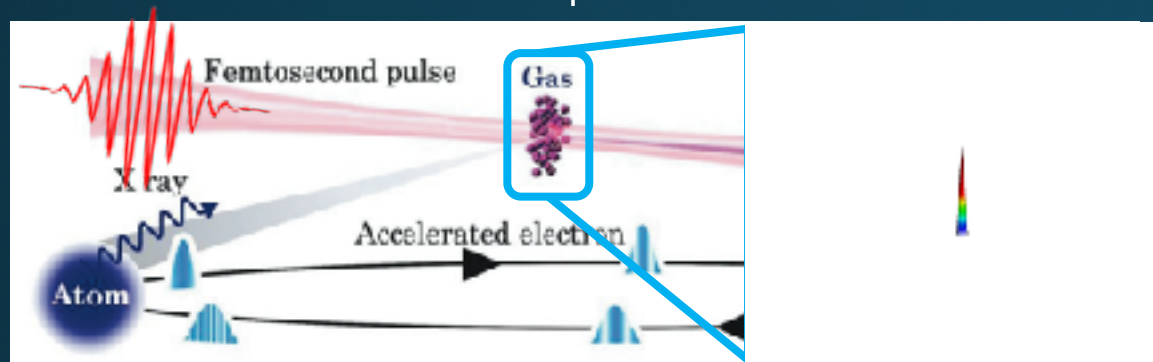
Popmintchev, Science, 6265, 2015



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## Microscopic Picture



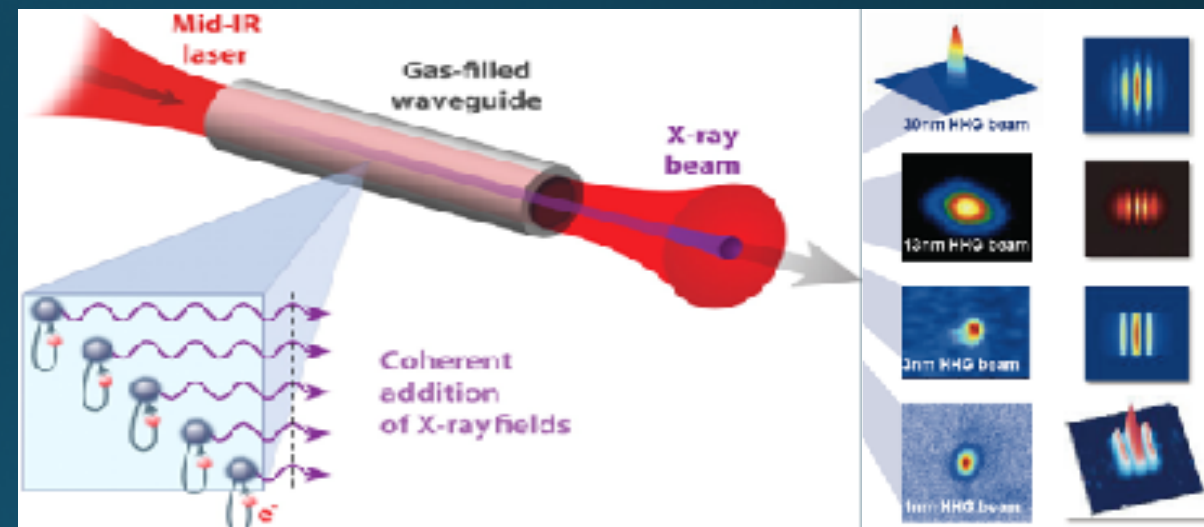
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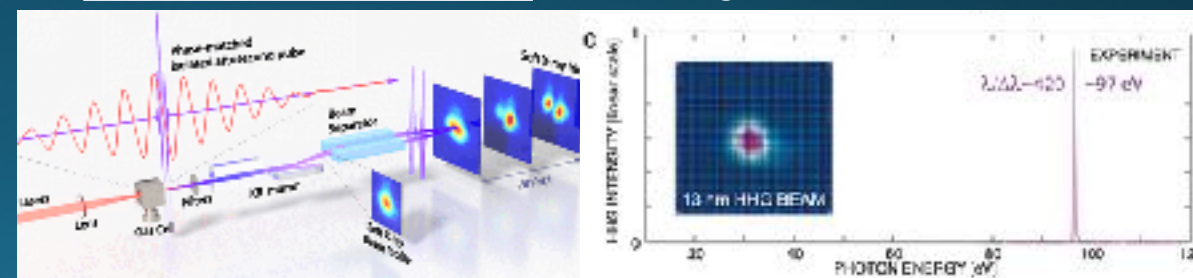
Video: Courtesy of Carlos Hernandez-Garcia

## Macroscopic Picture



## Isolated Attosecond Pulses

## Bright, Isolated Harmonics

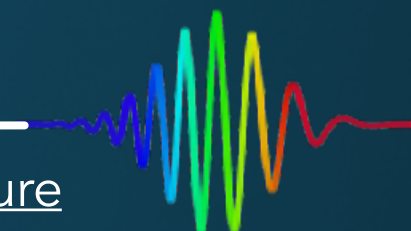


Chen, PNAS, 111, 2014

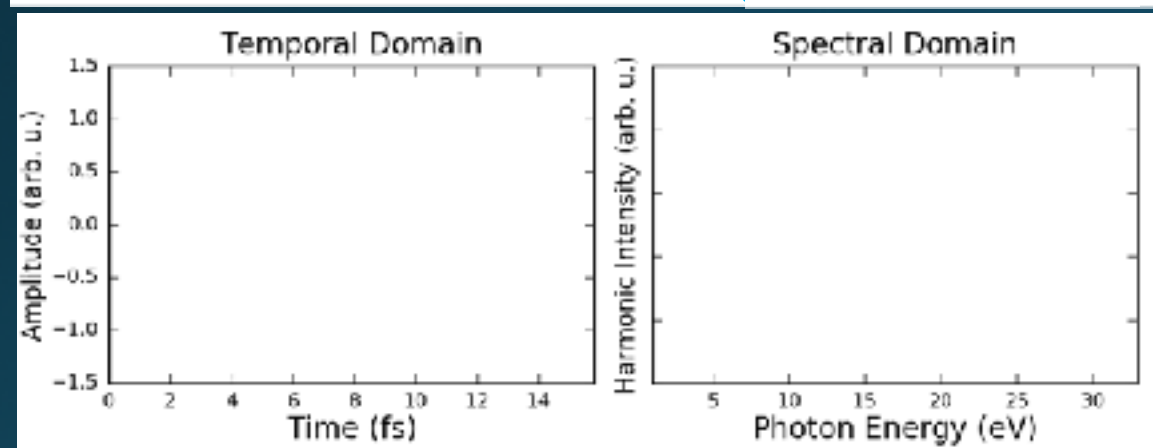
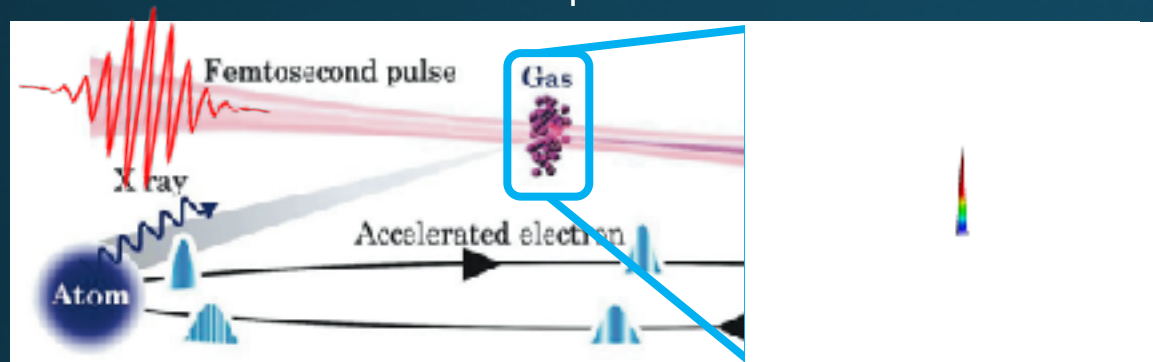
Hernandez-Garcia, Opt Exp, 106, 2017

Popmintchev, Science, 6265, 2015

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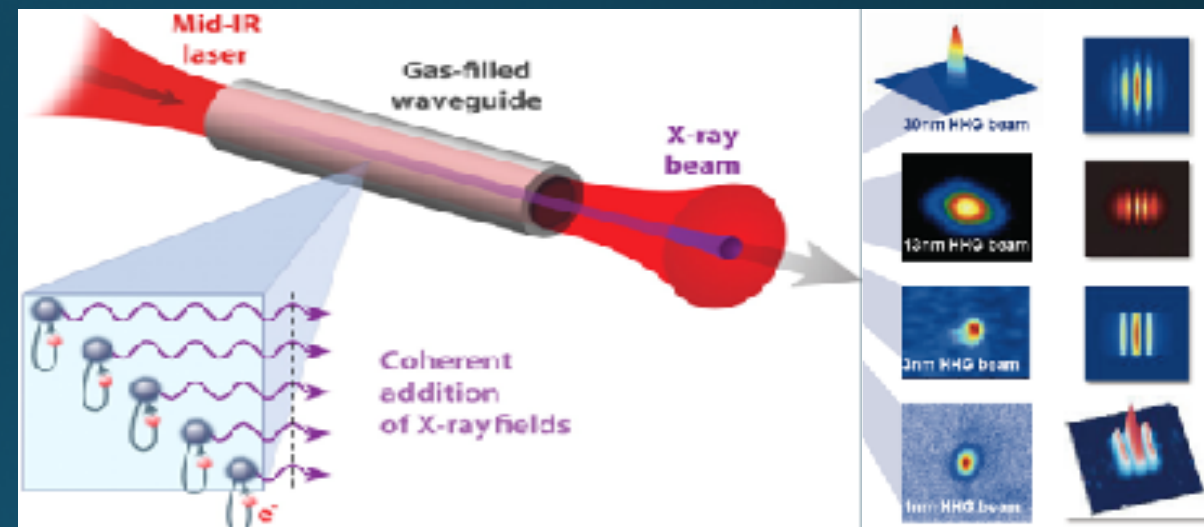
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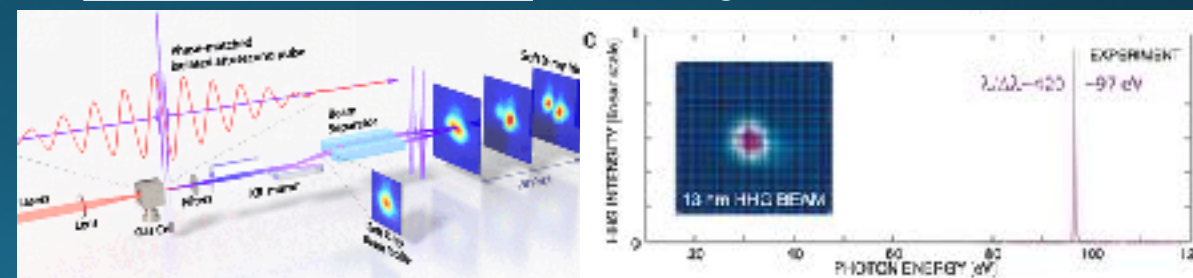
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## Macroscopic Picture



## Isolated Attosecond Pulses

## Bright, Isolated Harmonics



Chen, PNAS, 111, 2014

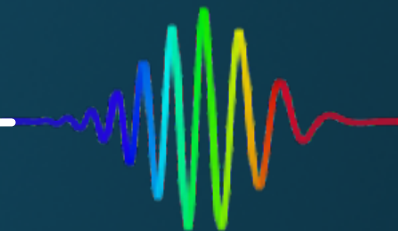
Hernandez-Garcia, Opt Exp, 106, 2017

Popmintchev, Science, 6265, 2015





# Polarization Control in HHG: Coming Full Circle in Quantum Control of High Harmonics



- External control of time, frequency, space in EUV and beyond... Polarization?

Video: Courtesy of Carlos Hernandez-Garcia

One Color Elliptical Driver

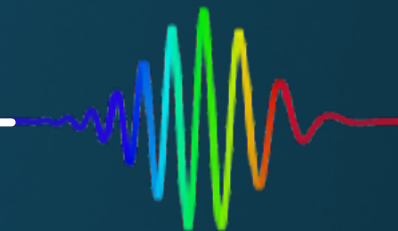


Budil, Phys. Rev. A. **48**, 1993

Weihe, J. Opt. Soc. Am. 13, 1996



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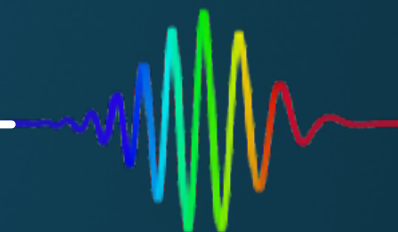


Budil, Phys. Rev. A. **48**, 1993

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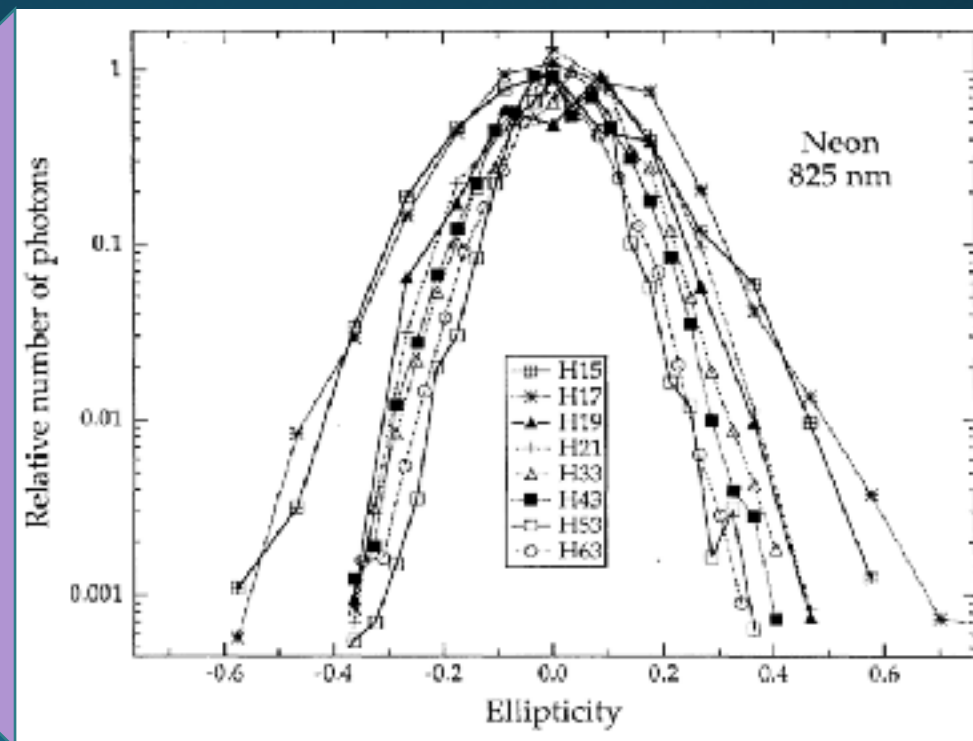
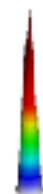
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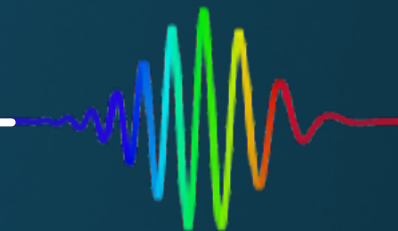
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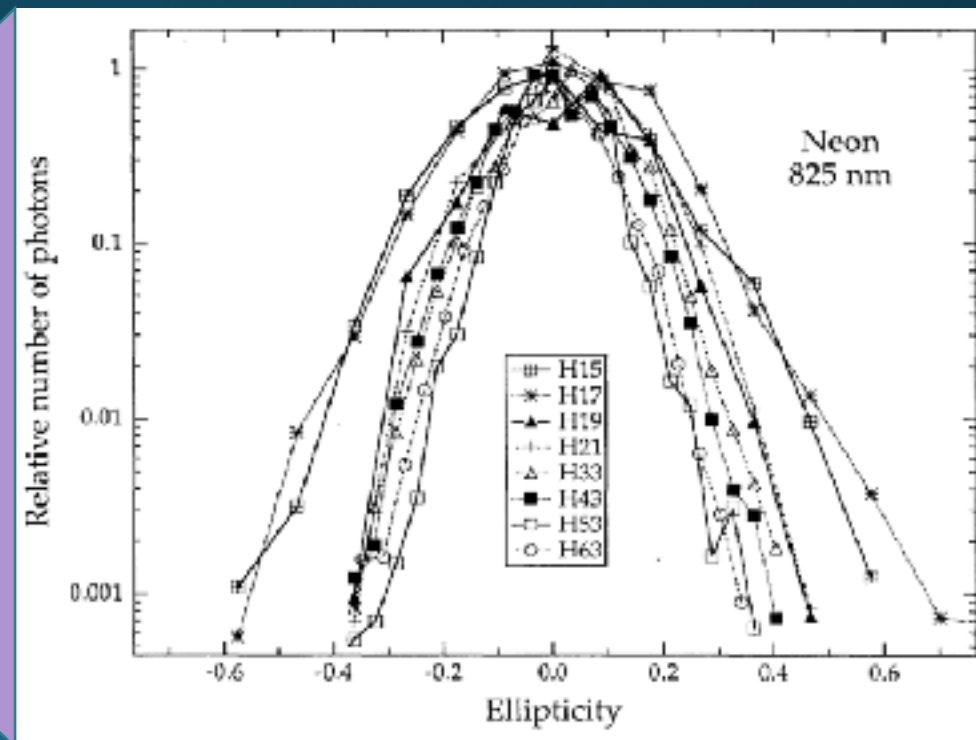
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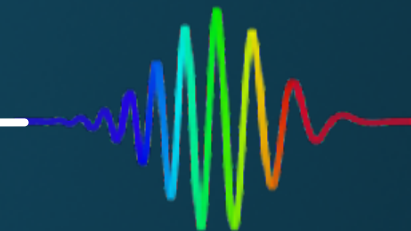


Budil, Phys. Rev. A. **48**, 1993

Weihe, J. Opt. Soc. Am. 13, 1996



# HHG in a Whole New Dimension: Reshaping the Frontier of Attosecond Science

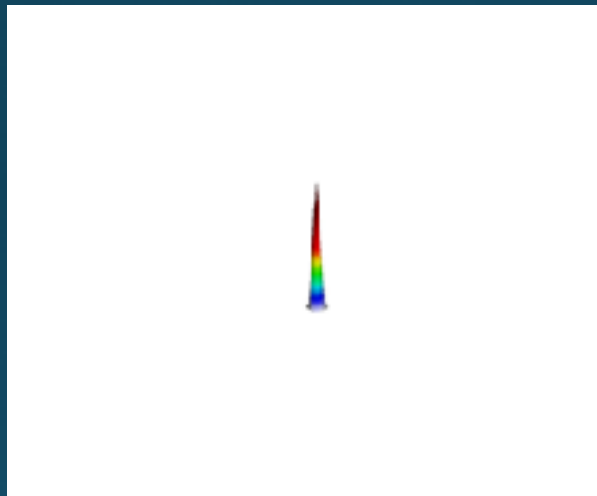


Videos: Courtesy of Carlos Hernandez-Garcia

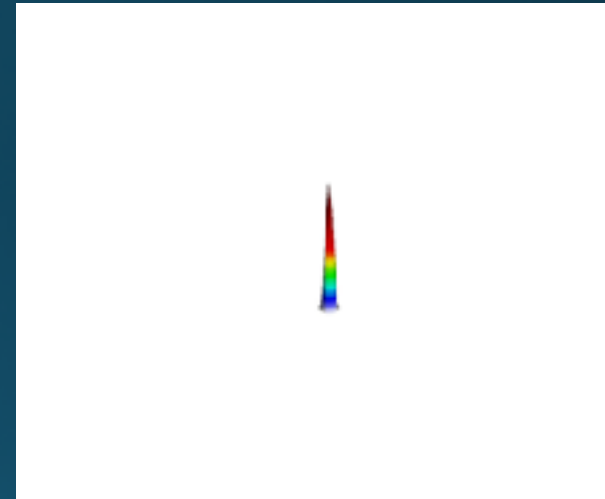
One Color Linear Driver



One-Color Circular Driver



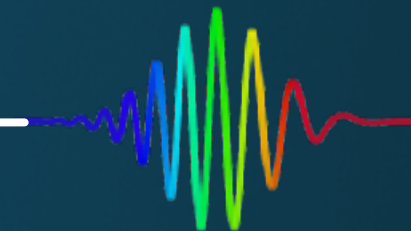
Two-Color Counter-Rotating







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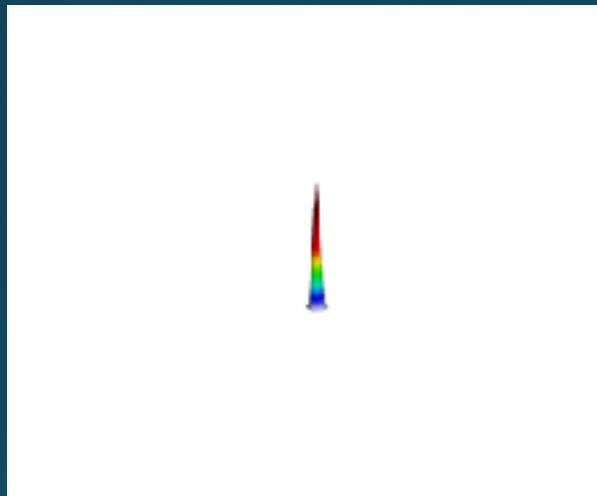


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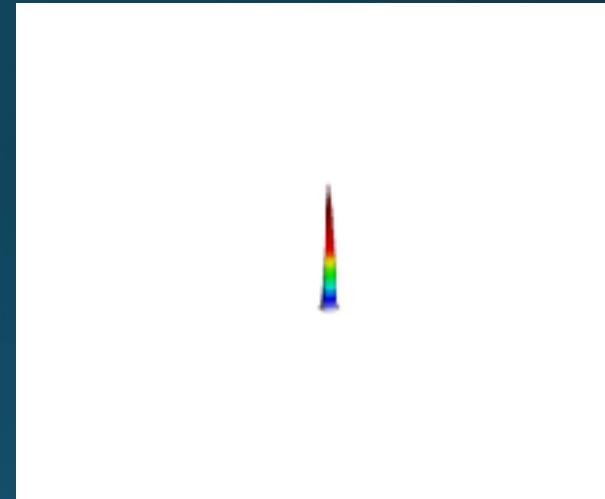
One Color Linear Driver



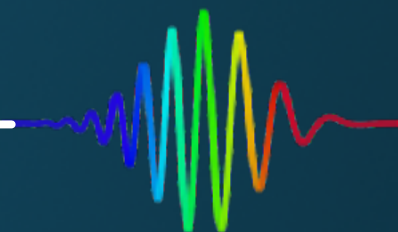
One-Color Circular Driver



Two-Color Counter-Rotating

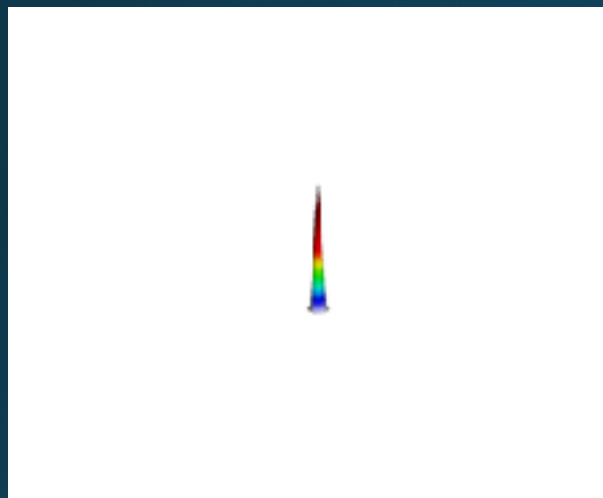


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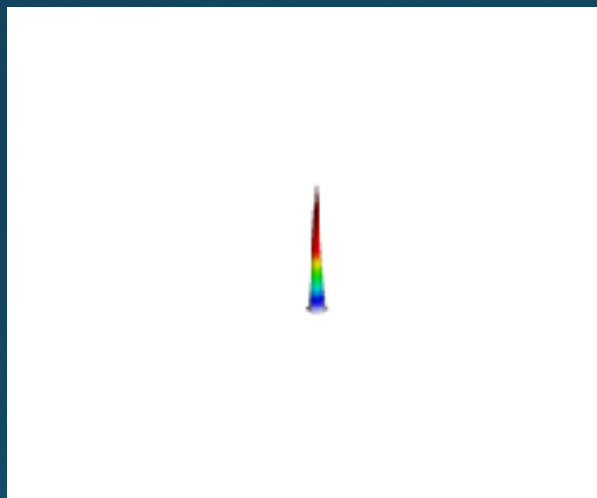


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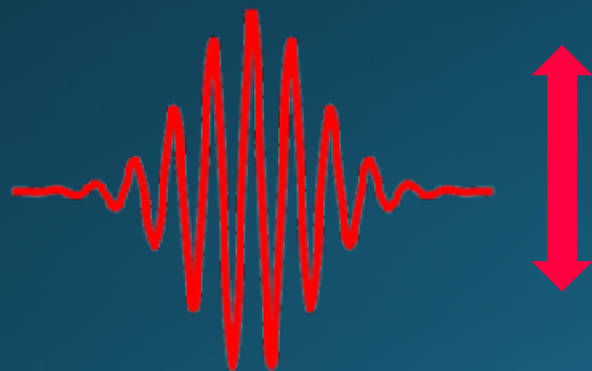
One Color Linear Driver



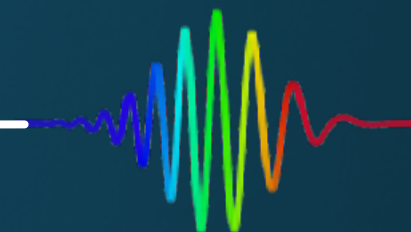
One-Color Circular Driver



Two-Color Counter-Rotating

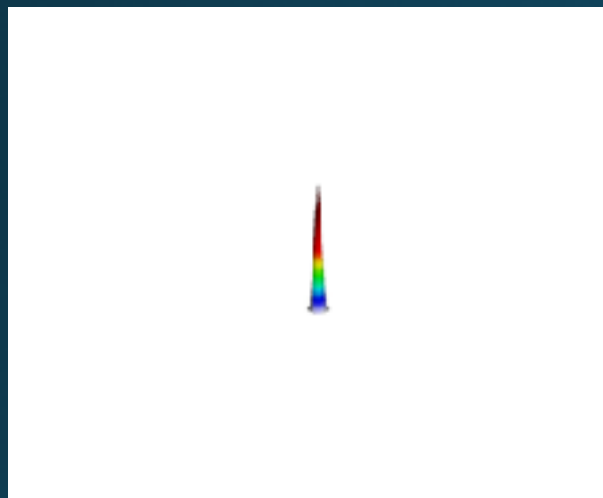


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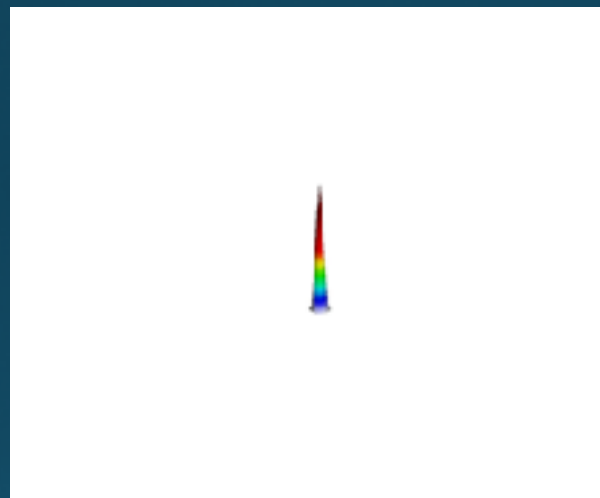


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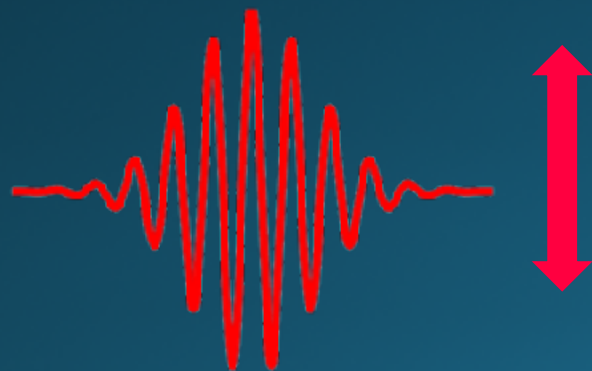
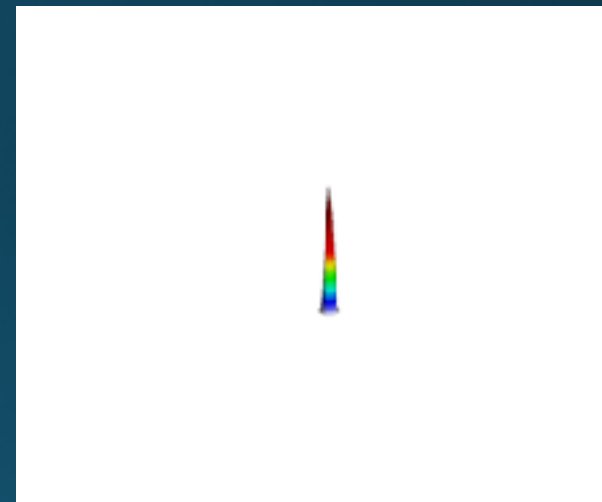
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One-Color Circular Driver

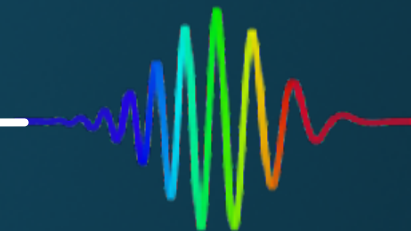


Two-Color Counter-Rotating





# HHG in a Whole New Dimension: Reshaping the Frontier of Attosecond Science

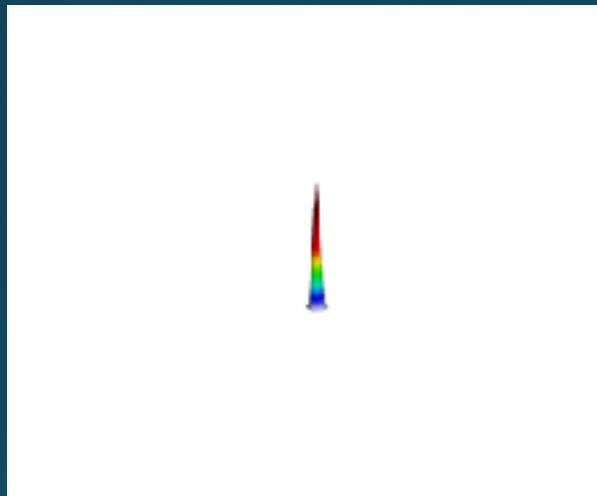


Videos: Courtesy of Carlos Hernandez-Garcia

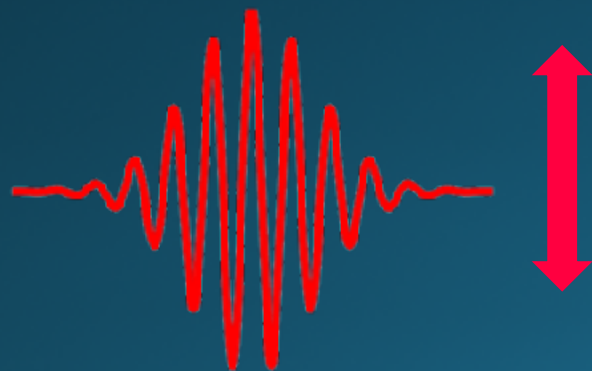
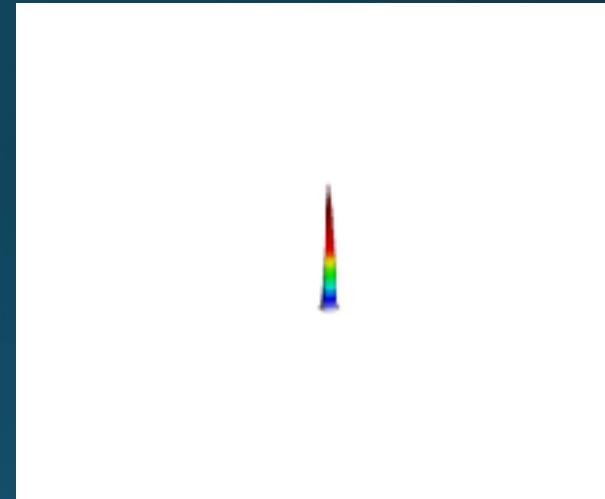
One Color Linear Driver



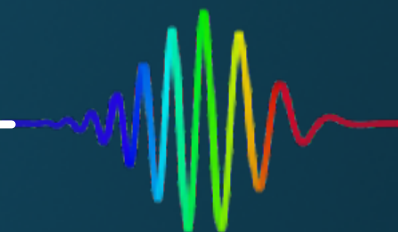
One-Color Circular Driver



Two-Color Counter-Rotating

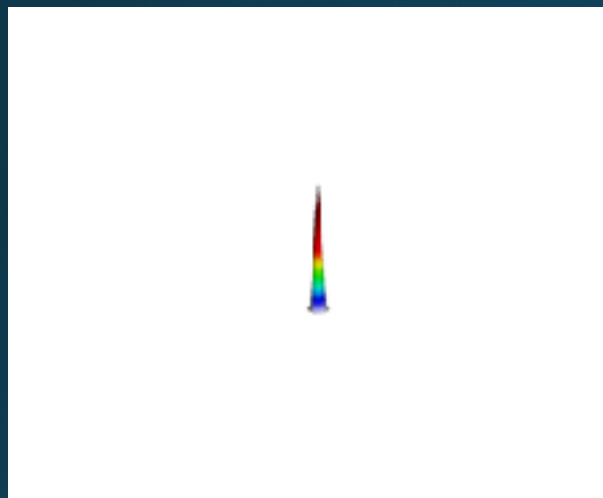


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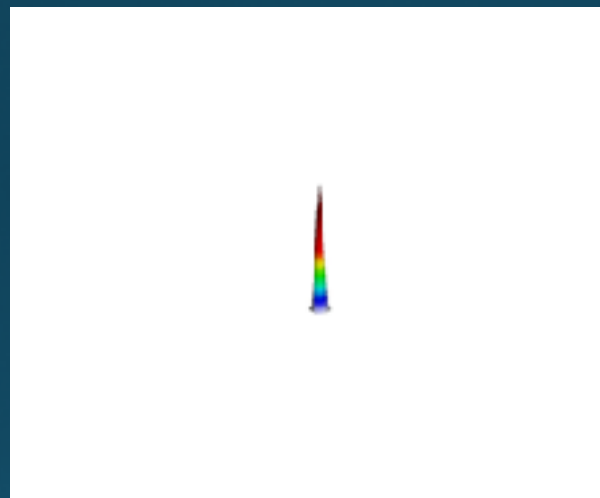


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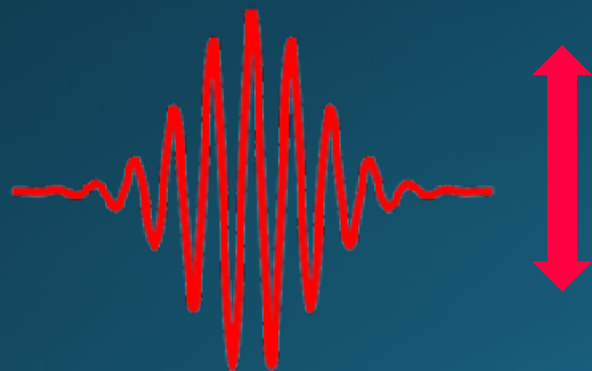
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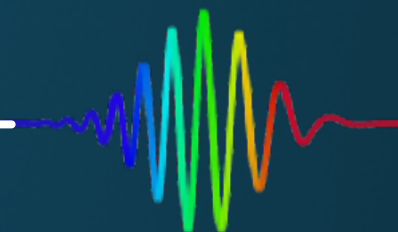
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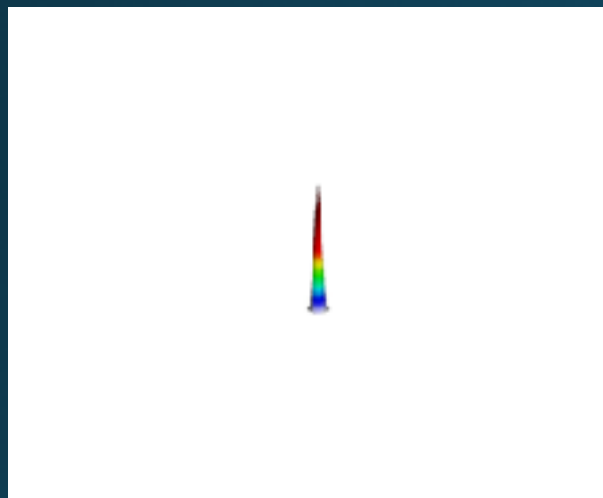


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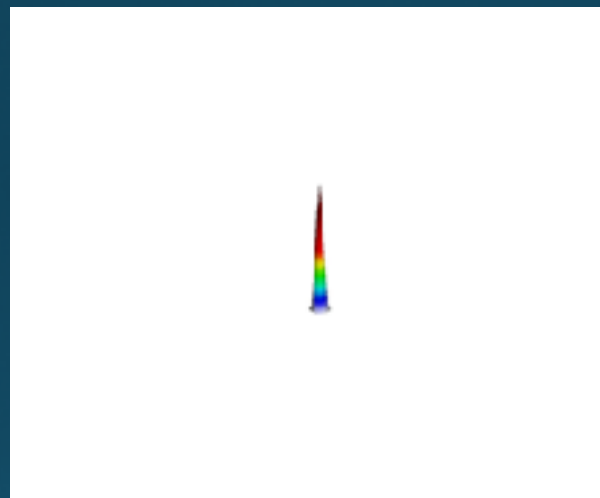


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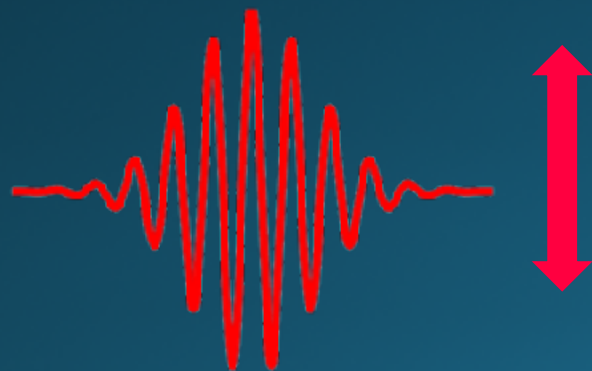
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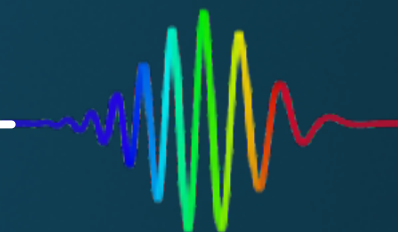


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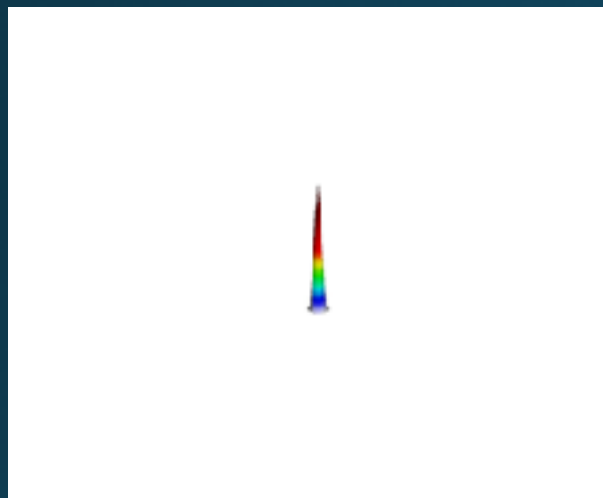


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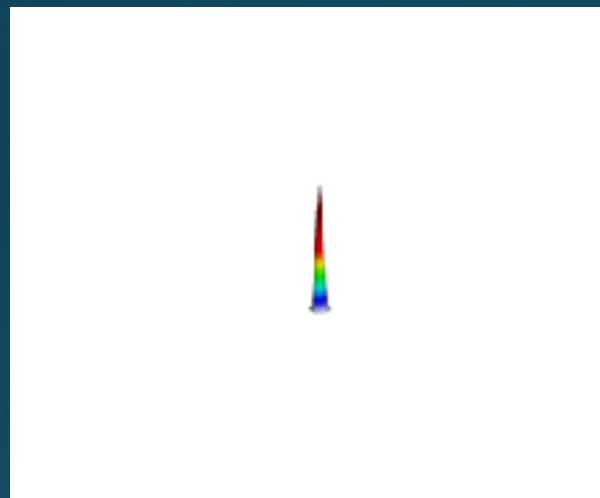


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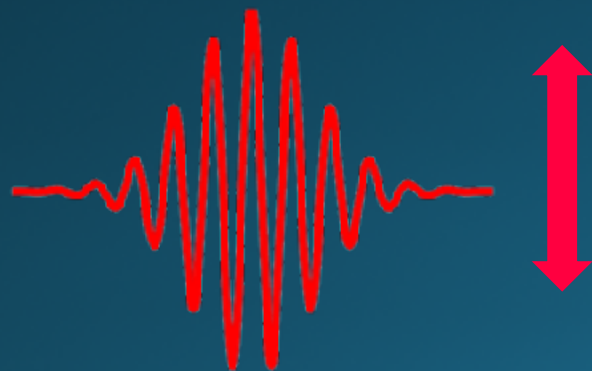
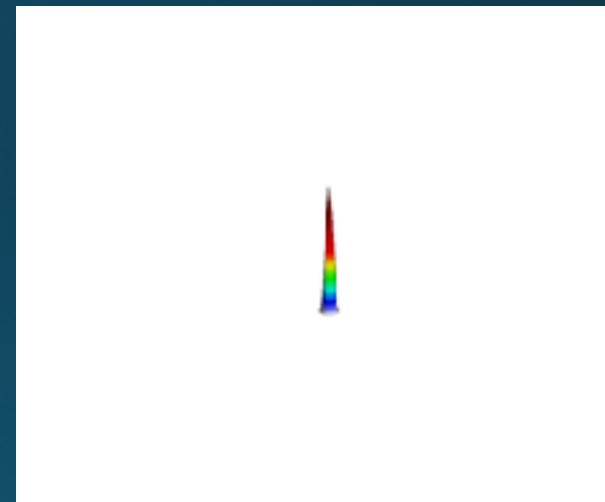
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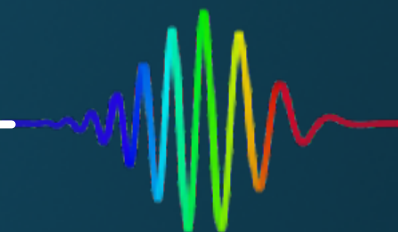
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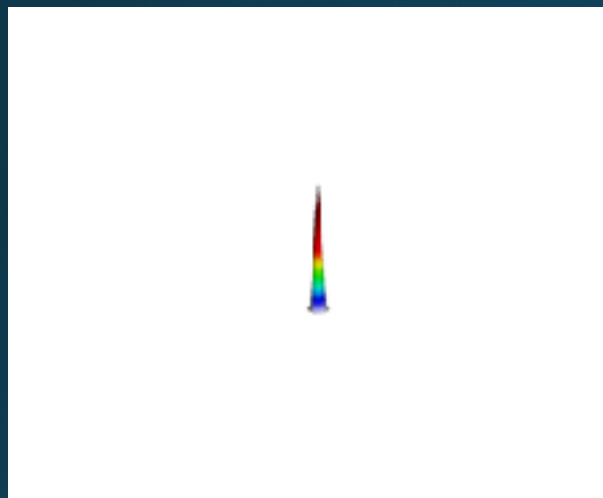


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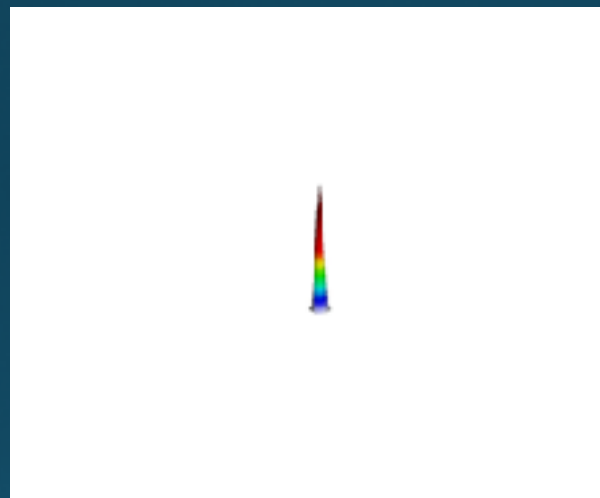


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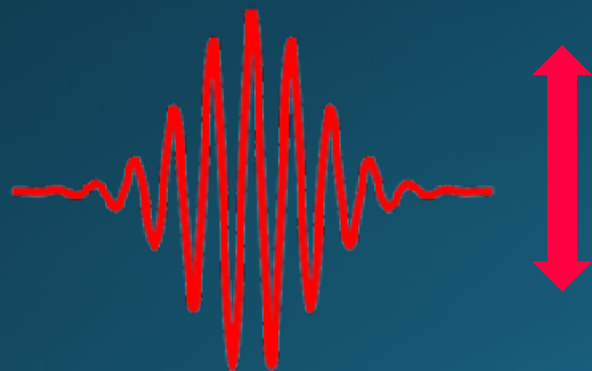
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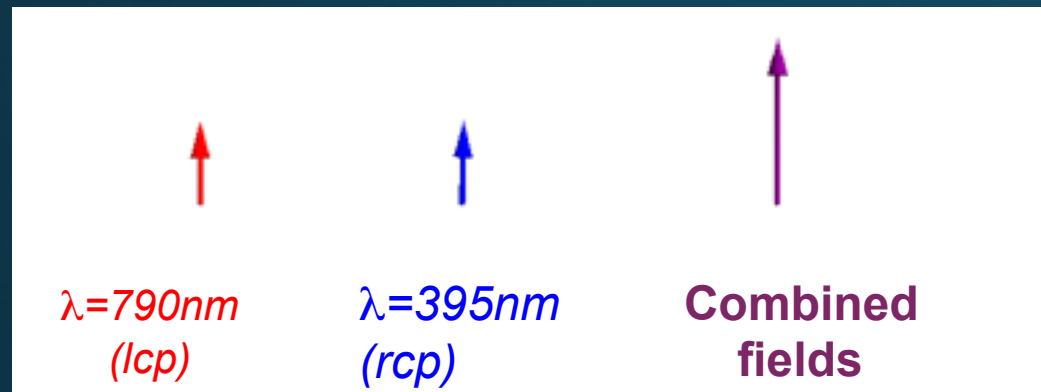
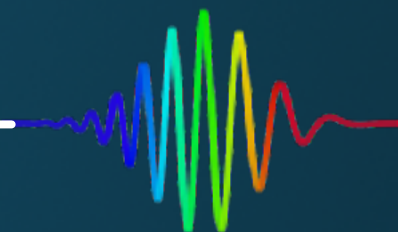


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# Circularly Polarized High Harmonic Generation (CPHHG): The Attosecond(s) From the Past!



Eichmann, Phys. Rev. A. 51, 1995

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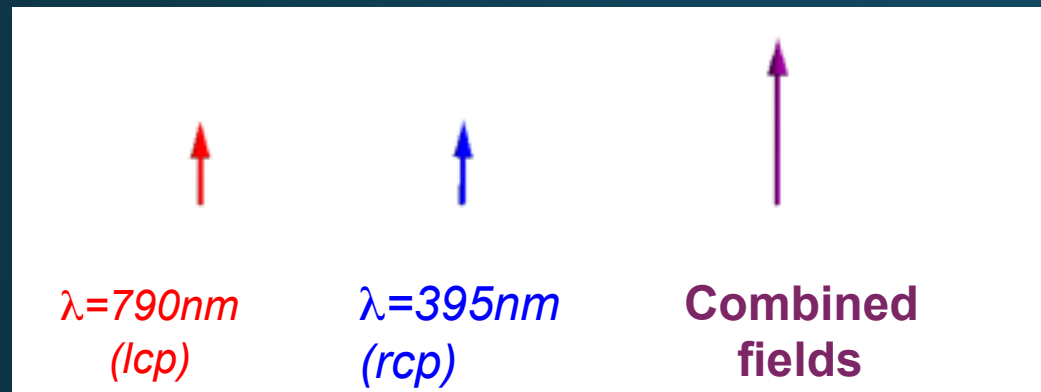
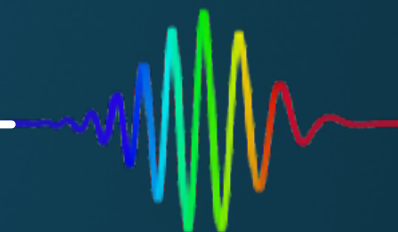
Dorney, PRL, 118, 2017

Many, many, more





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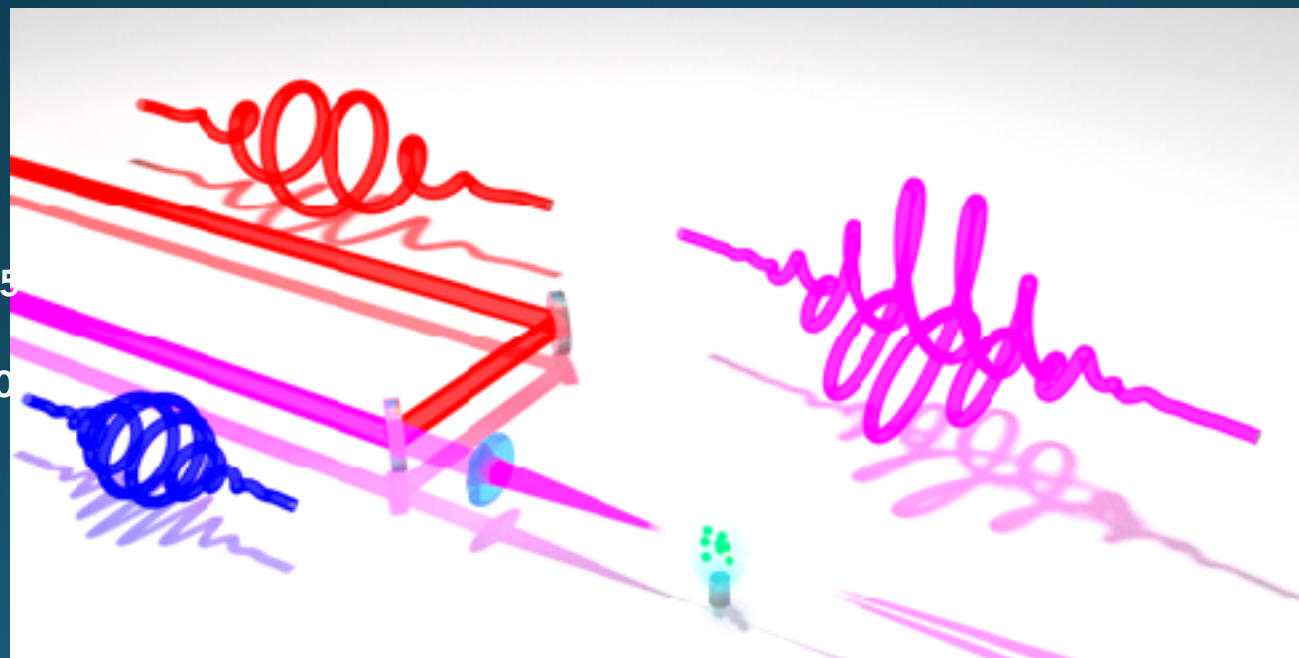
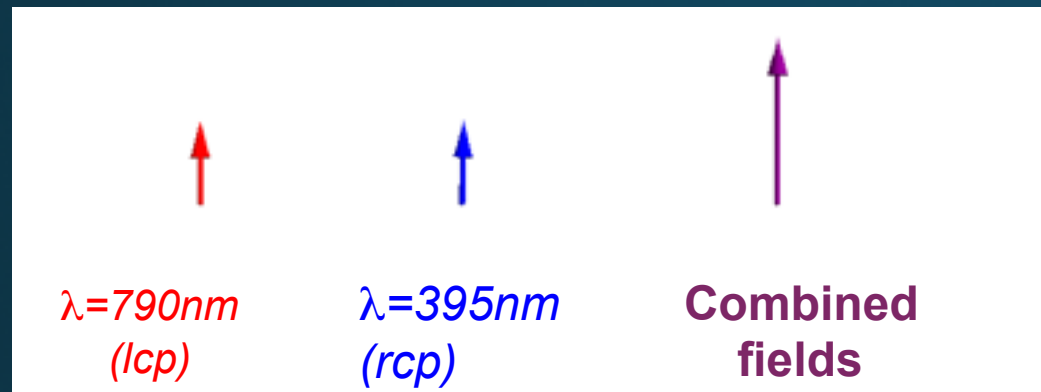
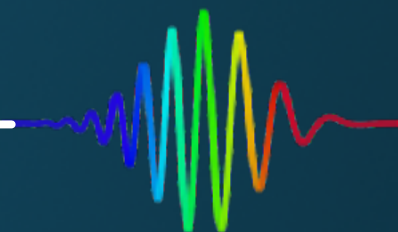
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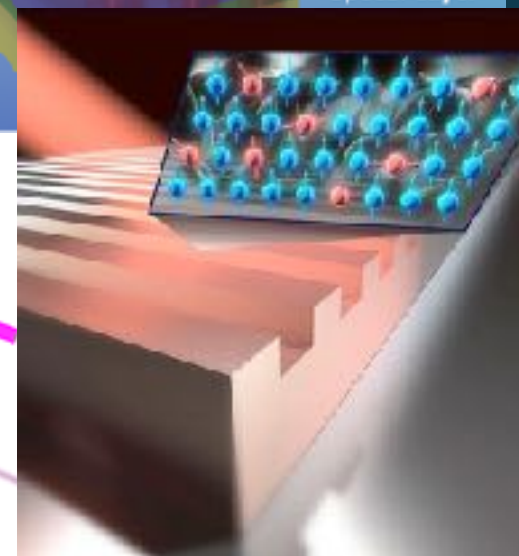
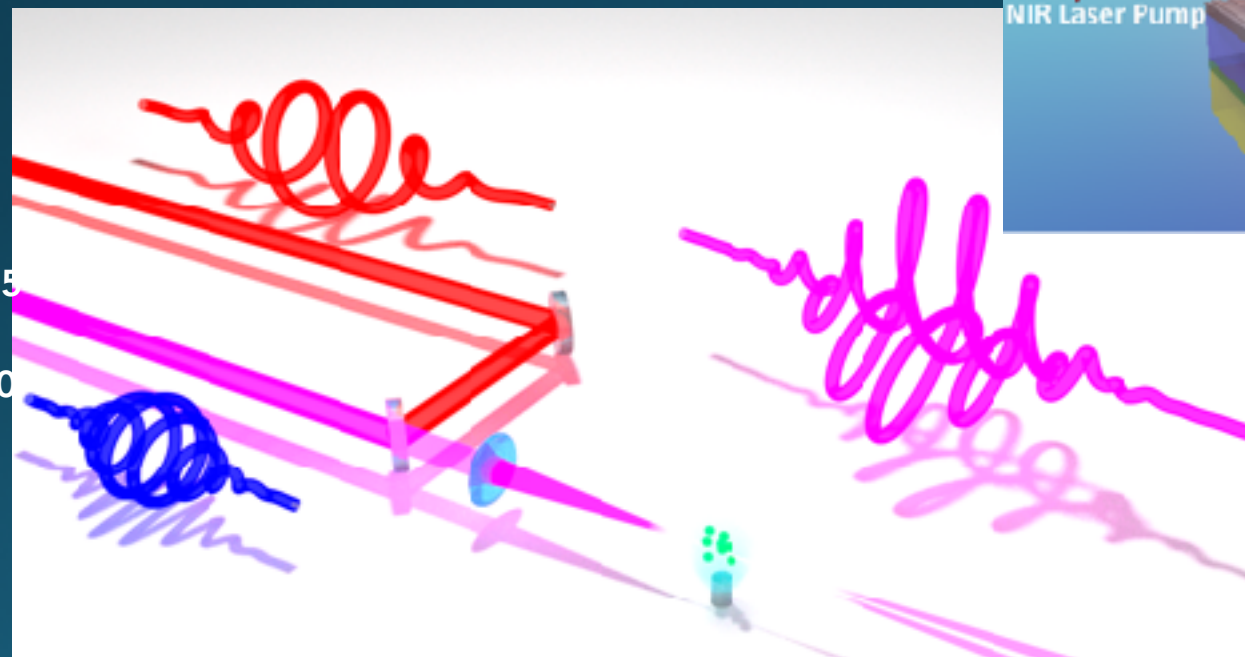
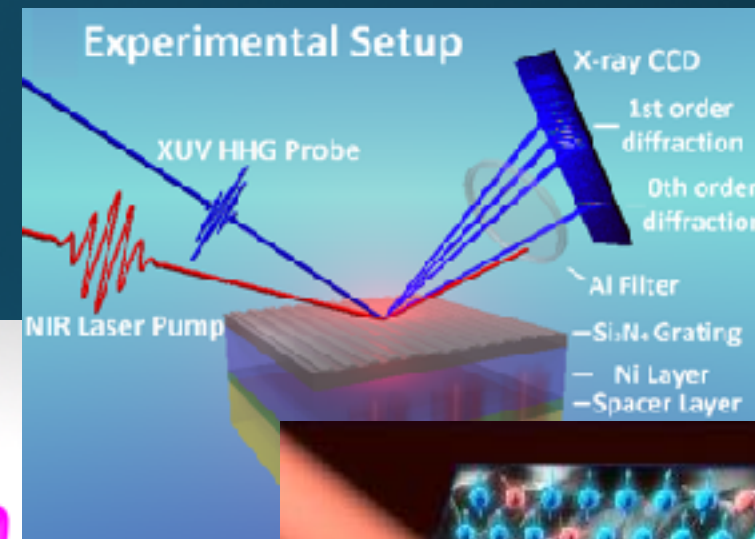
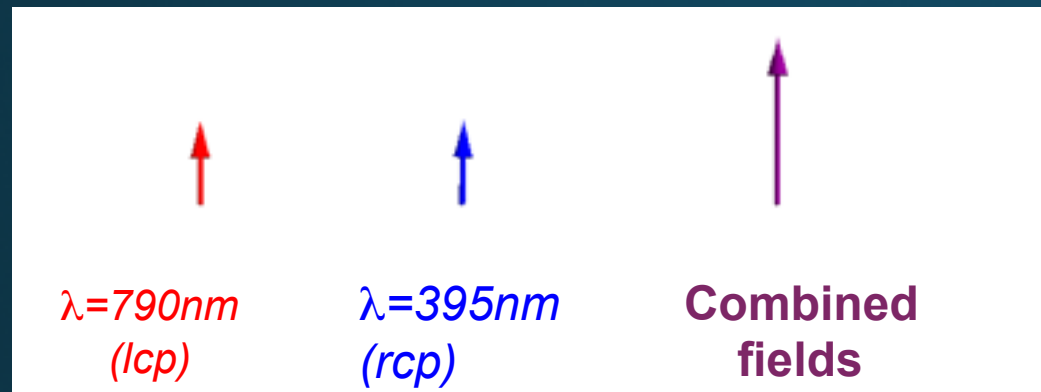
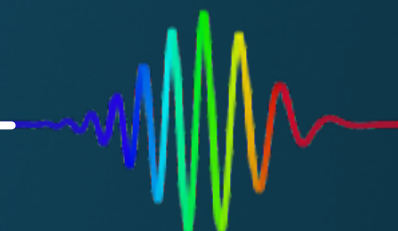
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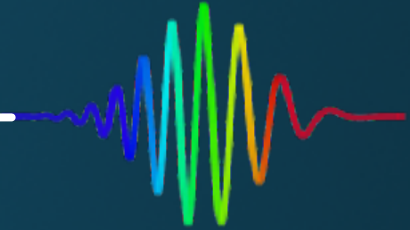


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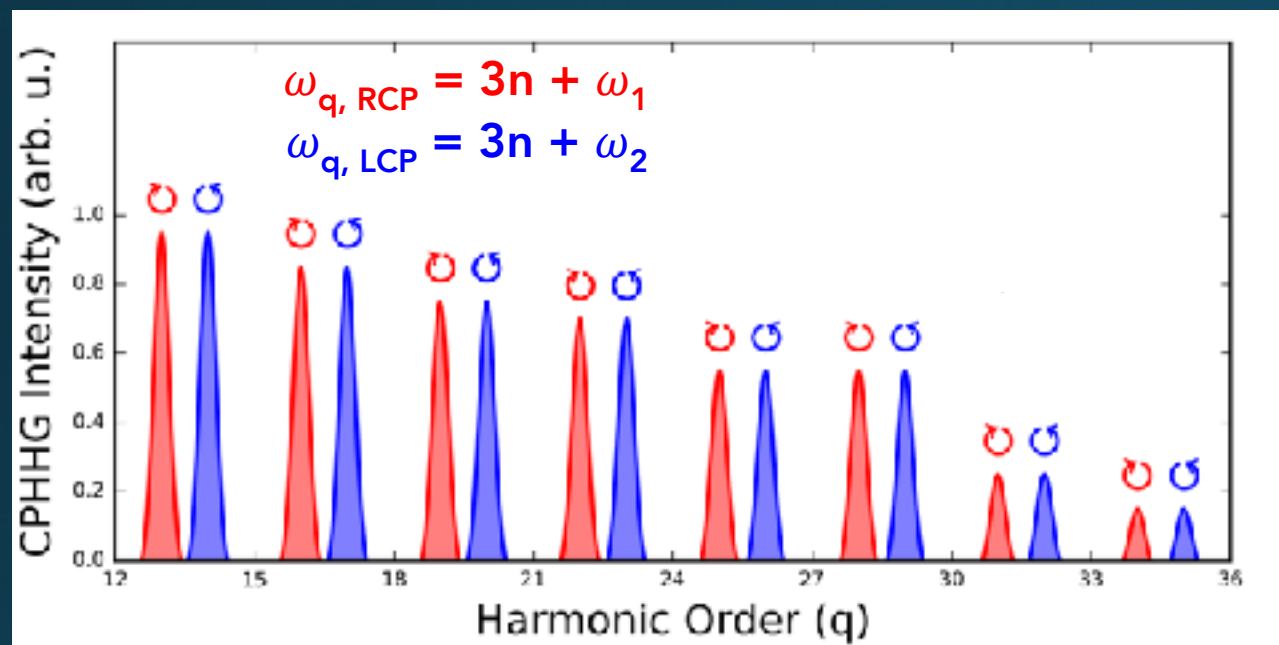
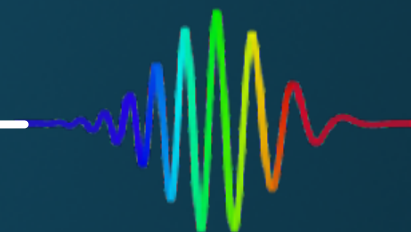


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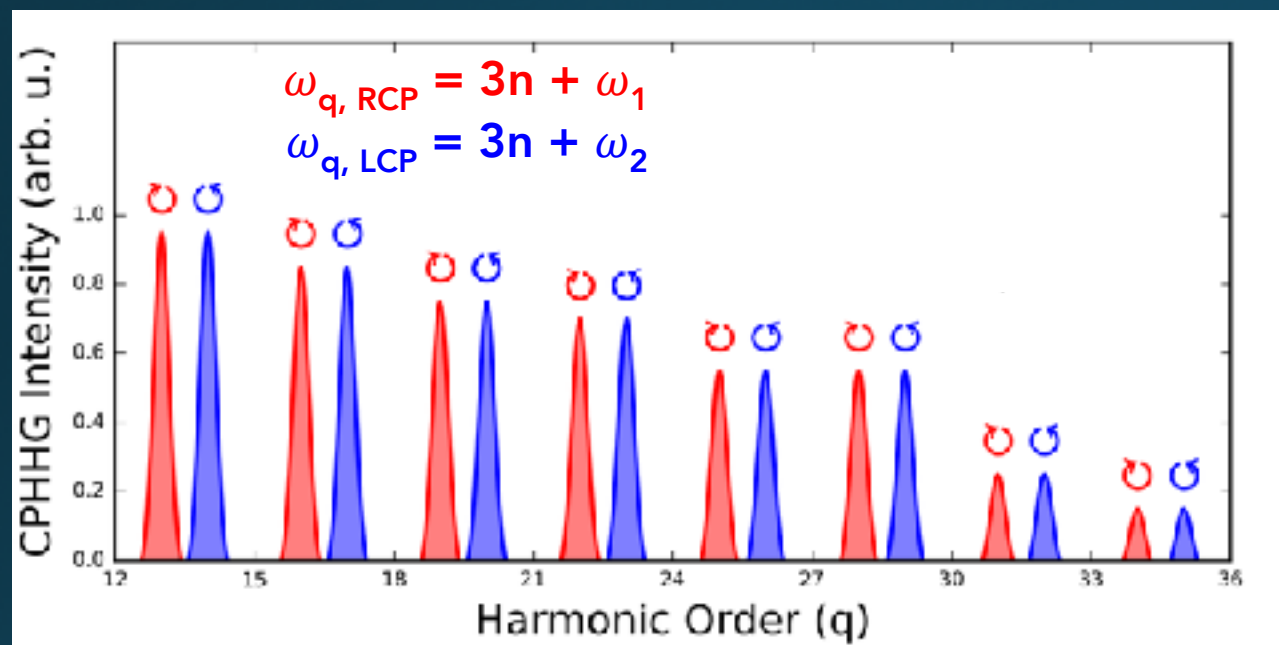
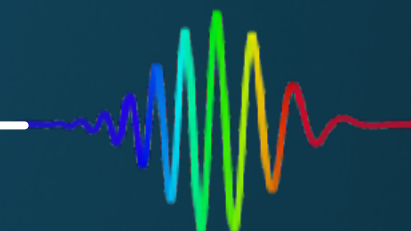
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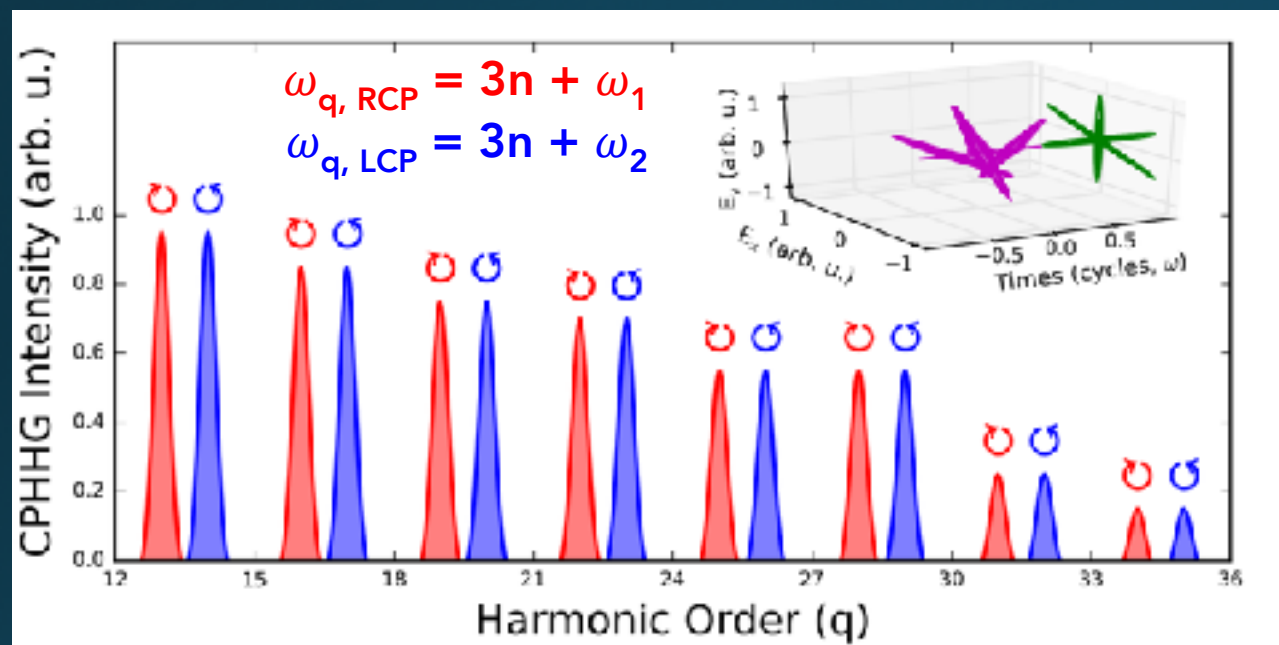
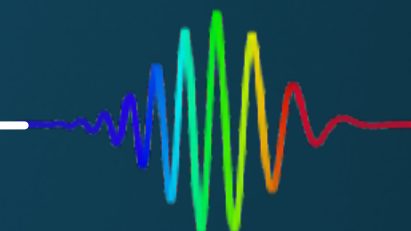
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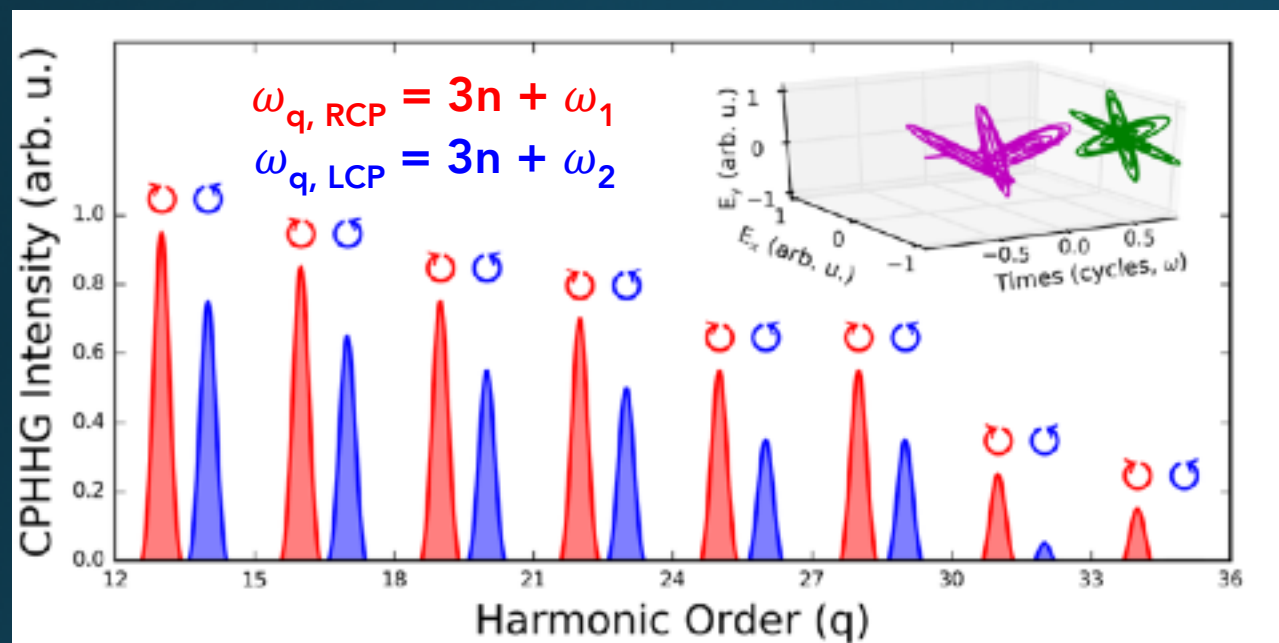
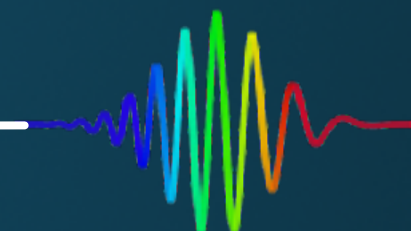
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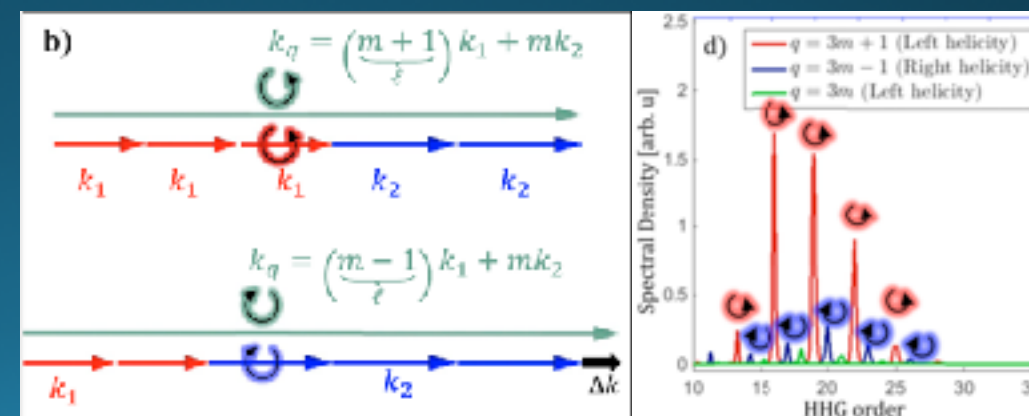
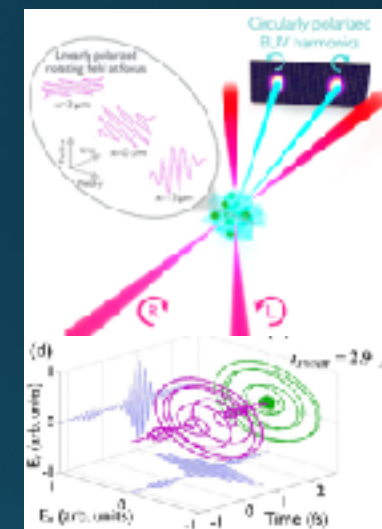
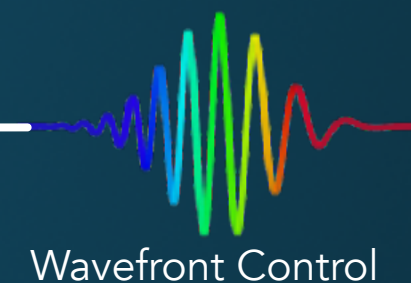
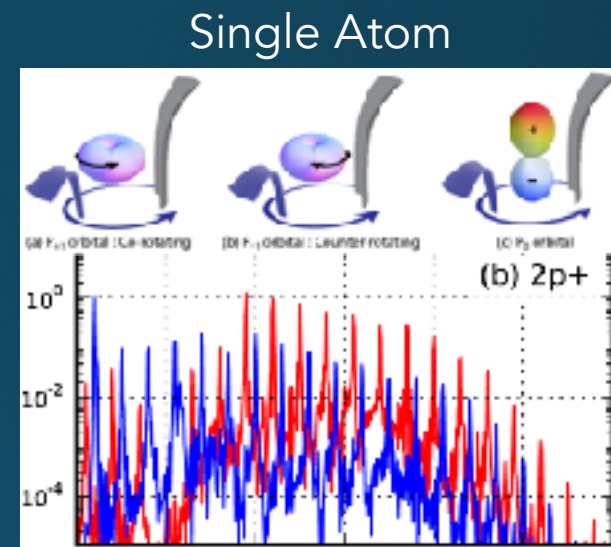
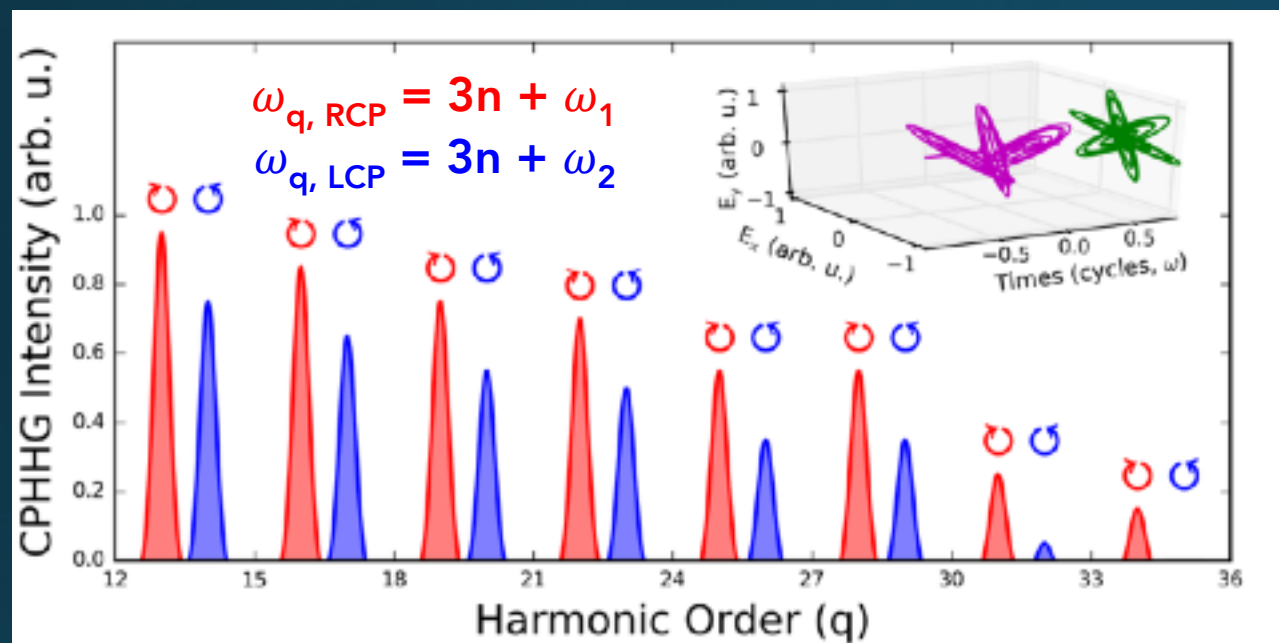
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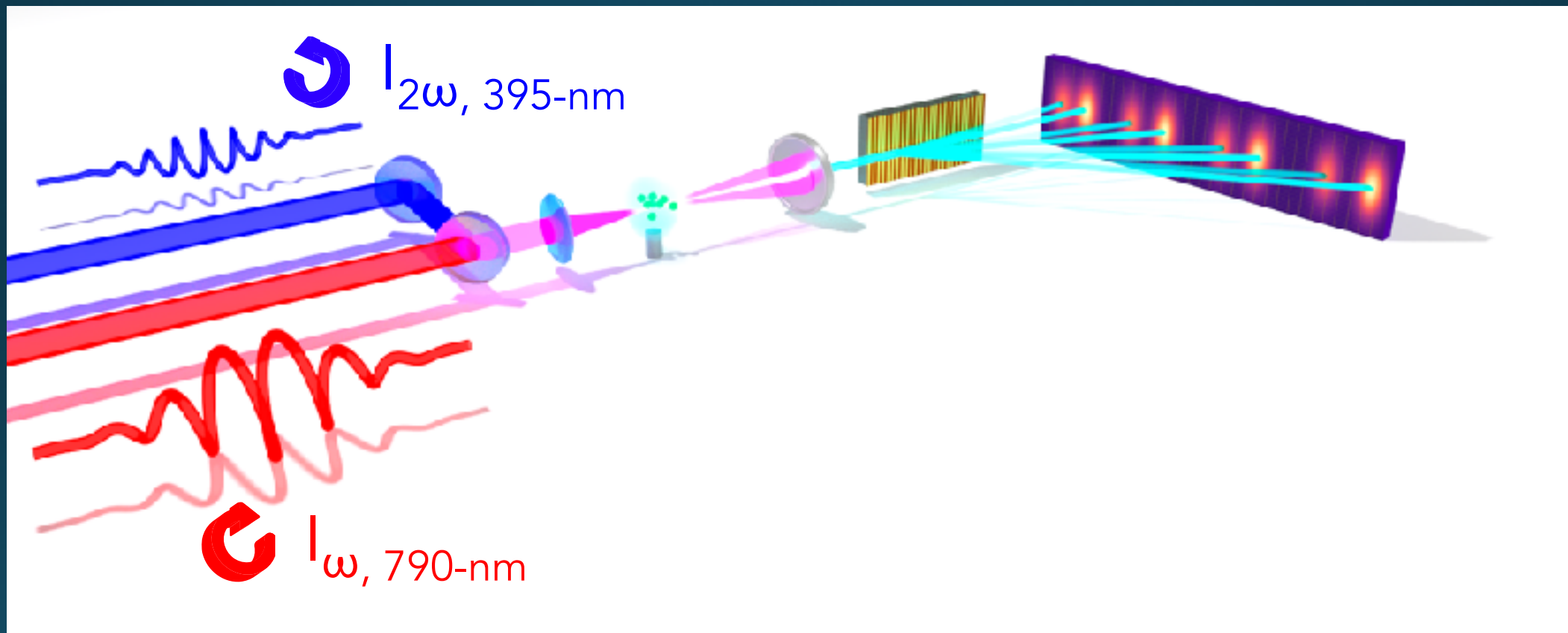
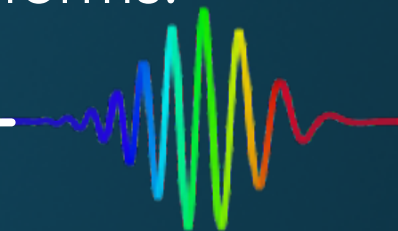


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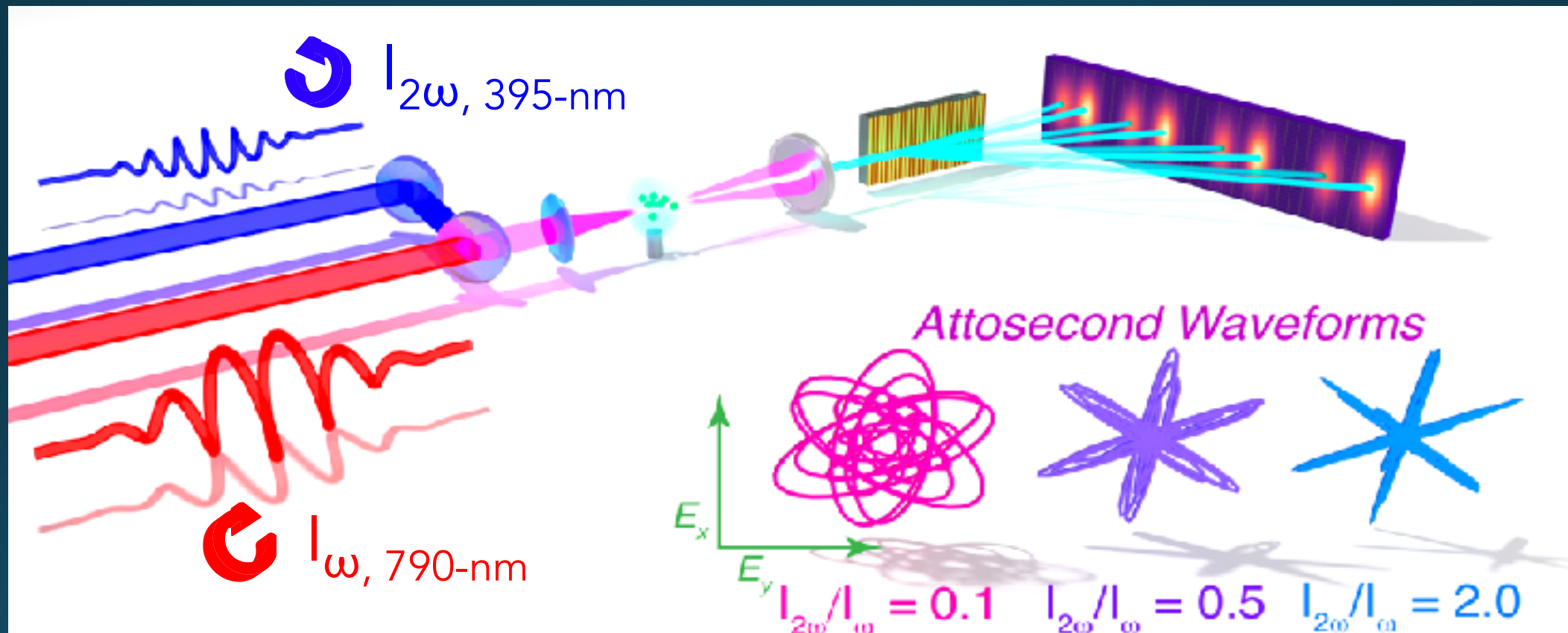
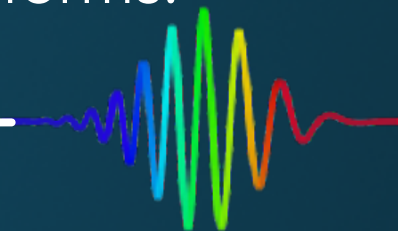
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# Active Control Over the Polarization of High-Harmonic Waveforms: Production of Elliptically Polarized Attosecond Pulse Trains!



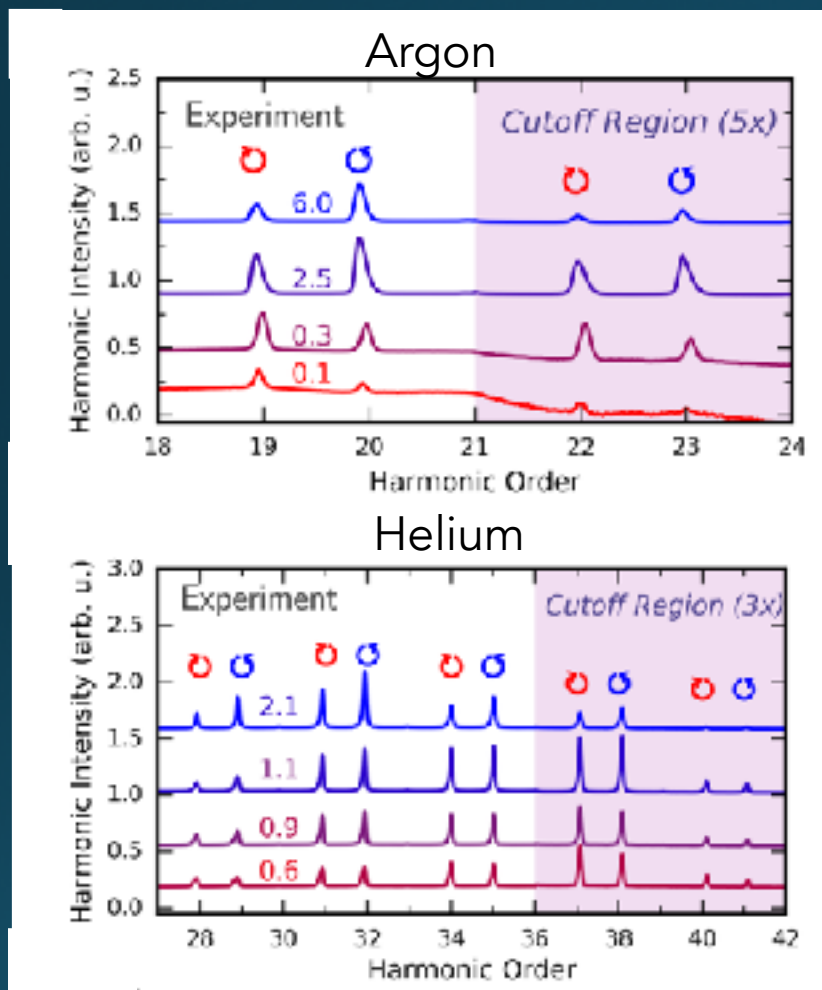
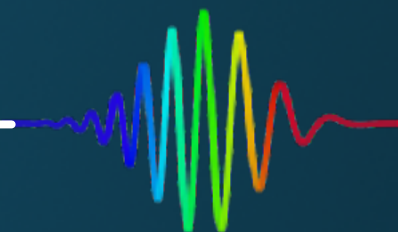
Dorney et. al., Phys. Rev. Lett., 118, 2017

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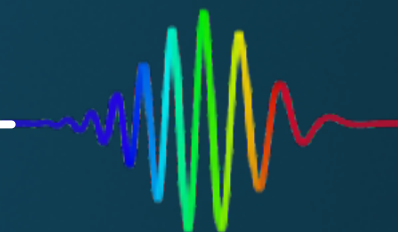
# Controlling the Driving Waveform for CPHHG: Active Control over Spectral Chirality



Dorney et. al., Phys. Rev. Lett., 118, 2017

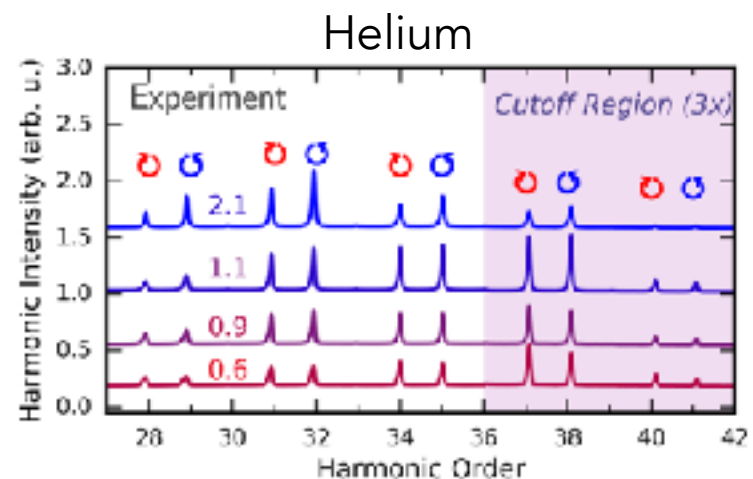
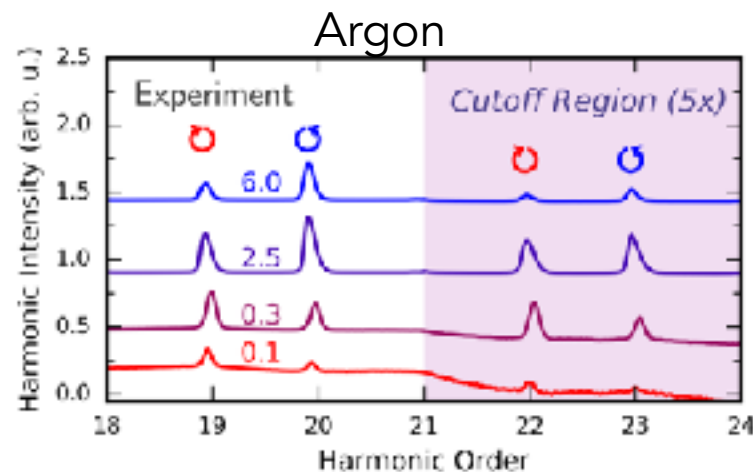
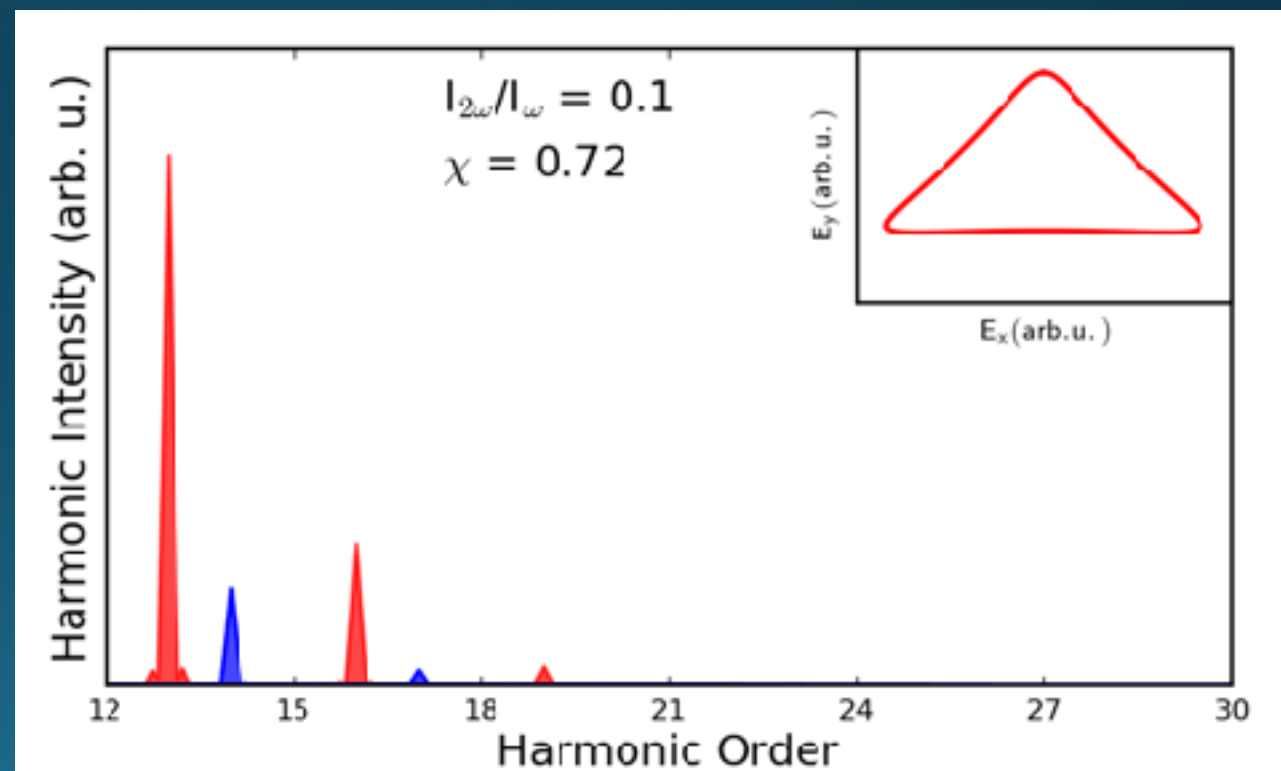


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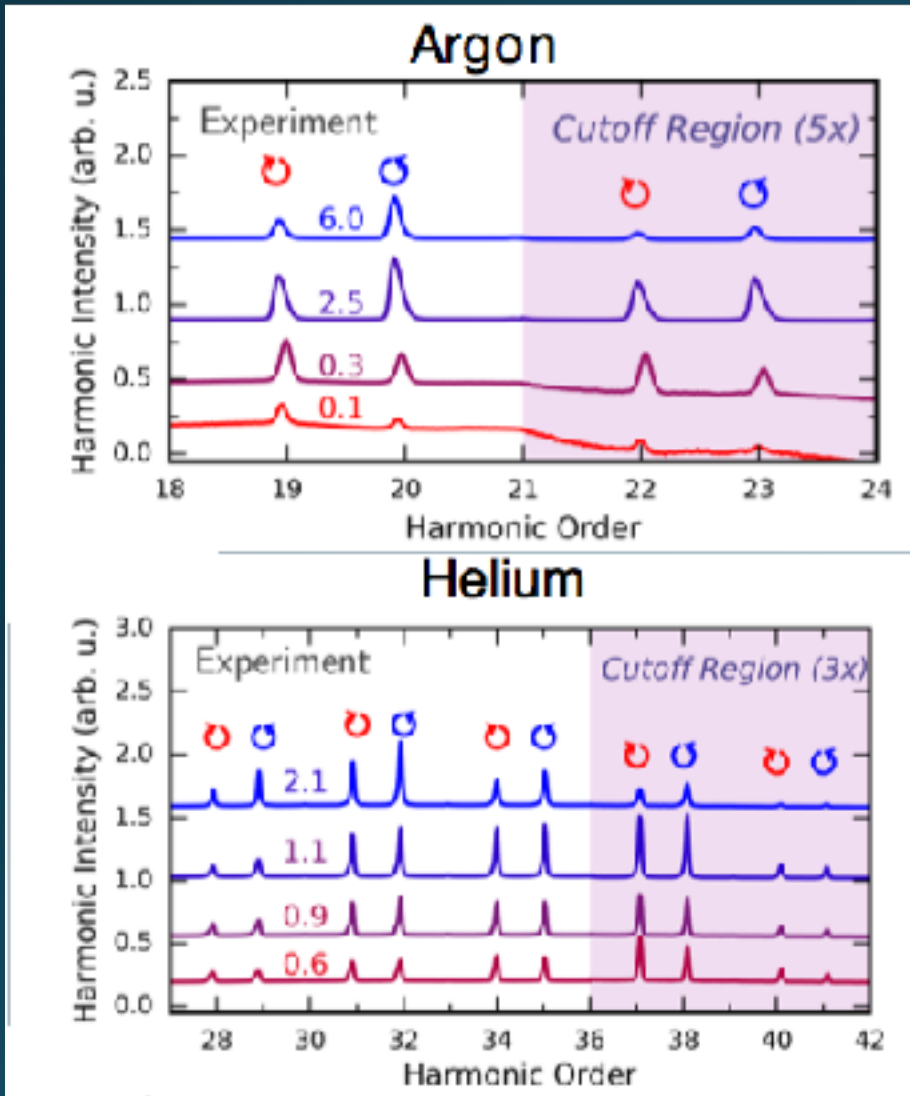
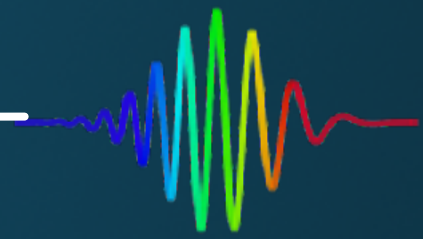


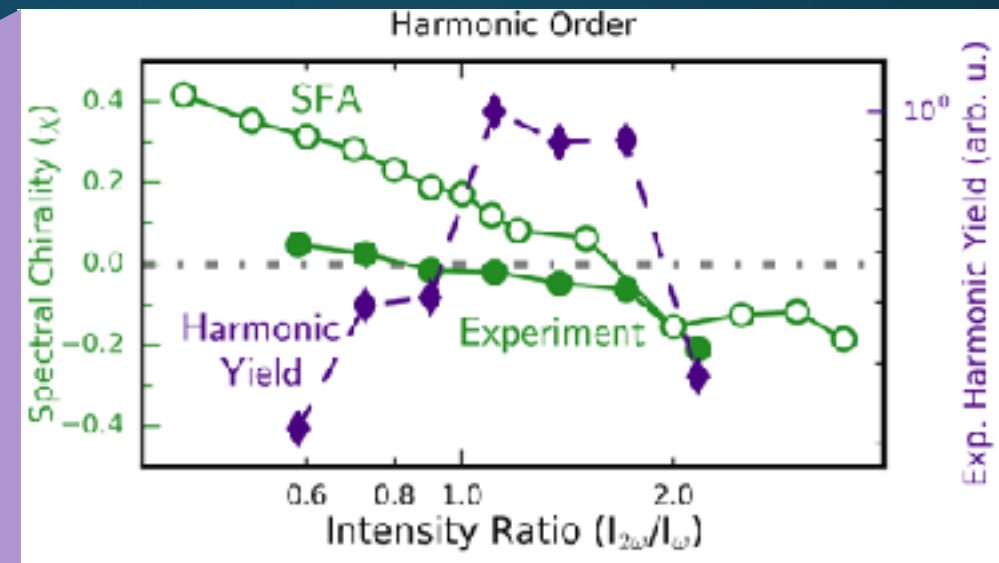
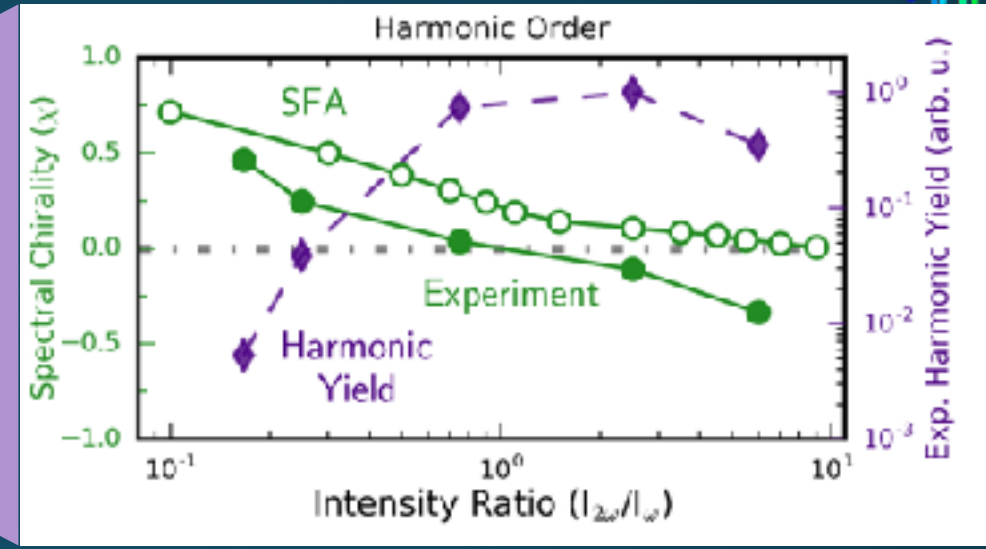
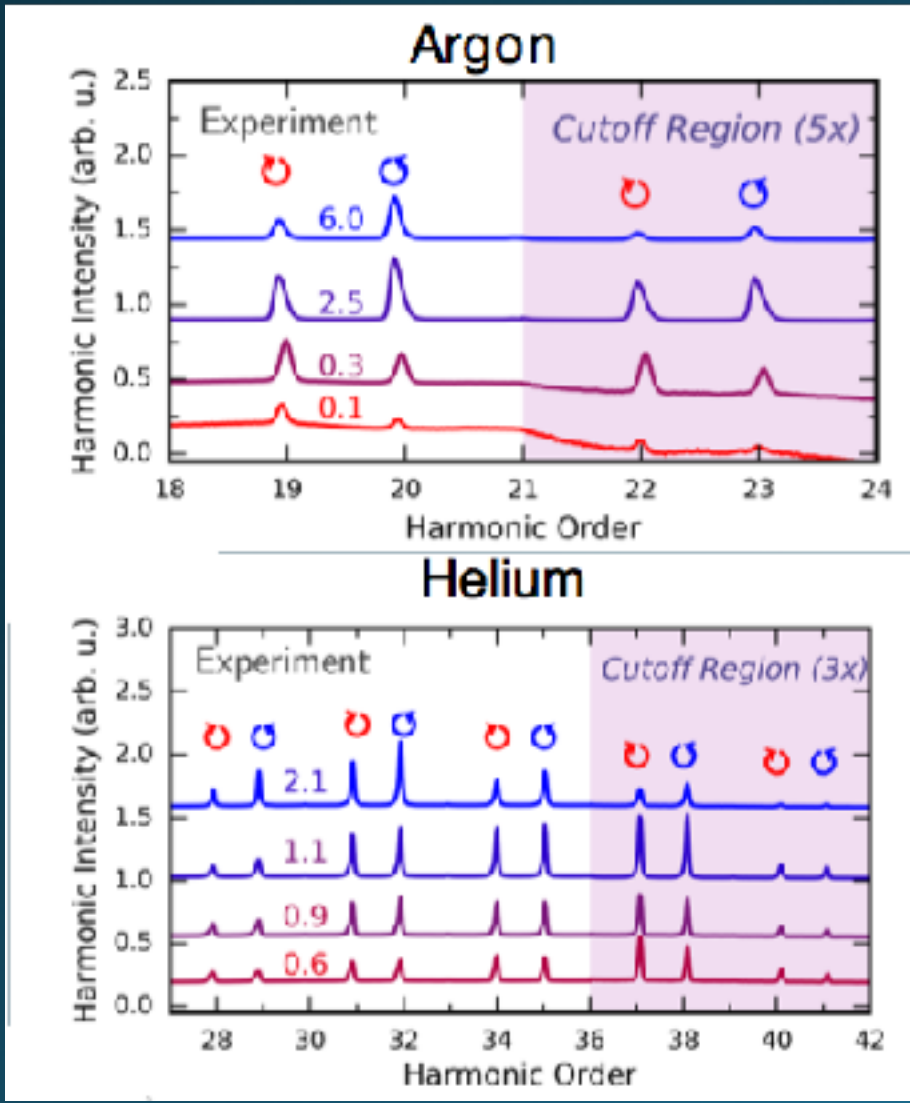
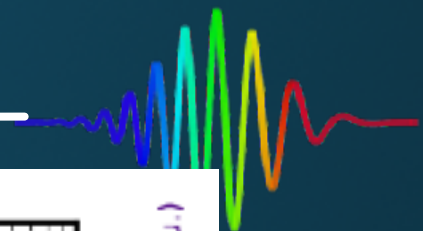
$$\chi = \frac{(I_{RCP} - I_{LCP})}{(I_{RCP} + I_{LCP})}$$

## SFA Simulation in Ar

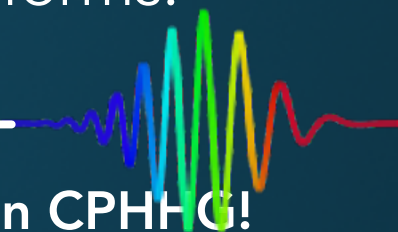


Dorney et. al., Phys. Rev. Lett., 118, 2017

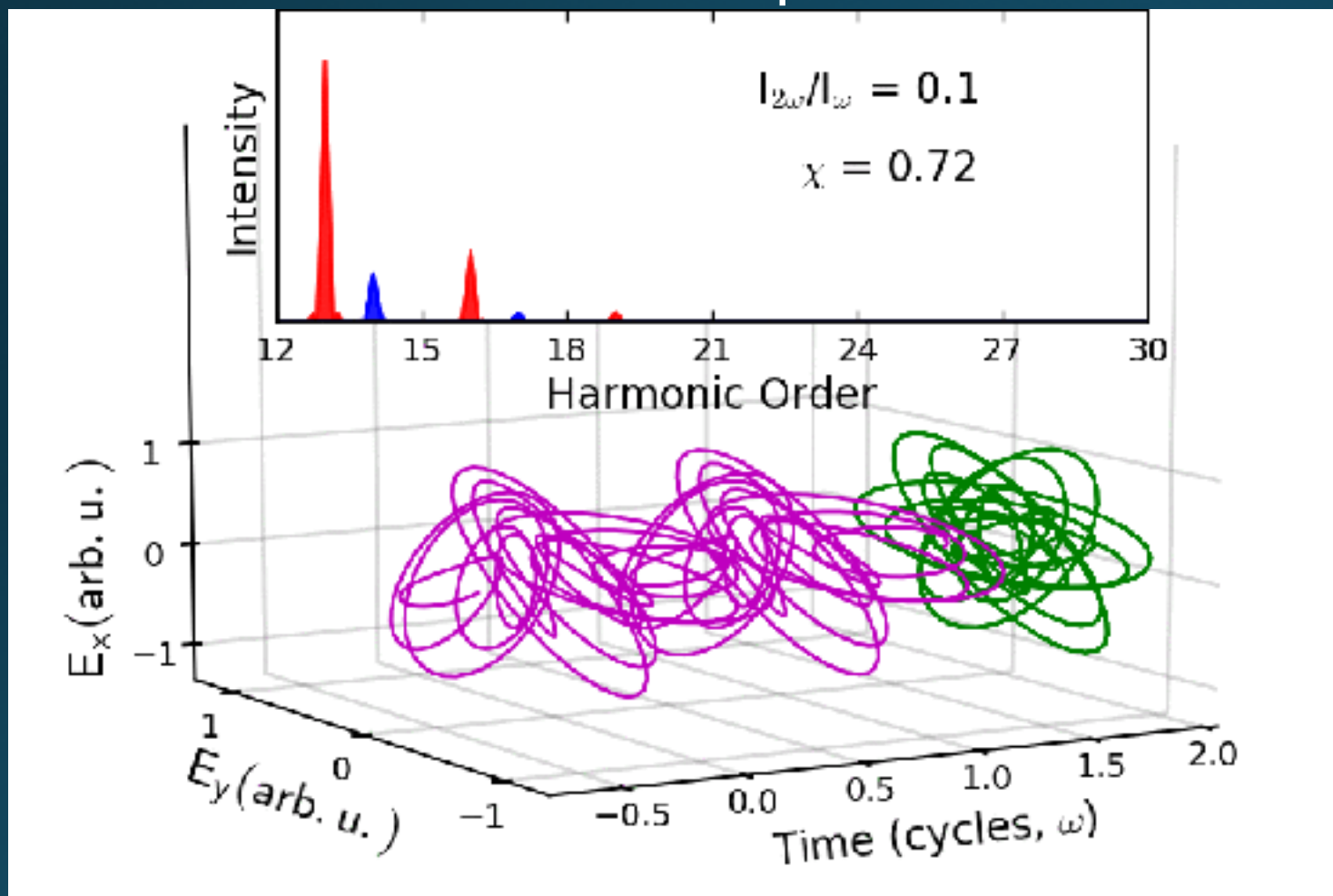




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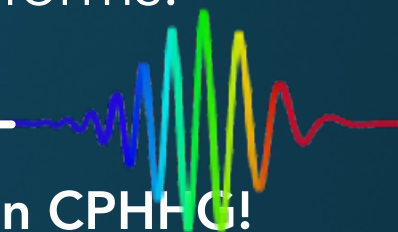


- First demonstration of real-time polarization control of attosecond pulse trains in CPHHG!

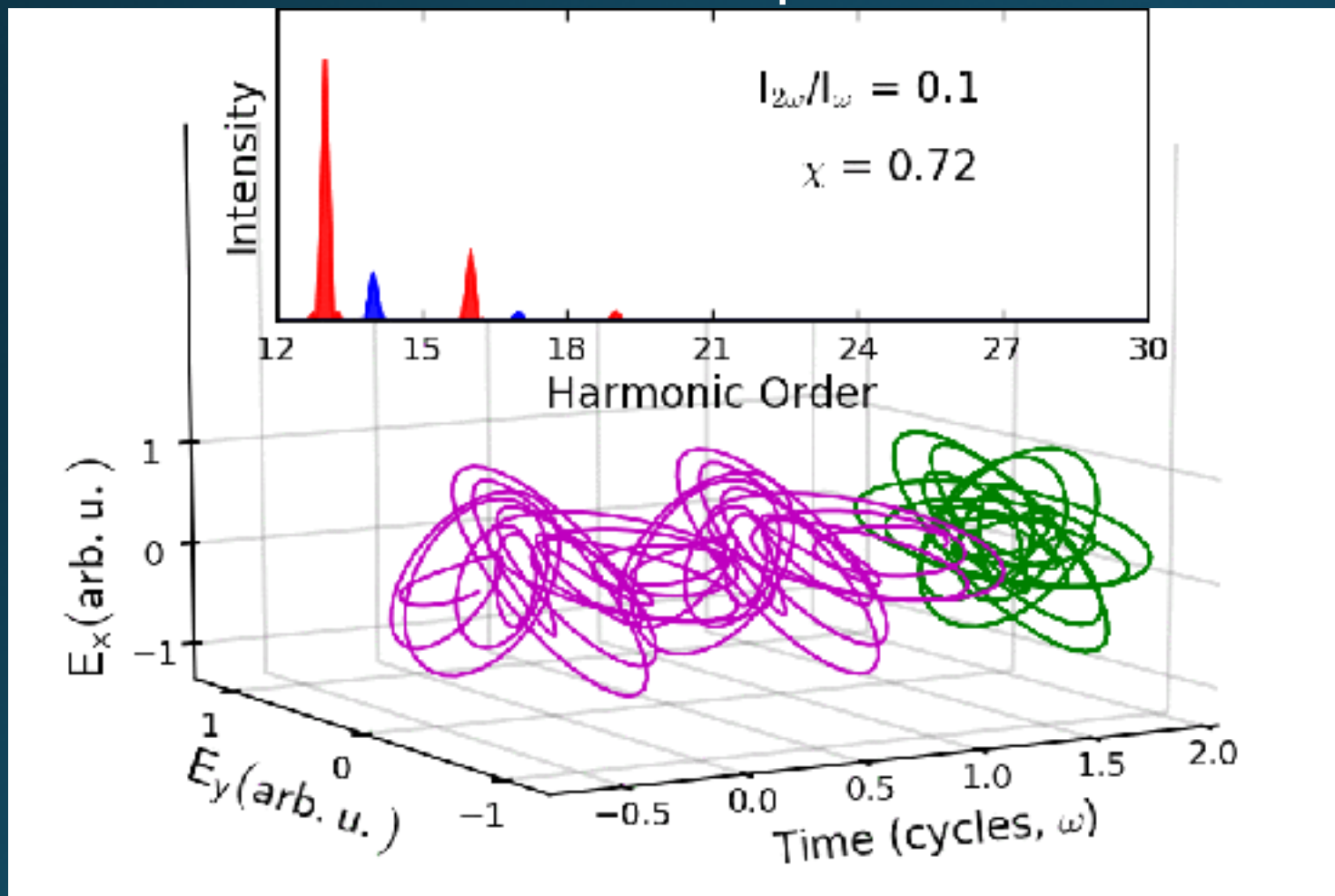




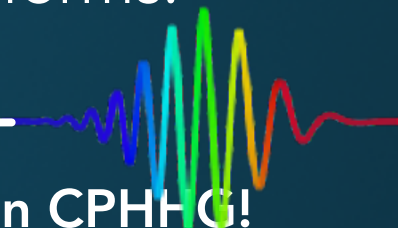
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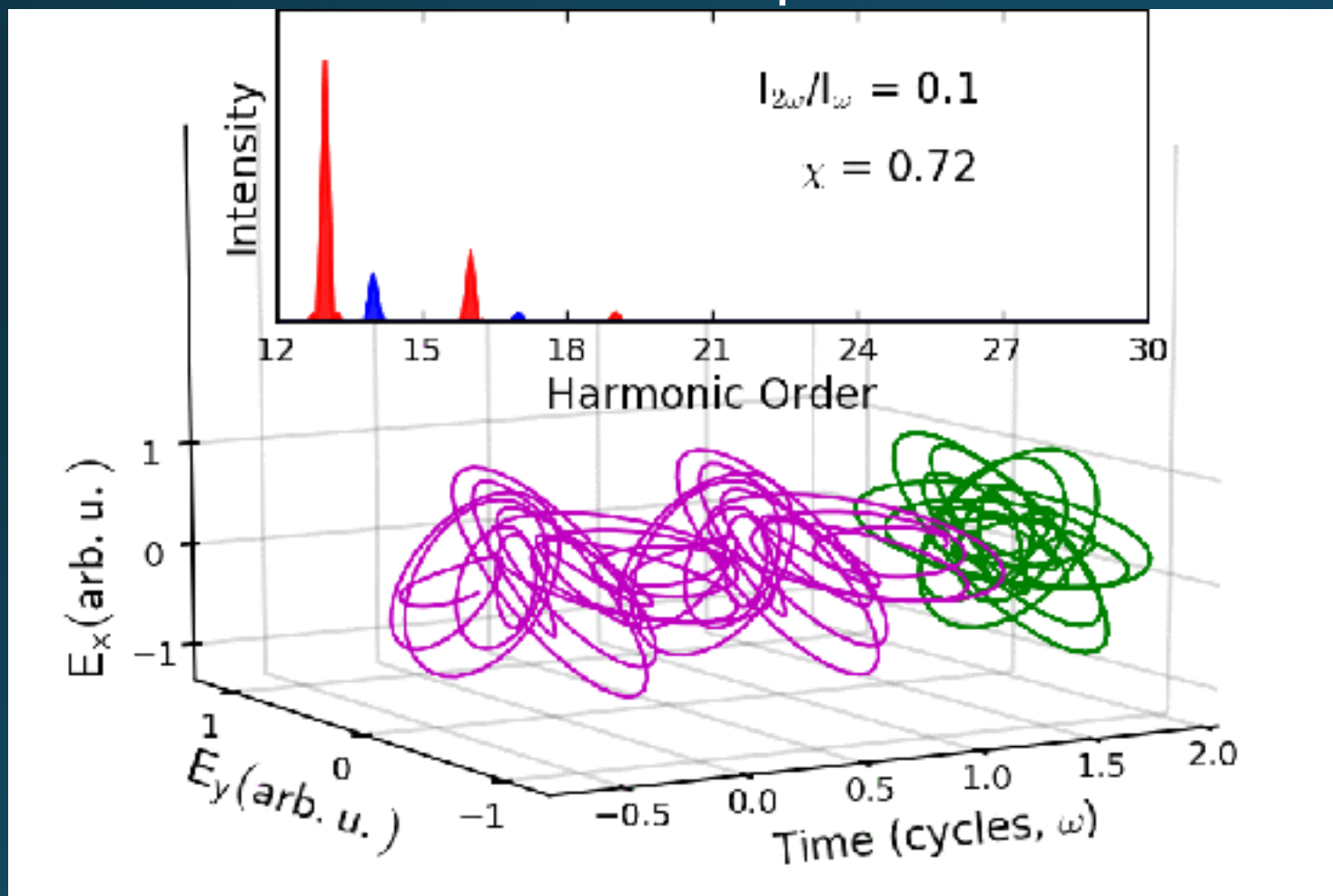
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Neufeild, et al., arXiv:1709.06261, 2017

Instantaneous Optical Chirality

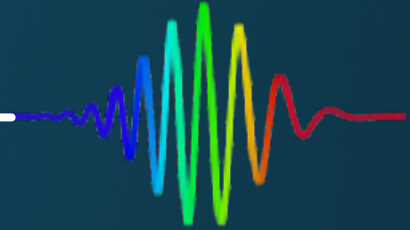
$$C_P = \frac{1}{c_0^2} \dot{\phi}(t) I(t)$$

Non-Instantaneous Optical Chirality

$$\chi(\Delta t) = \frac{1}{c_0^2} \frac{\phi(t + \Delta t) - \phi(t)}{\Delta t} \bar{I}(t, \Delta t)$$



# Simple Interpretation of Elliptical Control in CPHHG: Perturbative-(ish) Photon Absorption Model



$$I_q \propto P(\Omega) \propto \sum_{i=0}^{\infty} p_1^{|n_1^i|} p_2^{|n_2^i|}$$

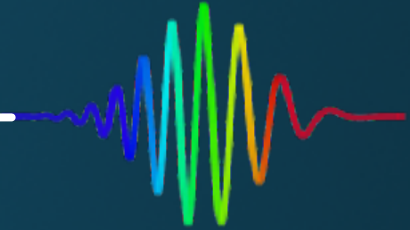
Dorney et al., Phys. Rev. Lett., 118, 2017

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Li, et al., arXiv:1702.04084, 2017



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$$I_q \propto P(\Omega) \propto \sum_{i=0}^{\infty} p_1^{|n_1^i|} p_2^{|n_2^i|} \quad p_{\omega} = \frac{I_{\omega}}{I_{\omega} + I_{2\omega}} = \frac{1}{1 + I_{ratio}} \quad p_{2\omega} = \frac{I_{2\omega}}{I_{\omega} + I_{2\omega}} = \frac{1}{1 + 1/I_{ratio}}$$

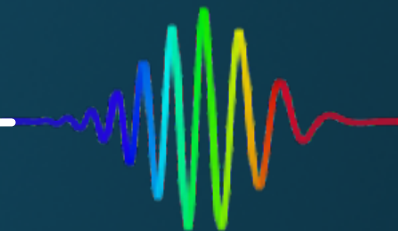
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n = channel number

s = number of "emitted" photons

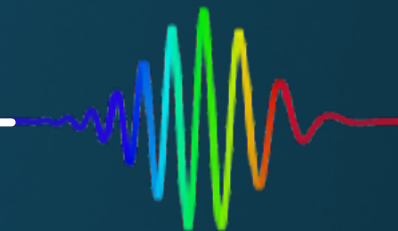
Channel	$H_{19, RCP} (7\omega_1 + 6\omega_2)$	
(n, s)	Total Photons	Statistical Scaling
(6, 0)	$7\omega_1 + 6\omega_2$	$p_{1\omega}$
(6, 1)	$7\omega_1 + 8\omega_2$	$p_{2\omega}$
(6, 2)	$9\omega_1 + 10\omega_2$	$p_{2\omega}$
(6, 3)	$11\omega_1 + 12\omega_2$	$p_{2\omega}$

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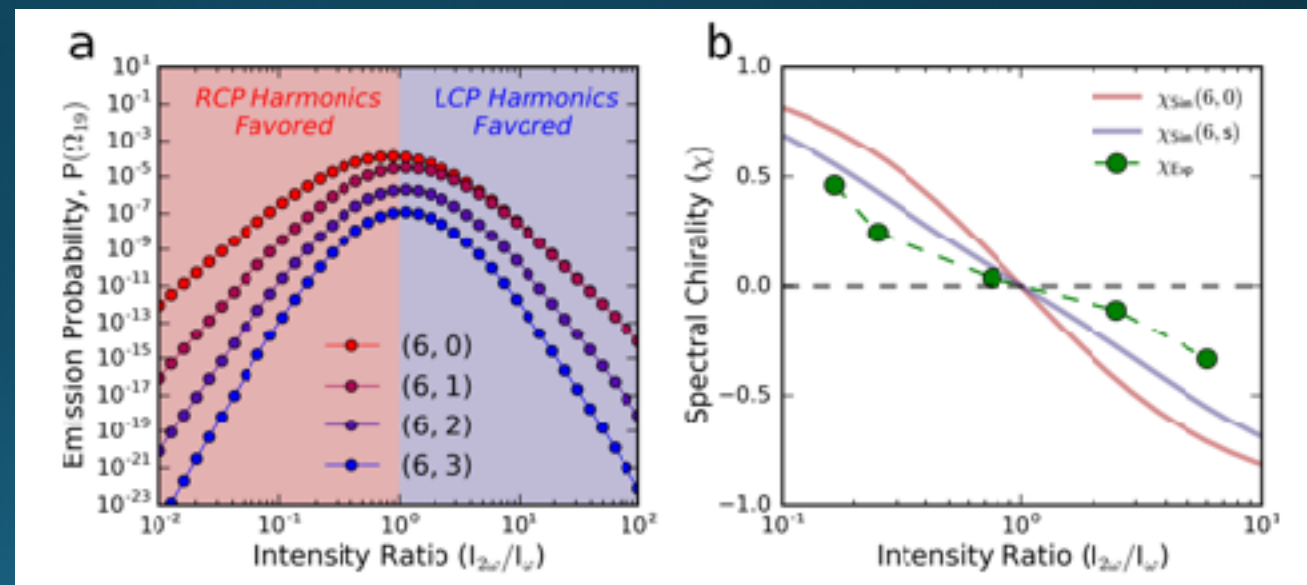


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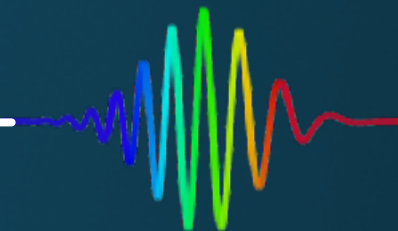
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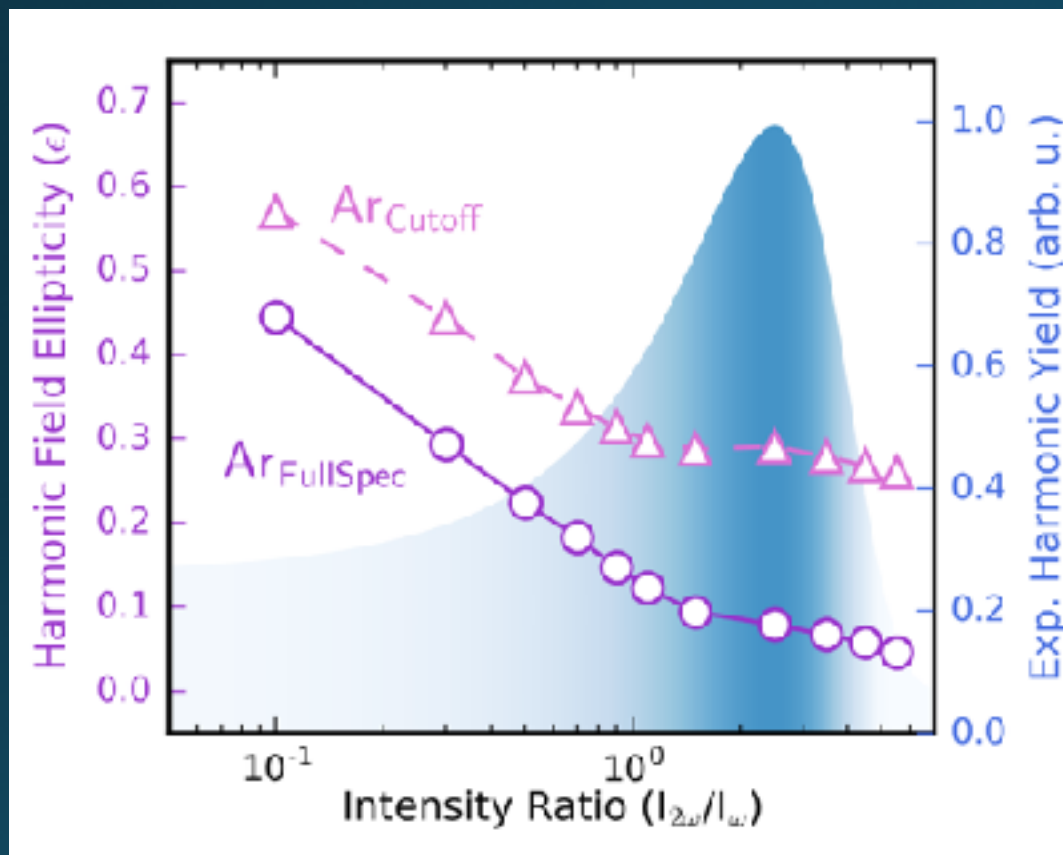
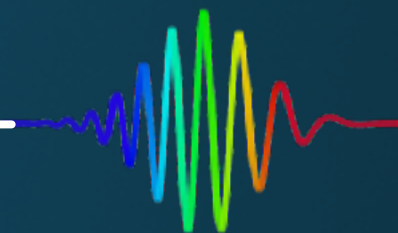
# Attosecond Polarization Control at a Price...

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Mancuso, Phys. Rev. A., 93, 2016

Dorney et. al., Phys. Rev. Lett., 118, 2017

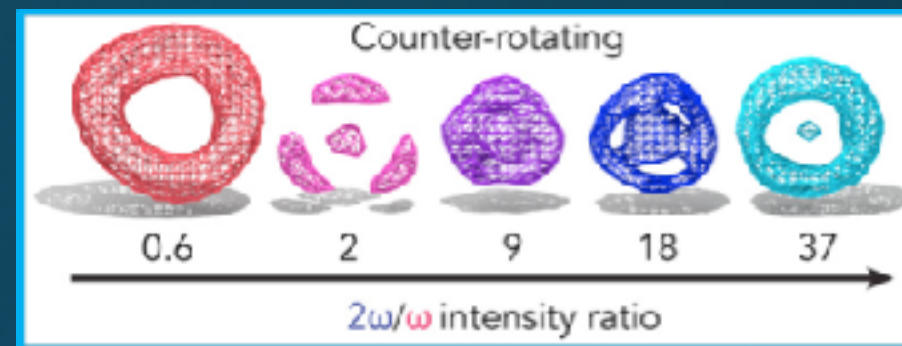
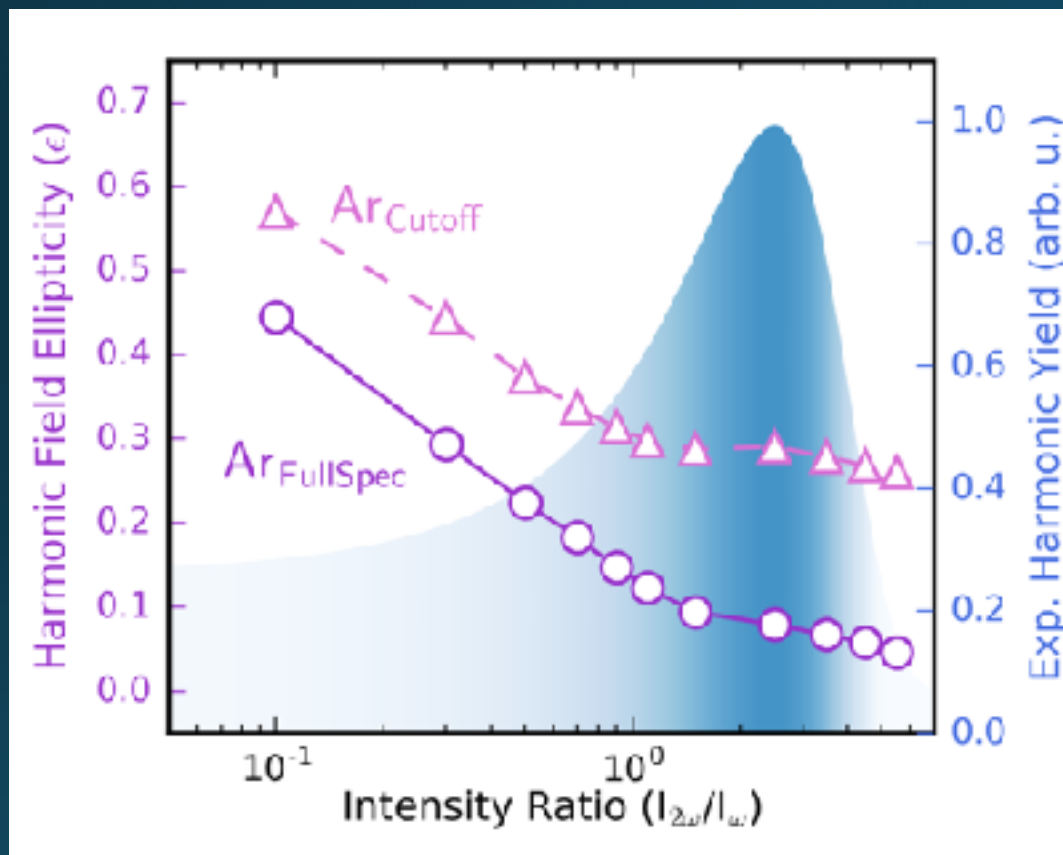
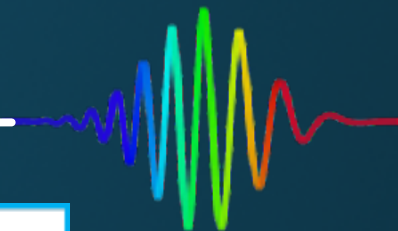


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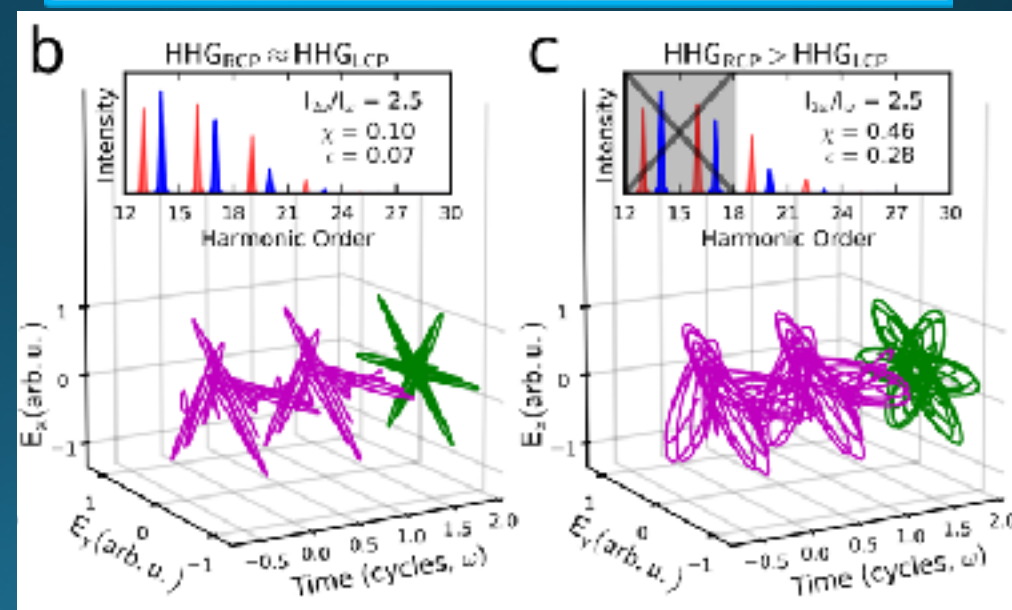
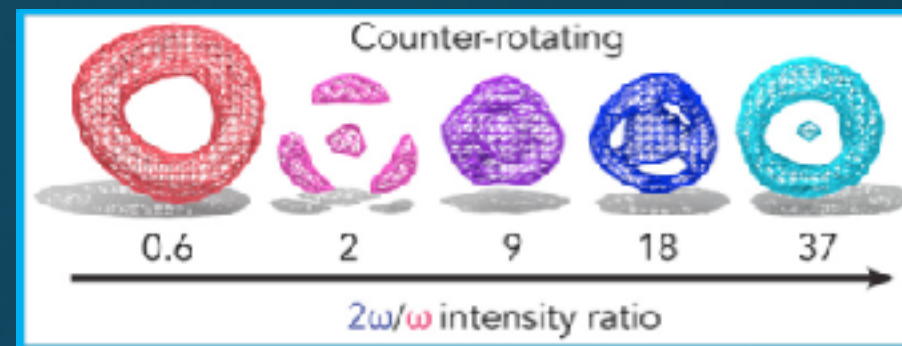
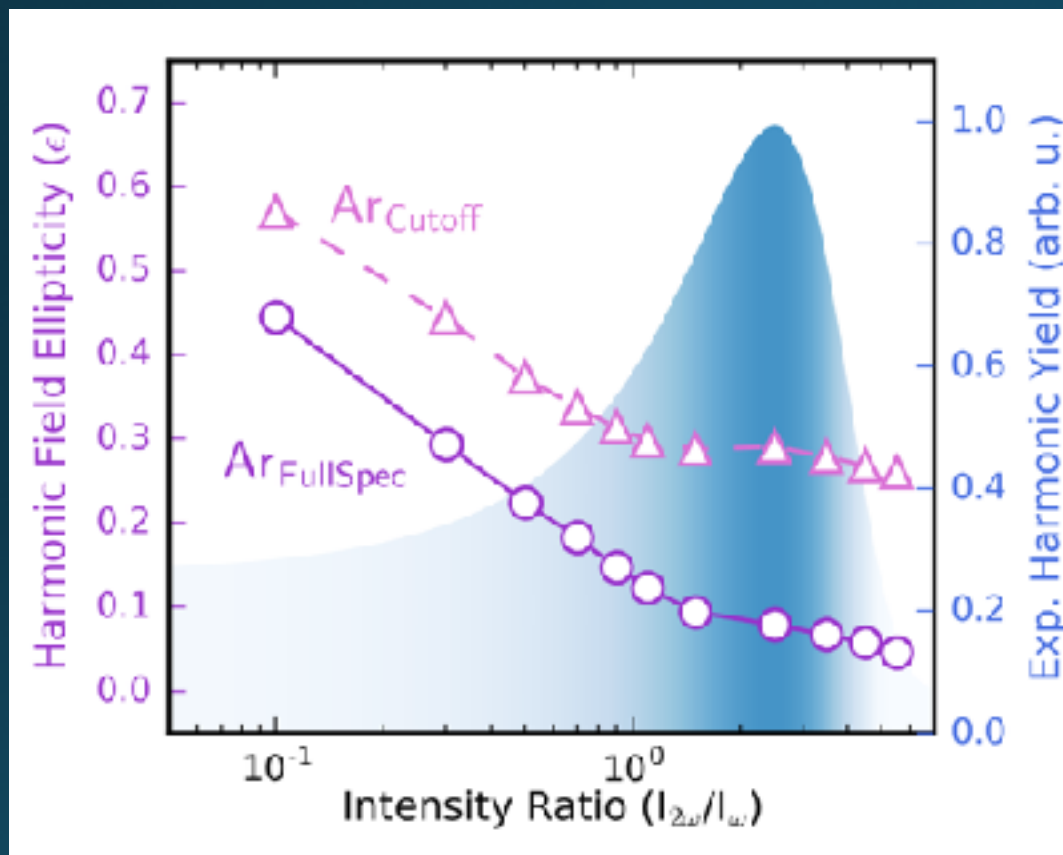
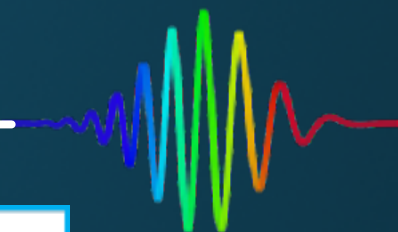
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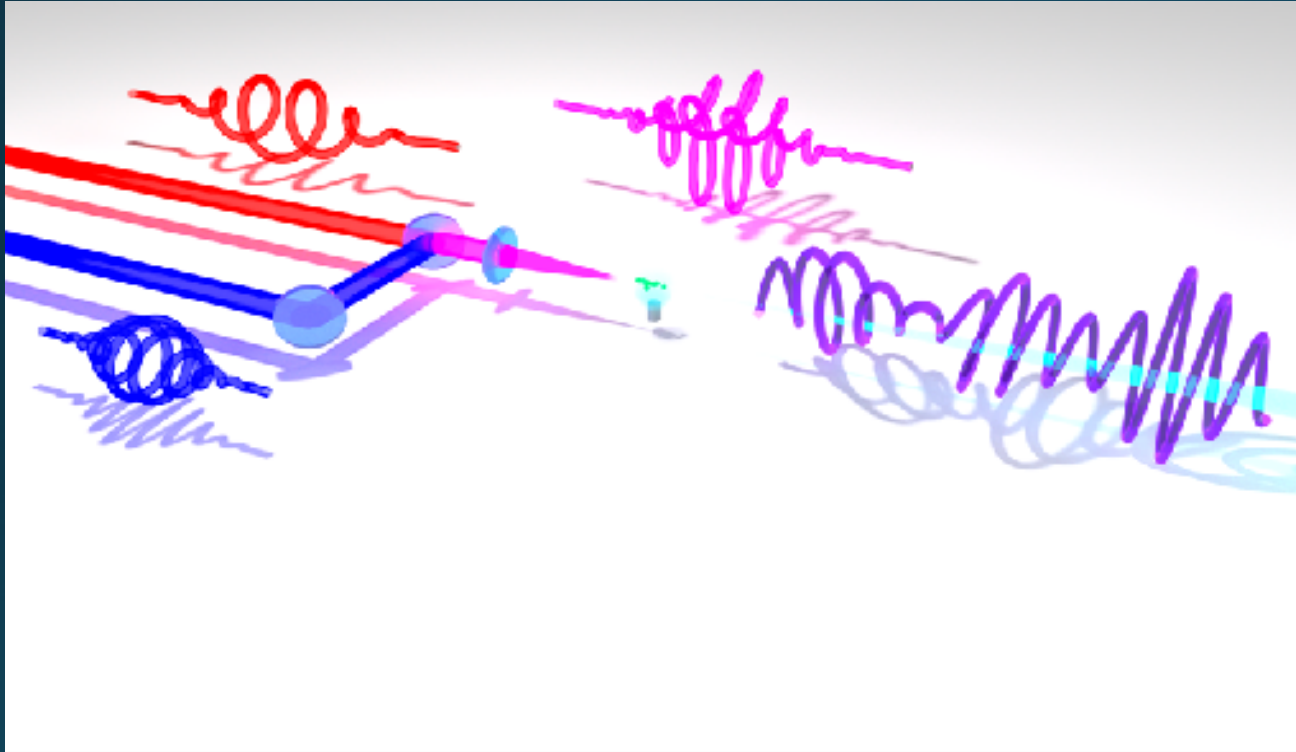
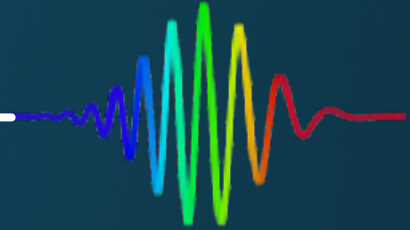
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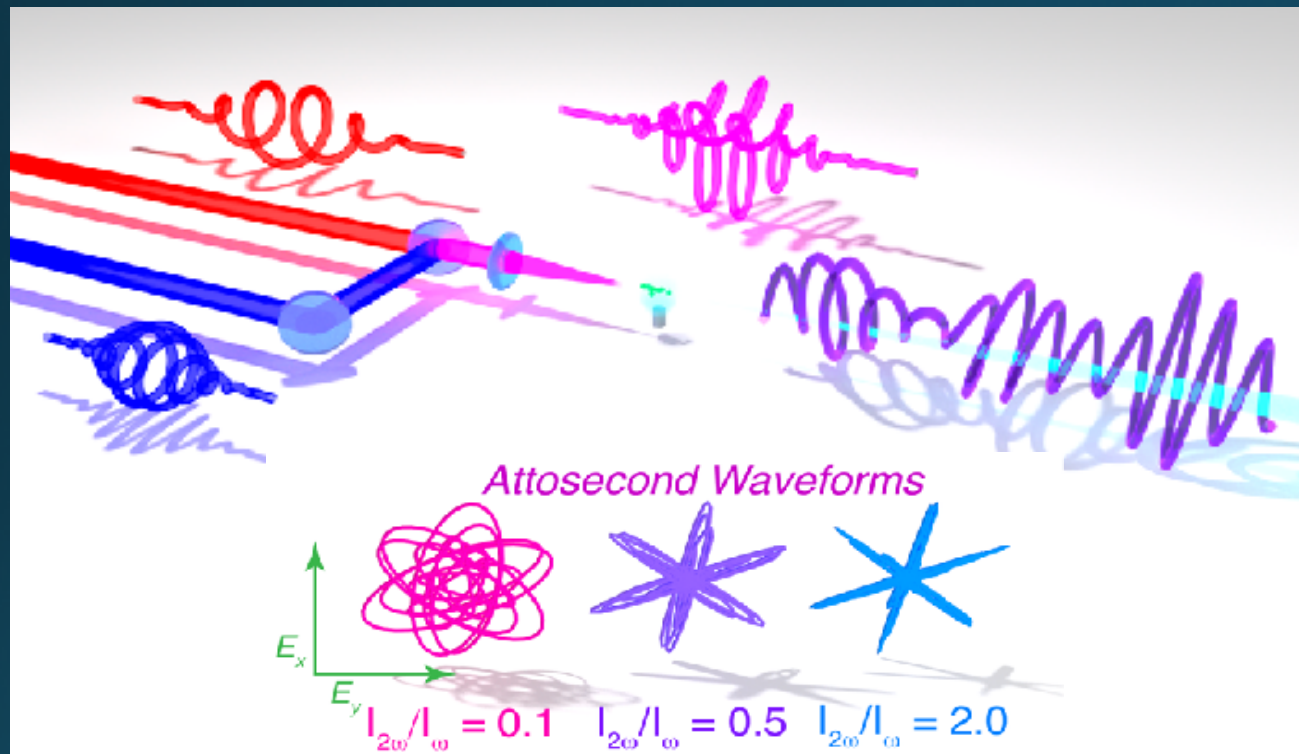
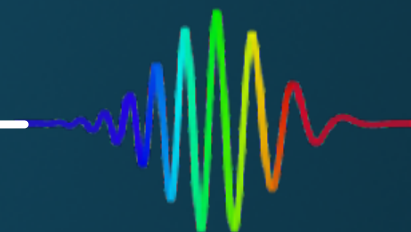
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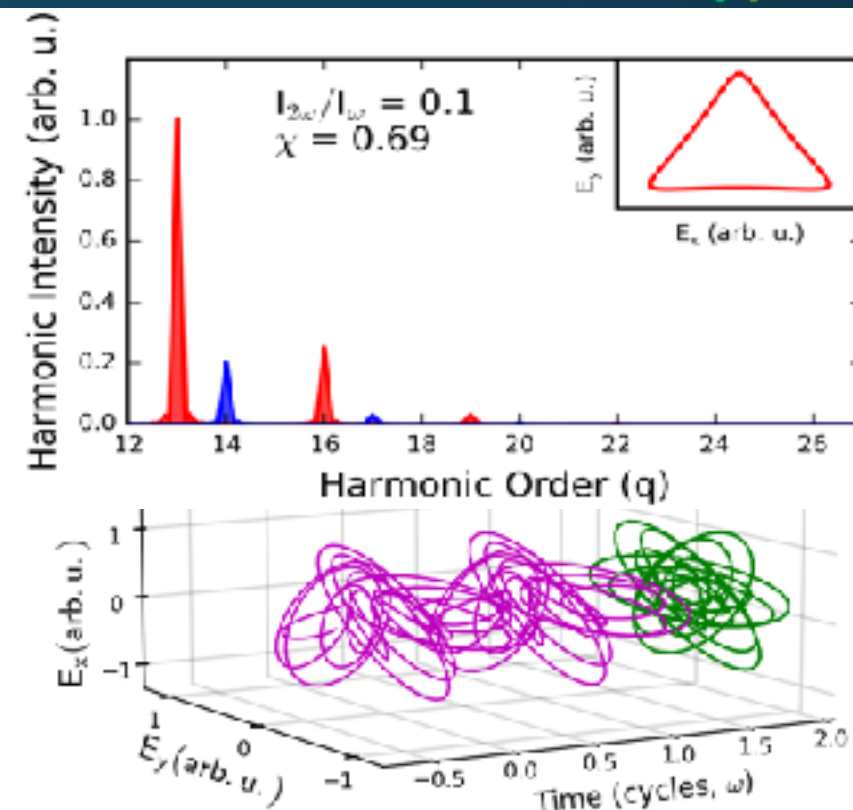
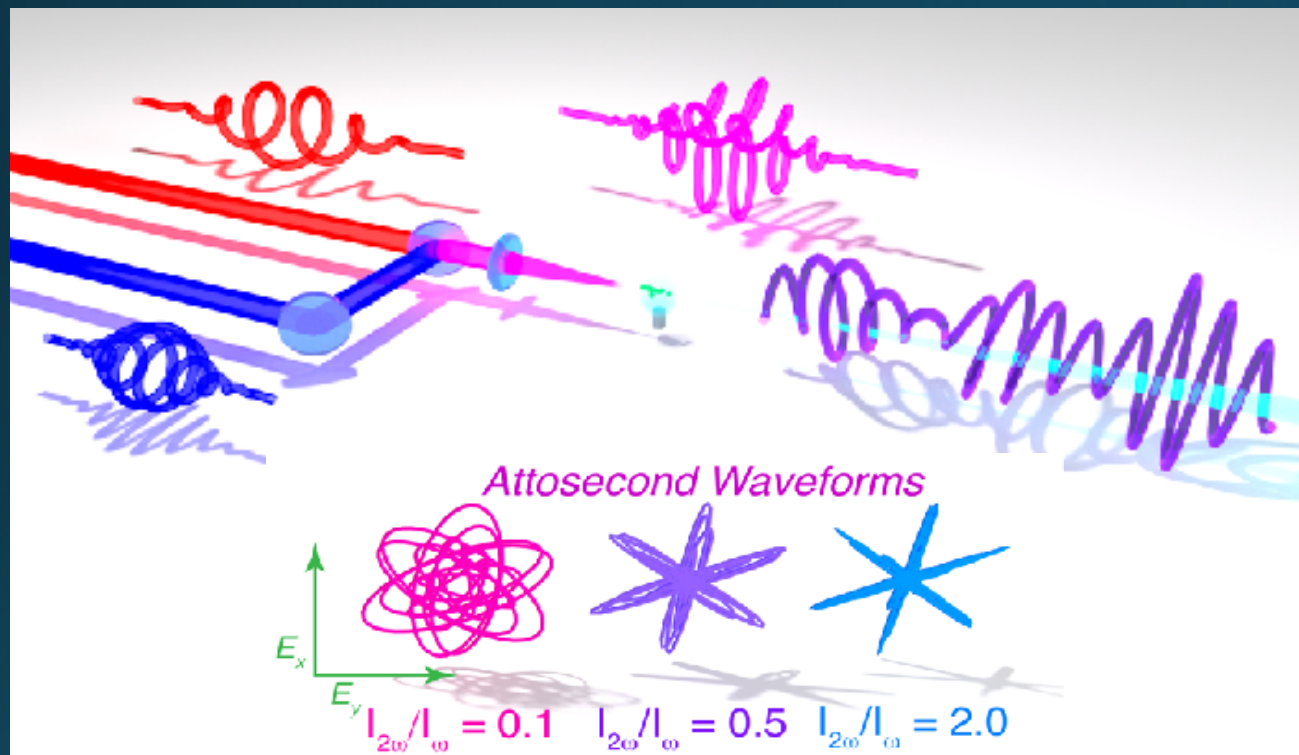
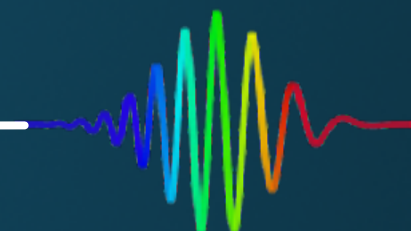
# Production of Elliptically Polarized Attosecond Waveforms: Custom Attosecond Pulses for Chiral Spectroscopies



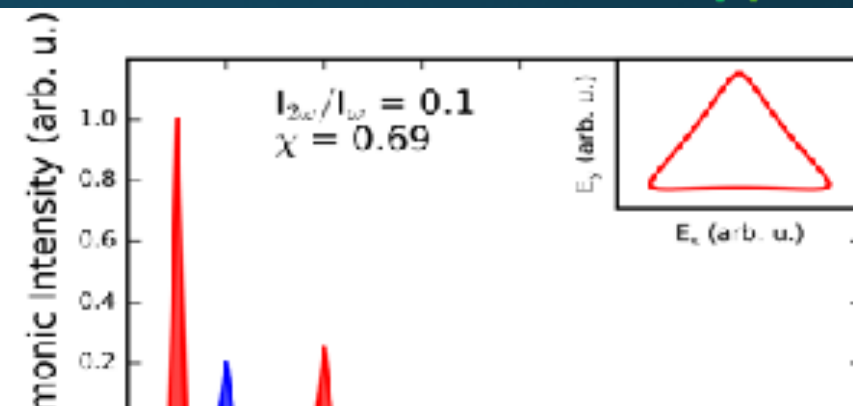
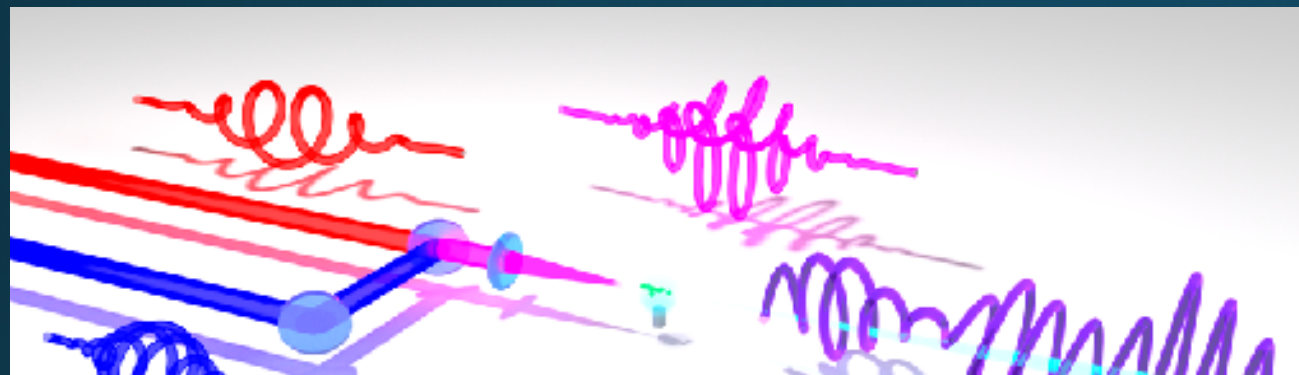
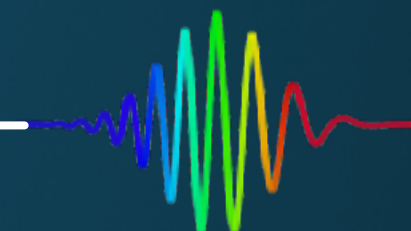
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PRL 119, 063201 (2017)

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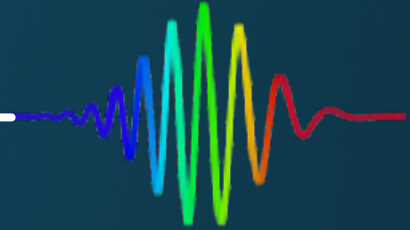
## Helicity-Selective Enhancement and Polarization Control of Attosecond High Harmonic Waveforms Driven by Bichromatic Circularly Polarized Laser Fields

Kevin M. Dorney,<sup>1,\*</sup> Jennifer L. Ellis,<sup>1</sup> Carlos Hernández-García,<sup>2</sup> Daniel D. Hickstein,<sup>1</sup> Christopher A. Mancuso,<sup>1</sup> Nathan Brooks,<sup>1</sup> Tingting Fan,<sup>1</sup> Guangyu Fan,<sup>3</sup> Dmitriy Zusin,<sup>1</sup> Christian Gentry,<sup>1</sup> Patrik Grychtol,<sup>1</sup> Henry C. Kapteyn,<sup>1</sup> and Margaret M. Murnane<sup>1</sup>



# Production and Control of Elliptically Polarized Waveforms: Future Demos and Applications

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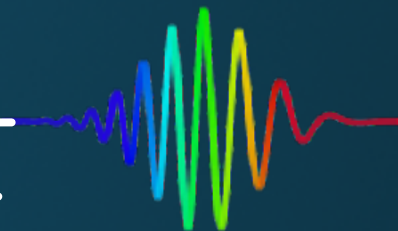




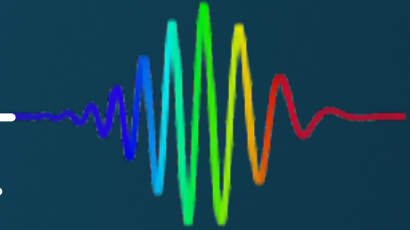
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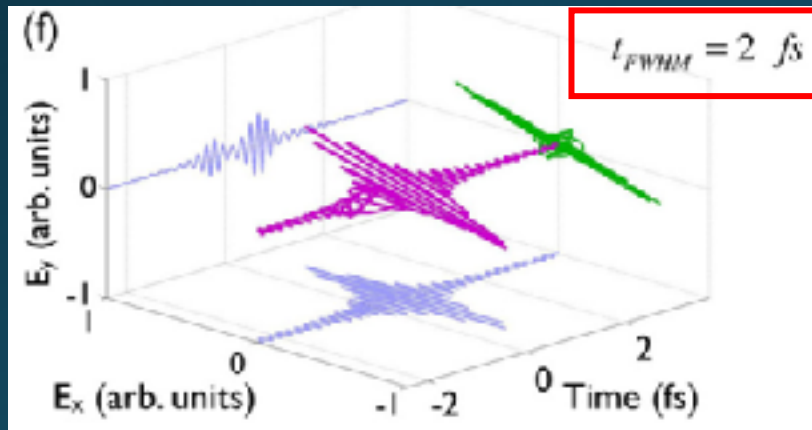
- **Helicity-selective CPHHG is completely compatible with time-gating techniques.**
  - Intensity gating, ionization gating, external field gate

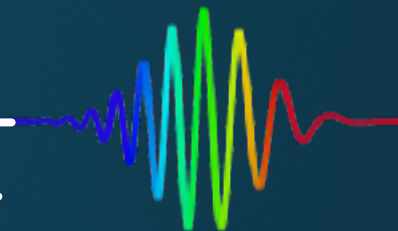




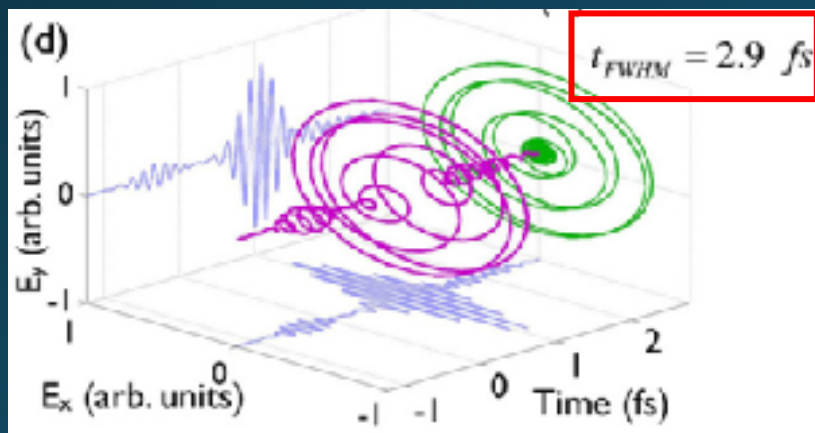


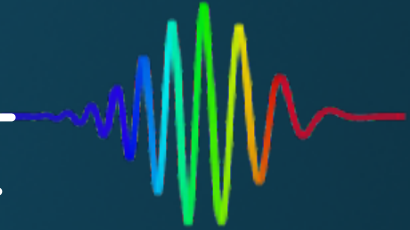
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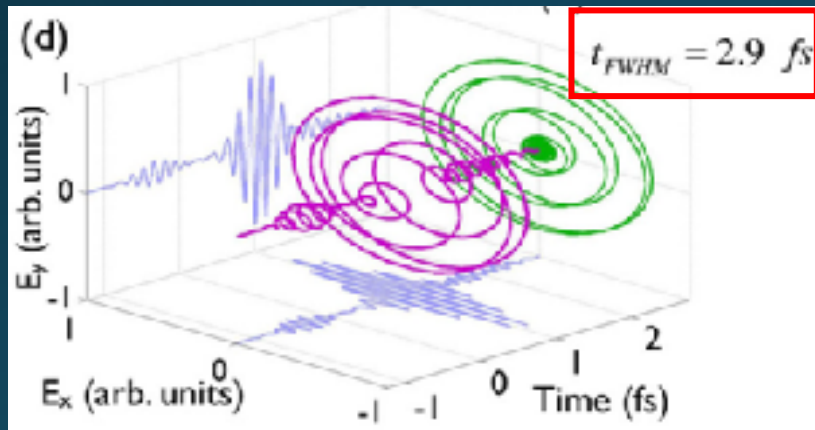


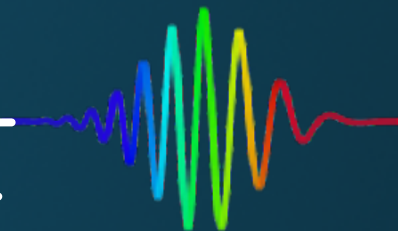
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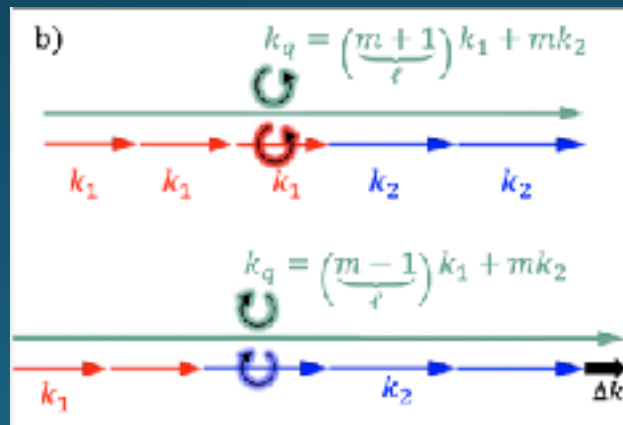
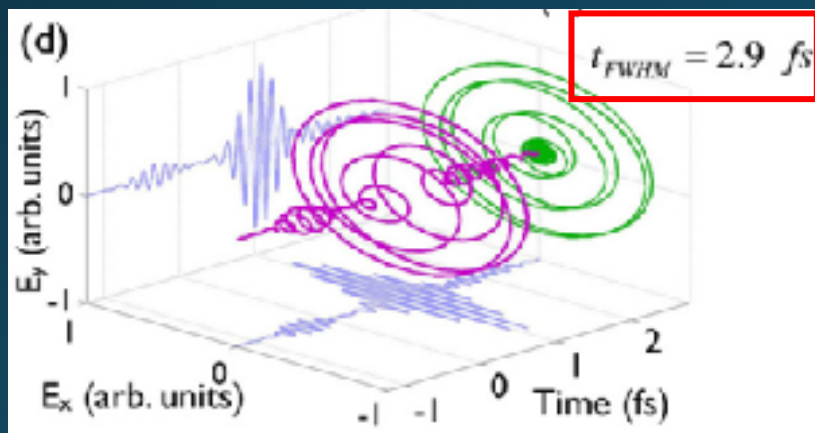


- Helicity-selective CPHHG is completely compatible with time-gating techniques.
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- Macroscopic control can yield APTs with even higher ellipticity.
  - Phase-matching conditions can be manipulated to increase spectral chirality

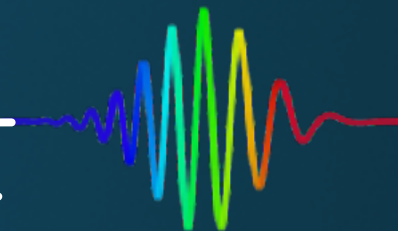




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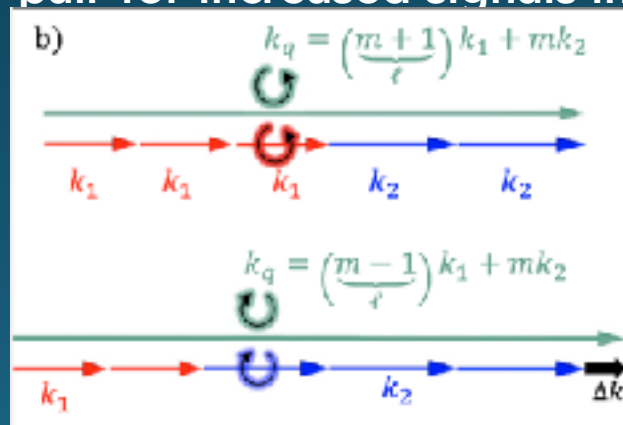
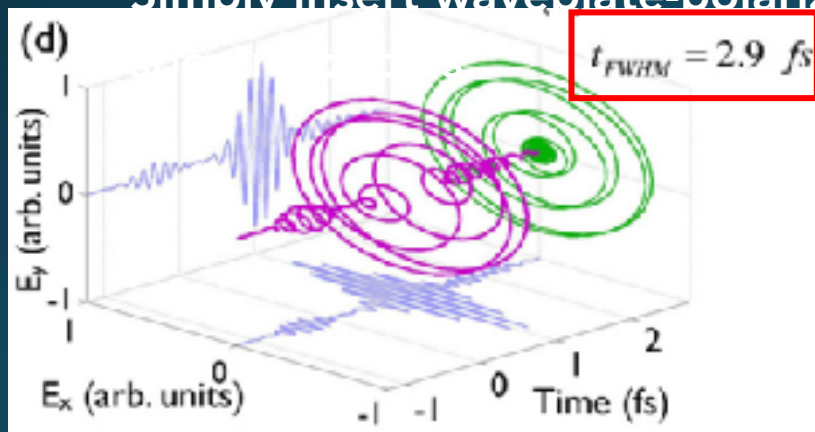
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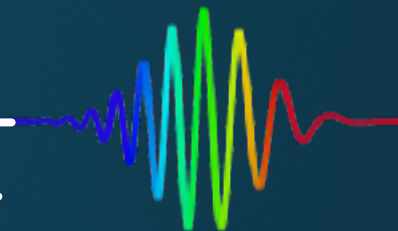
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➤ Straight-forward and robust methodology: elliptical APTs from existing systems

- Simply insert waveplate-polarizer-pair for increased signals in chiral





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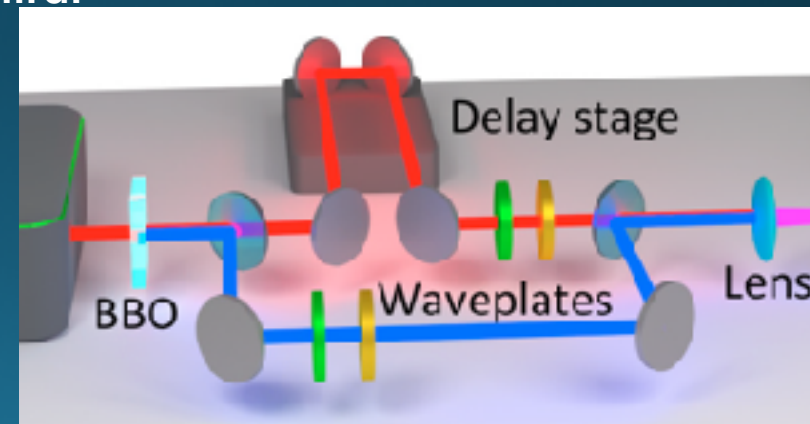
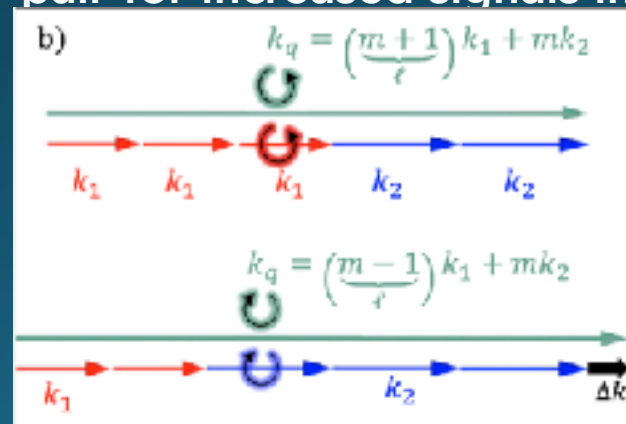
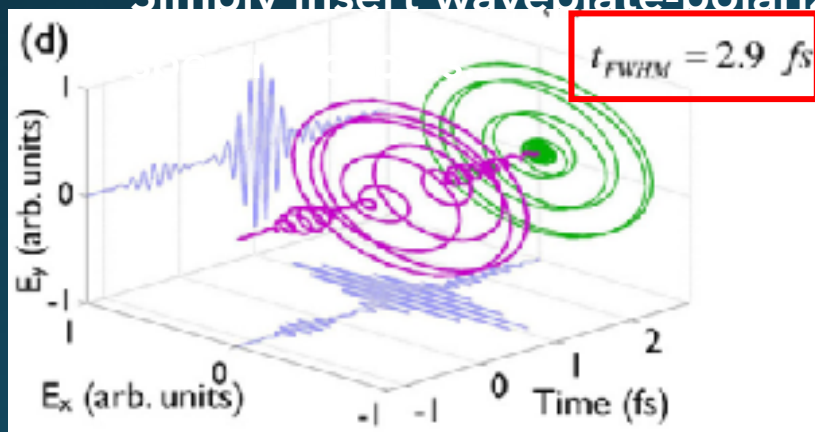
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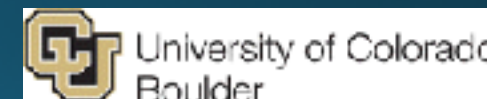
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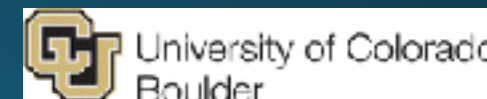
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