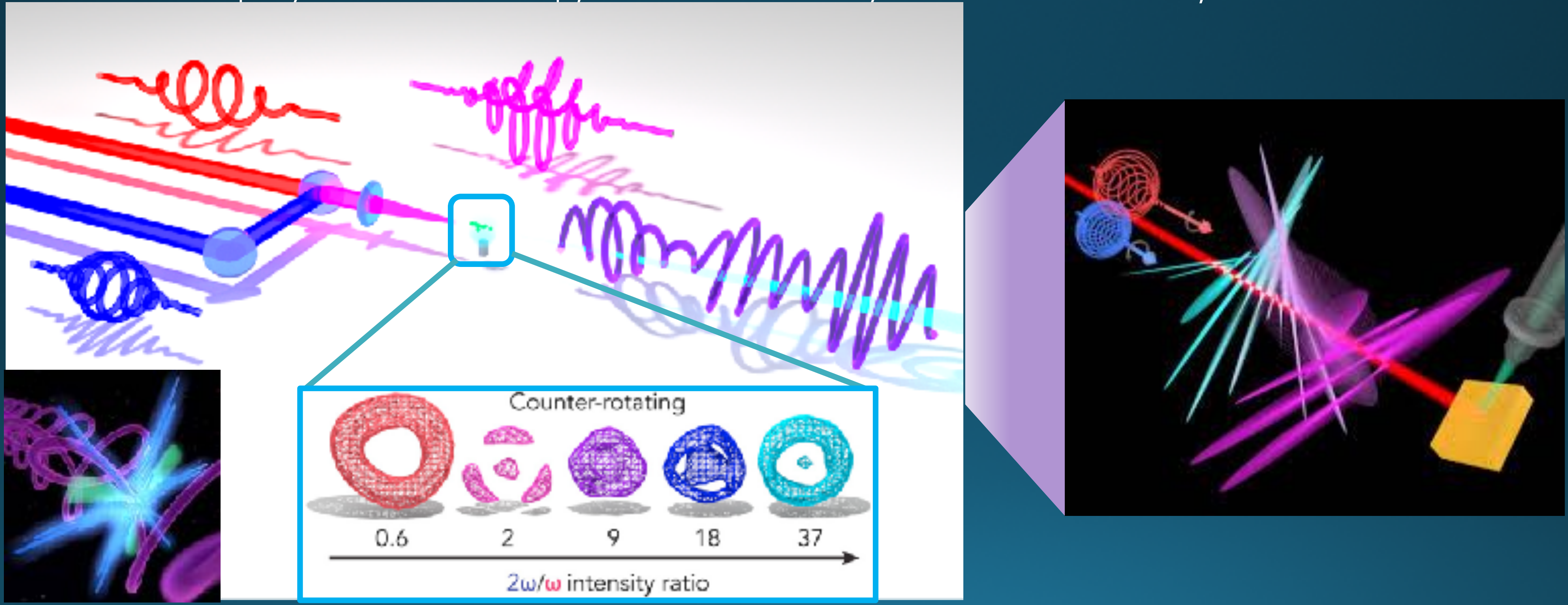
Elliptically Polarized Attosecond Pulse Trains Produced via Circularly Polarized High Harmonic Generation

Kevin M. Dorney

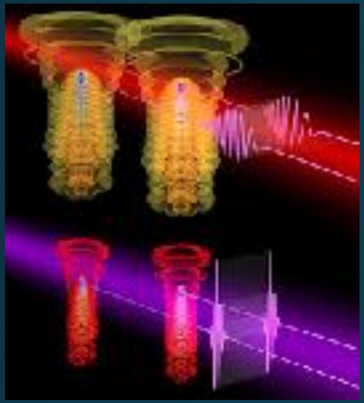
Kapteyn-Murnane Group, JILA and University of Colorado Boulder, USA



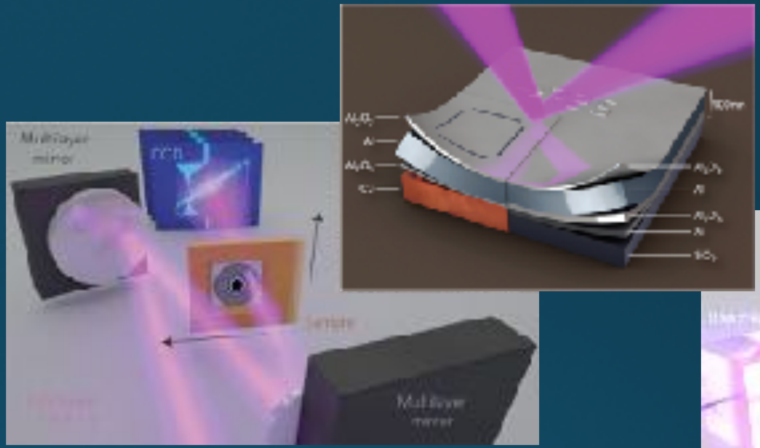
A&T Topical Review on Extreme Ultraviolet and Soft X-ray Sources and Application II, CLEO 2017

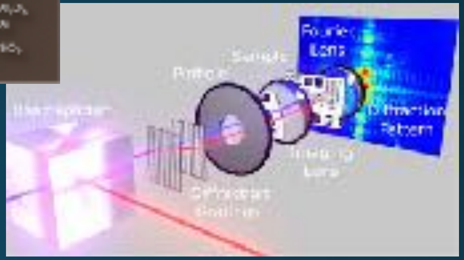
Light and Materials Science in the KM Group: AMO Dynamics at Extreme Spatial and Temporal Scales

Attosecond Extreme Nonlinear Optics

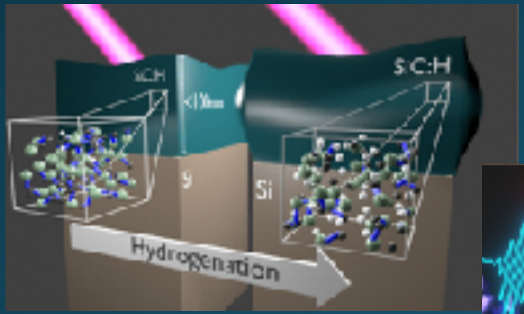


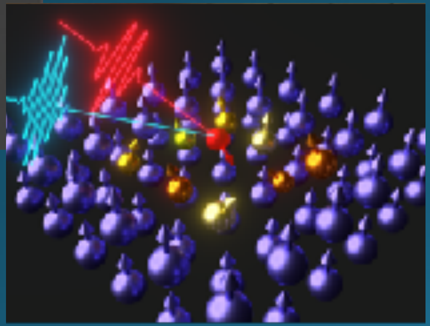
Coherent x-ray Imaging

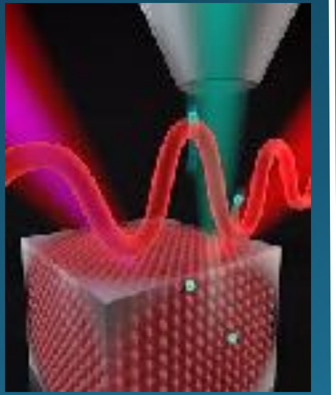




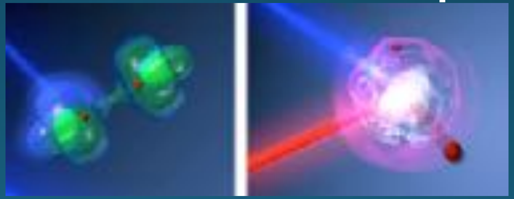
Ultrafast Materials Science

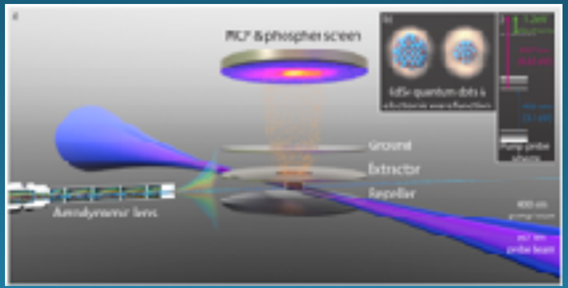


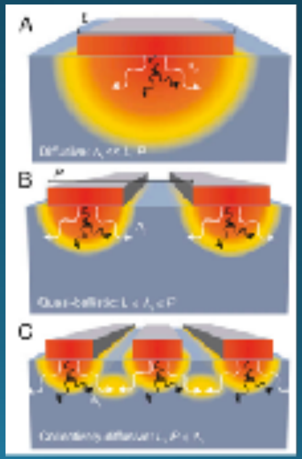


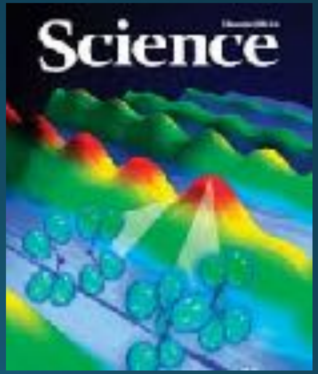


Nano-Molecular Spectroscopy and Dynamics





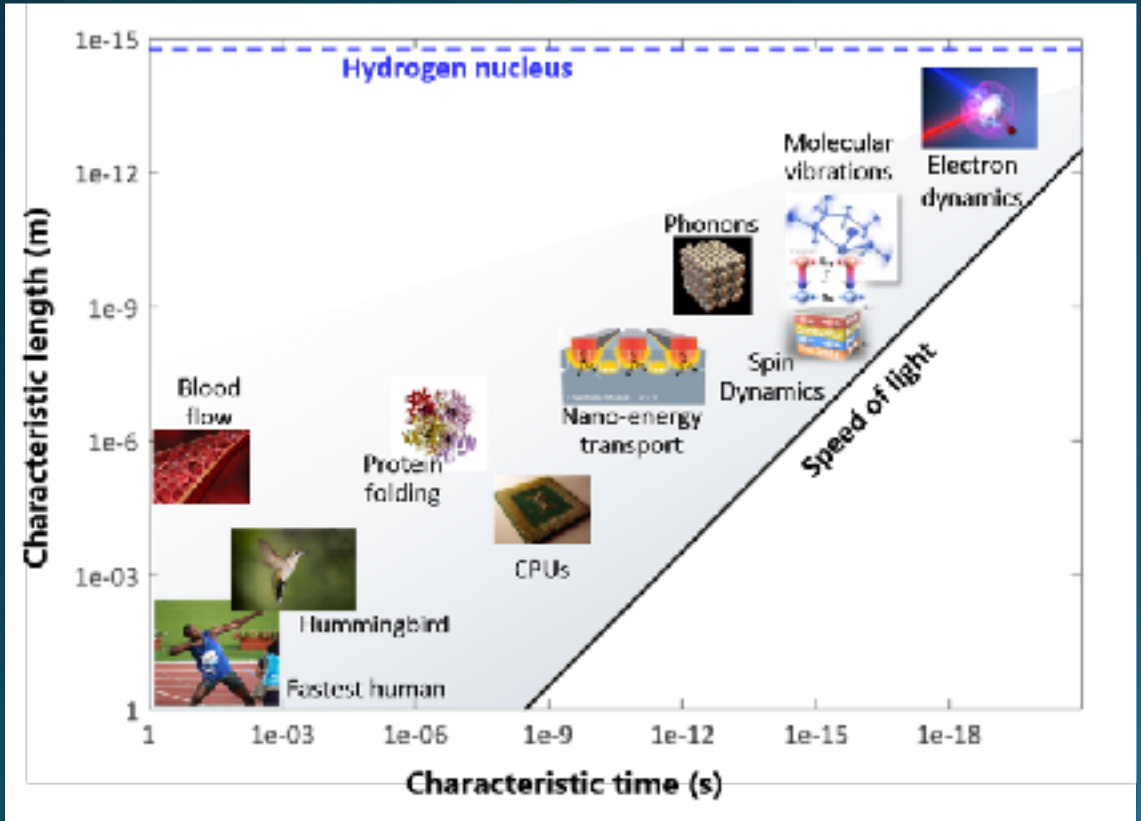




High Harmonic Generation (HHG): Light Science at the Atomic Frontier and Beyond

The White Whale of the Physical Sciences

- Direct observation of atomic and molecular scale transformations at their natural **time** and **length scales**.



Courtesy: Nico Hernandez Chupak, KM Group

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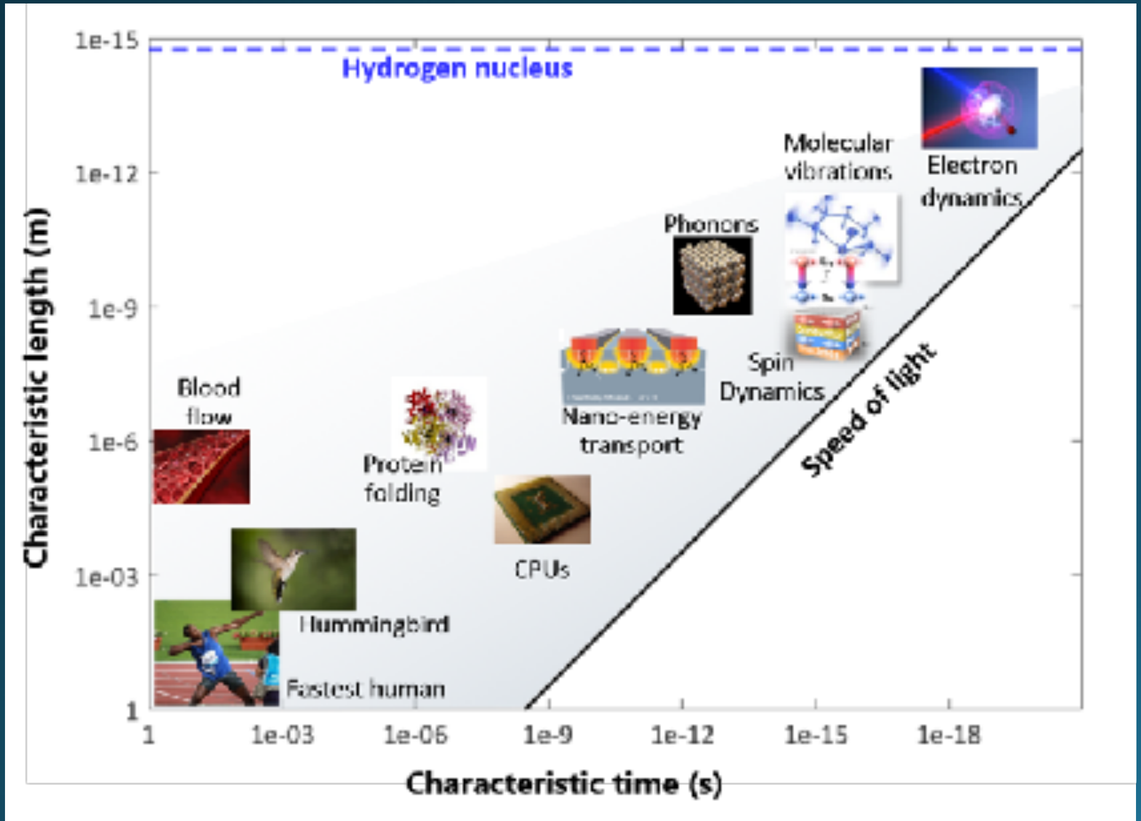

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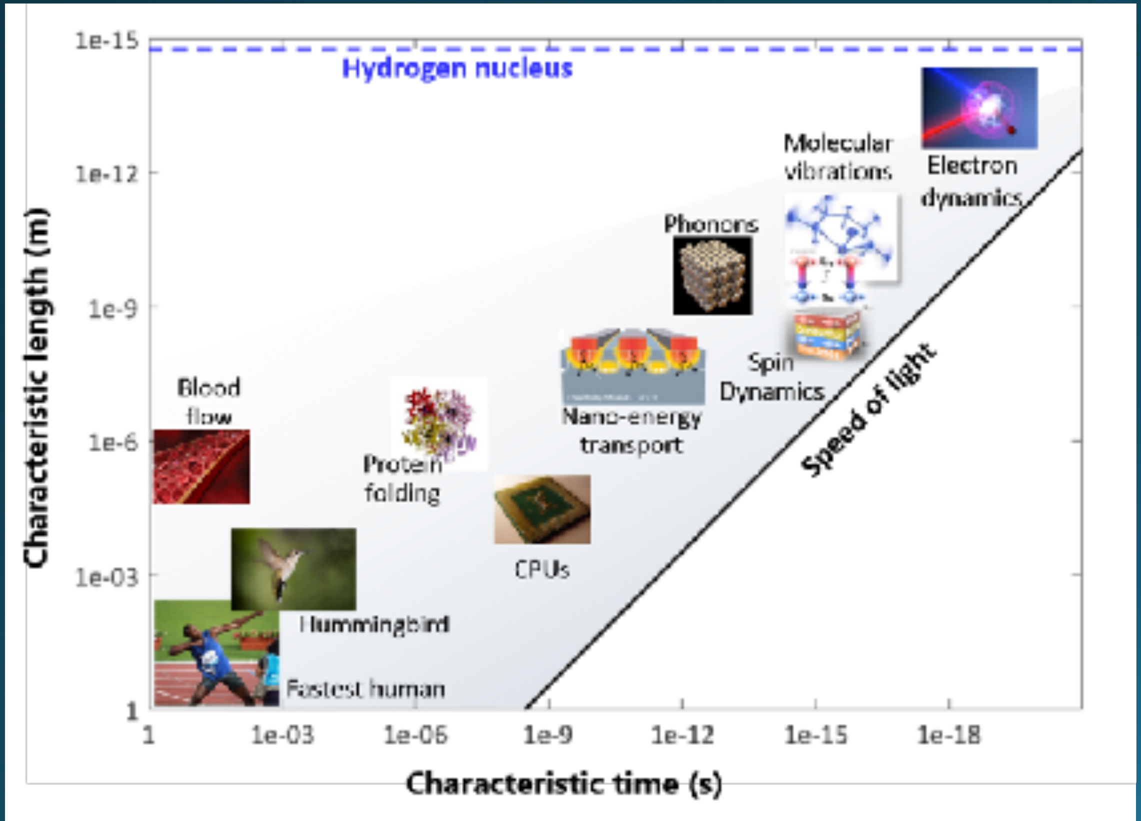

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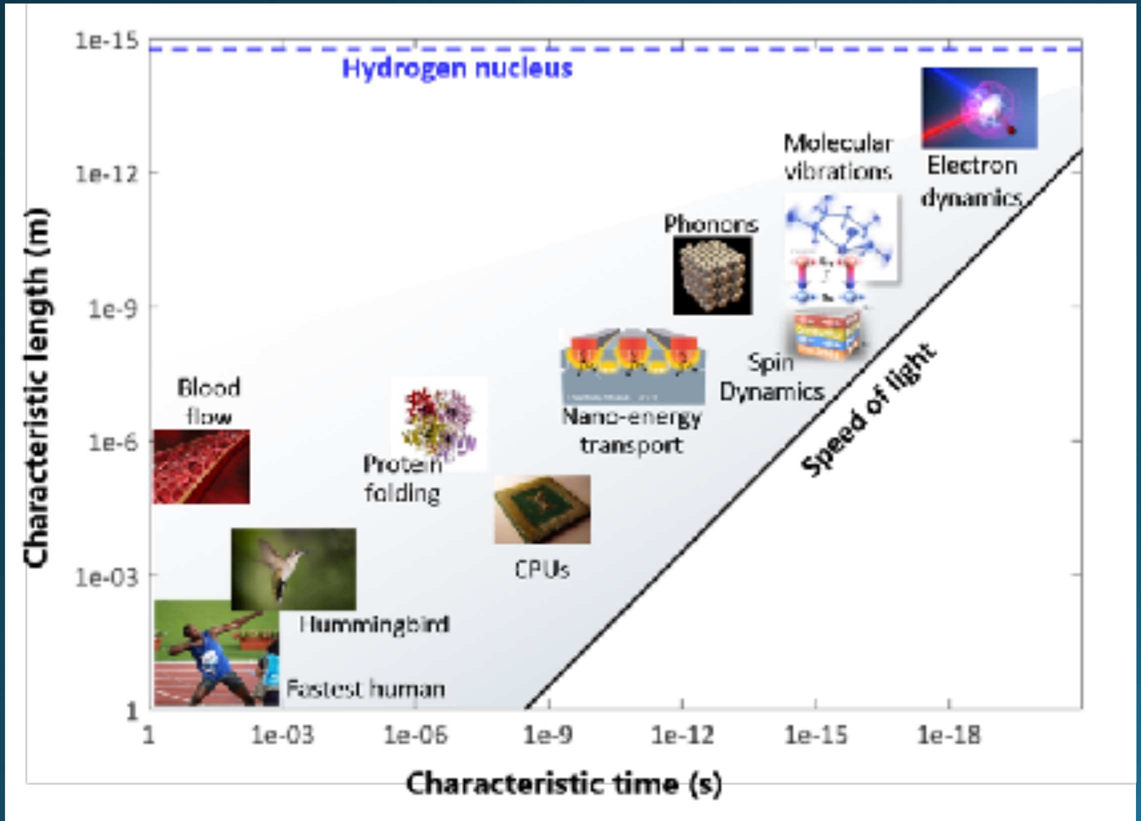

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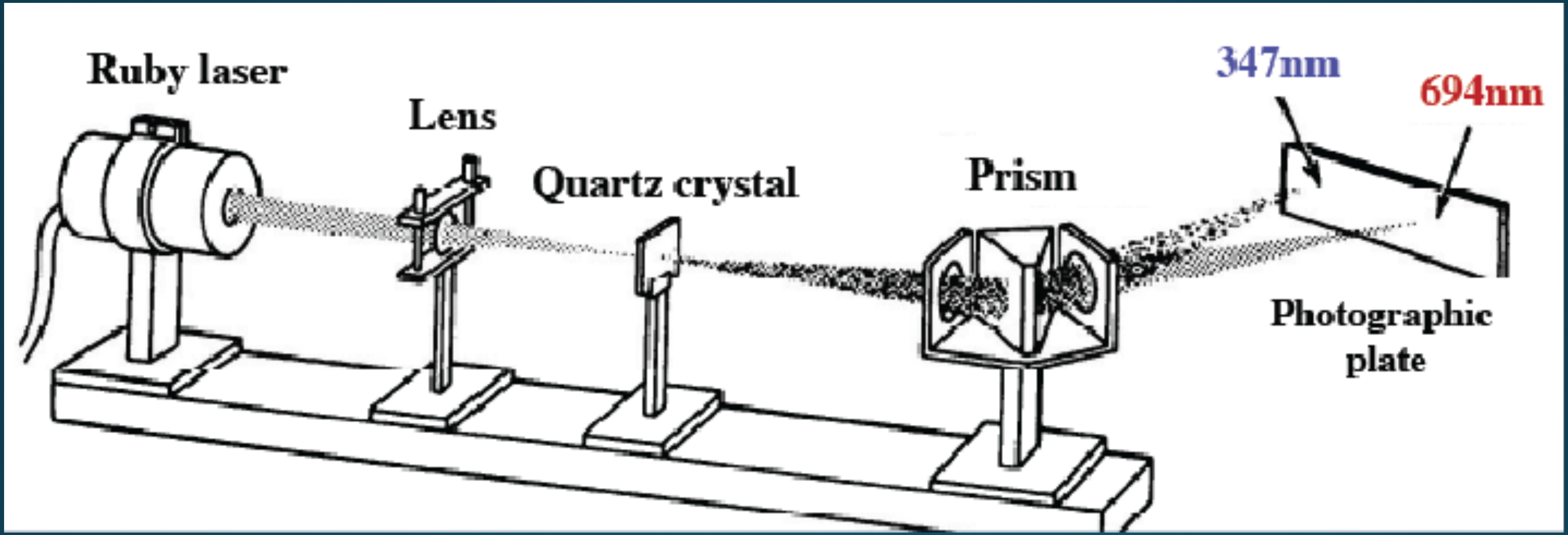
HHG in the Beginning:
A Barely Visible "Smudge" Revolutionizes Optical Science



Franken et al. PRL, **7**, 1961

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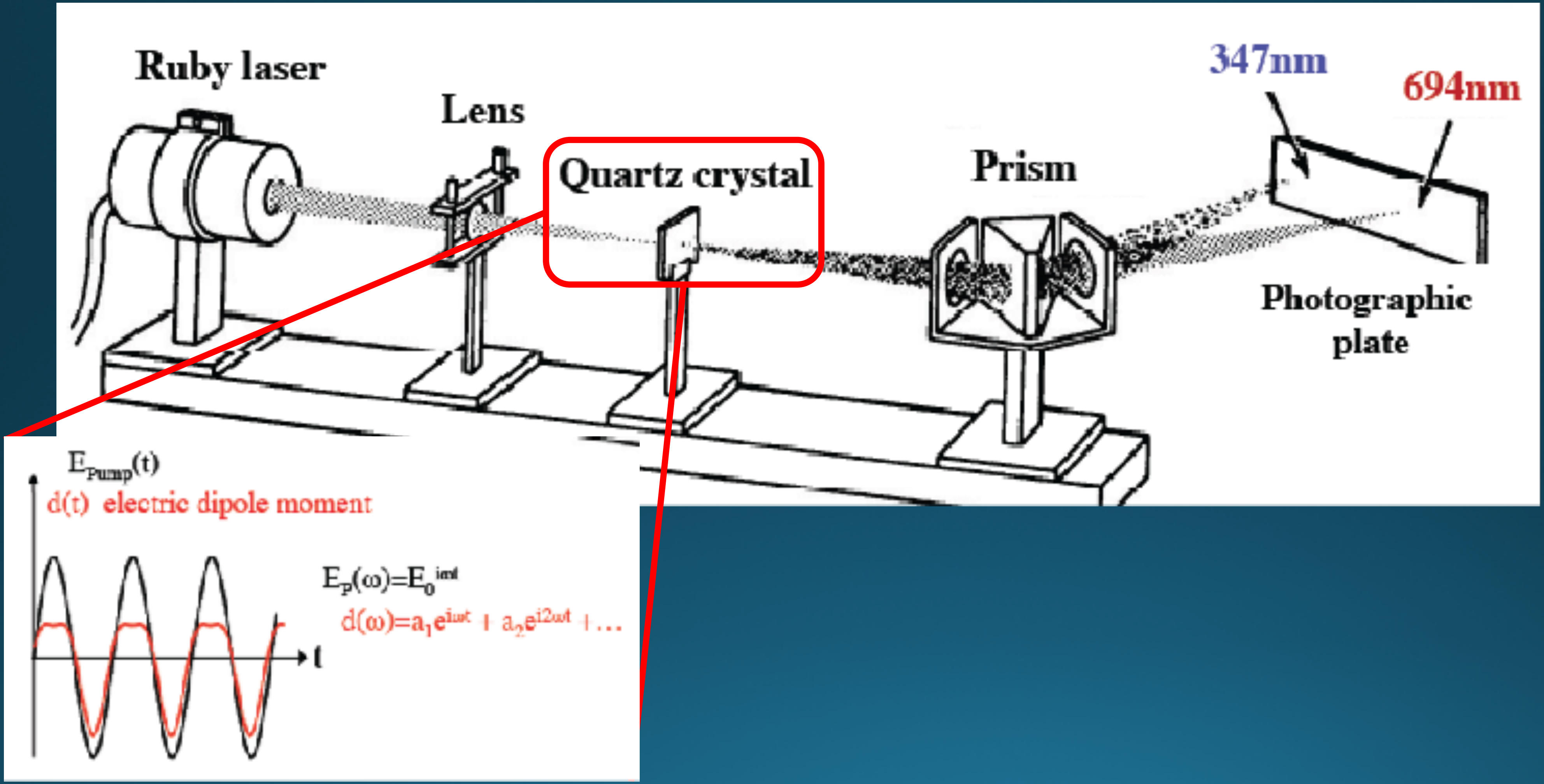

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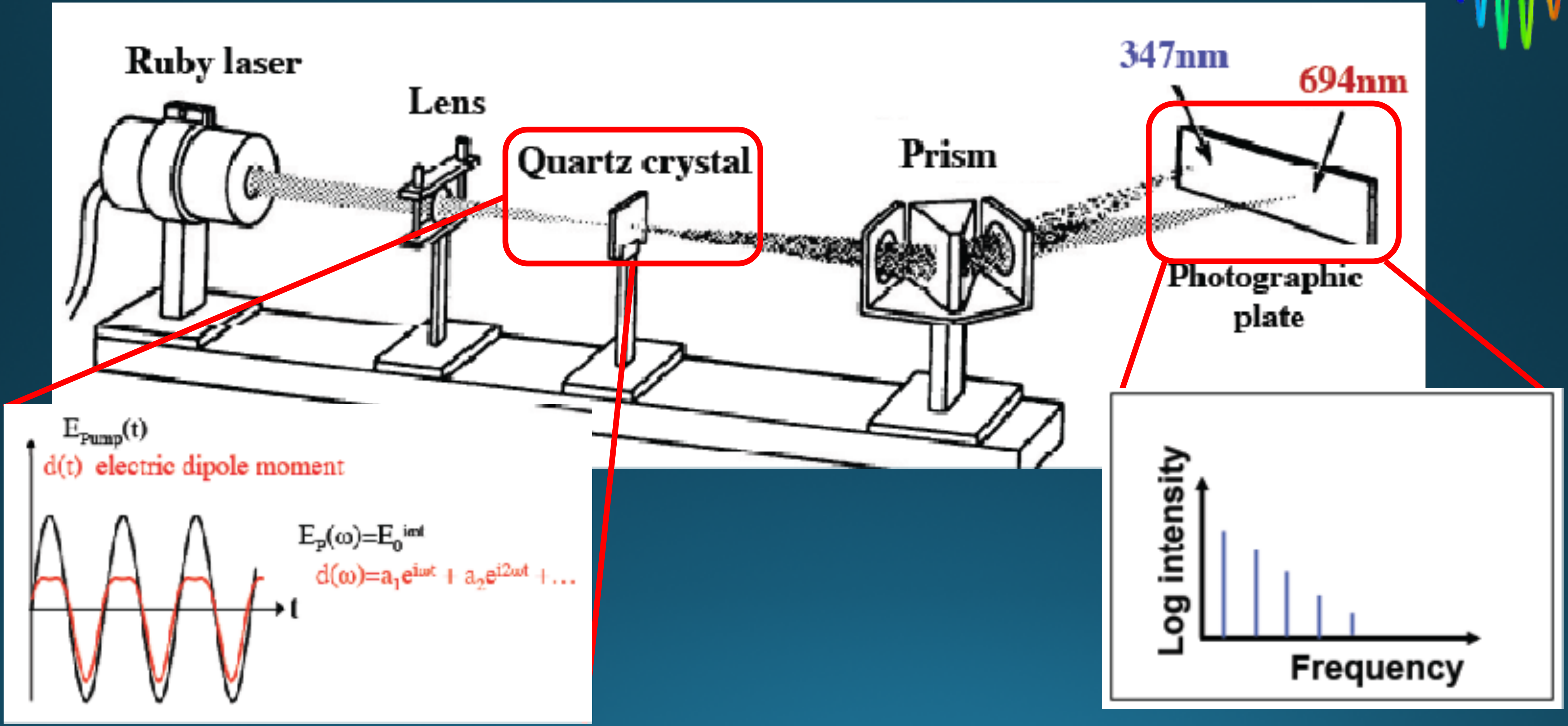

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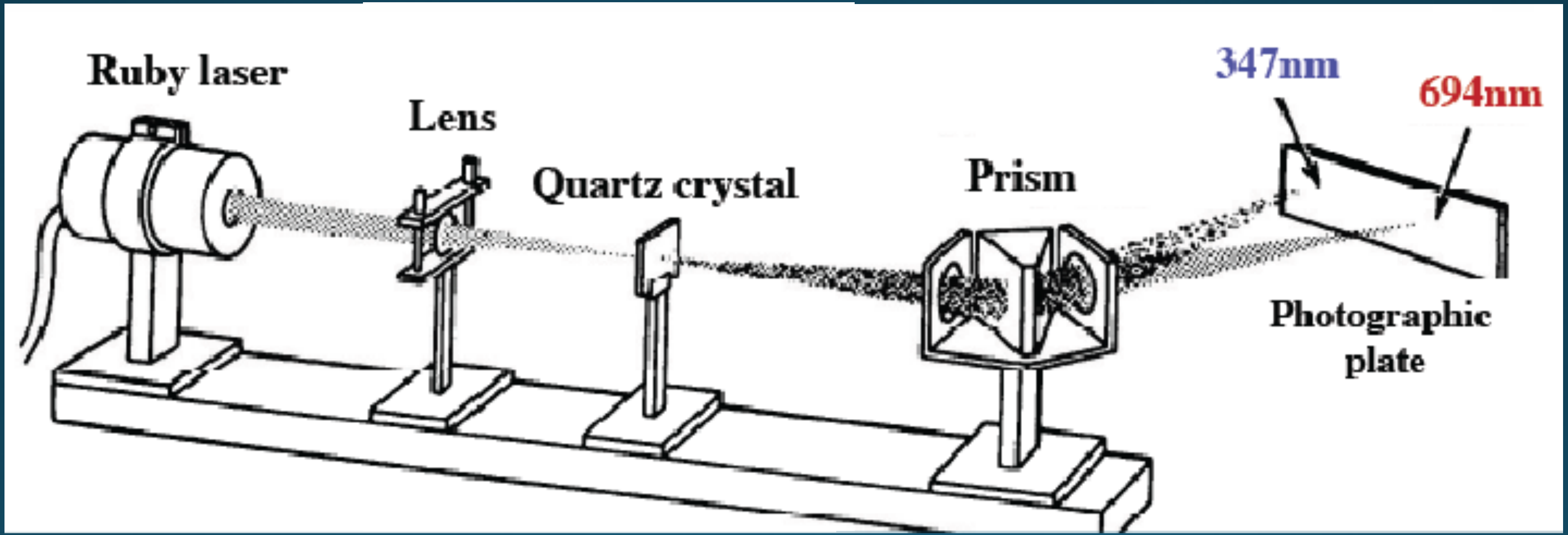
Evolution of HHG: Perturbative Optics to Extreme Nonlinear Optical Science

- From perturbative optics to extreme nonlinear optical science.



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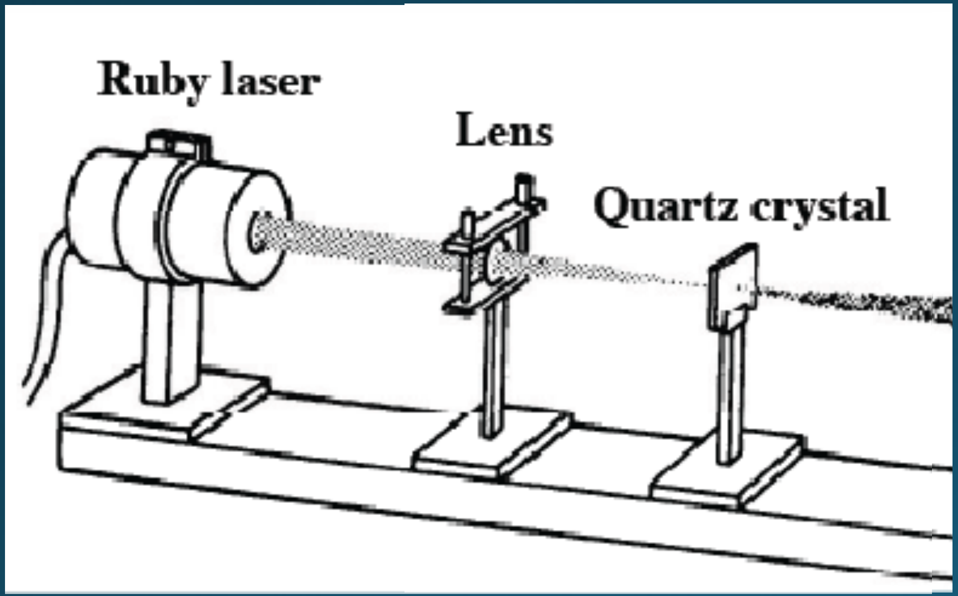

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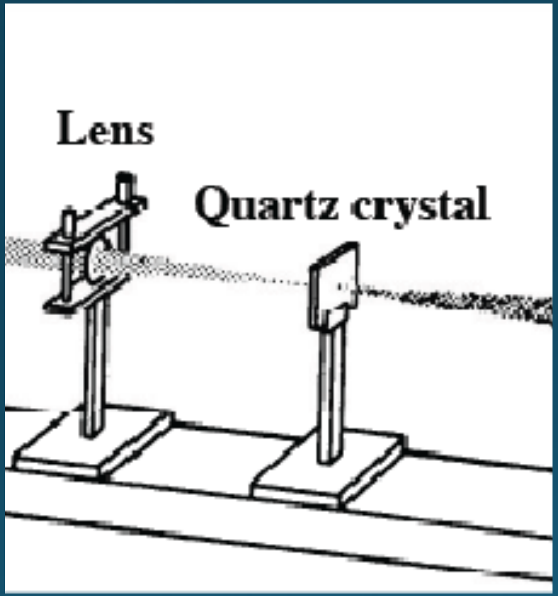

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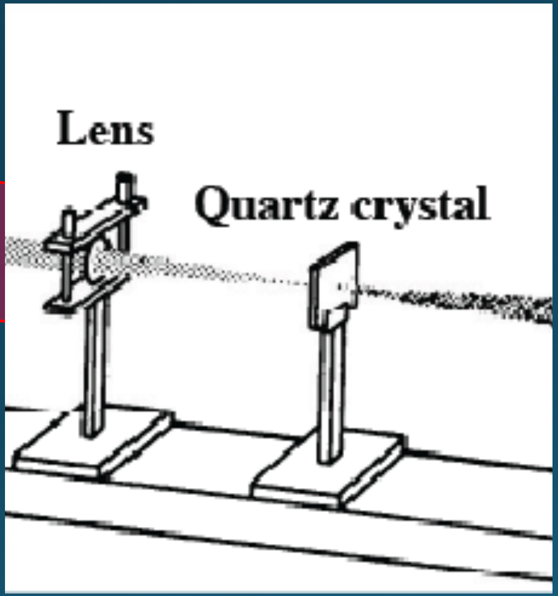

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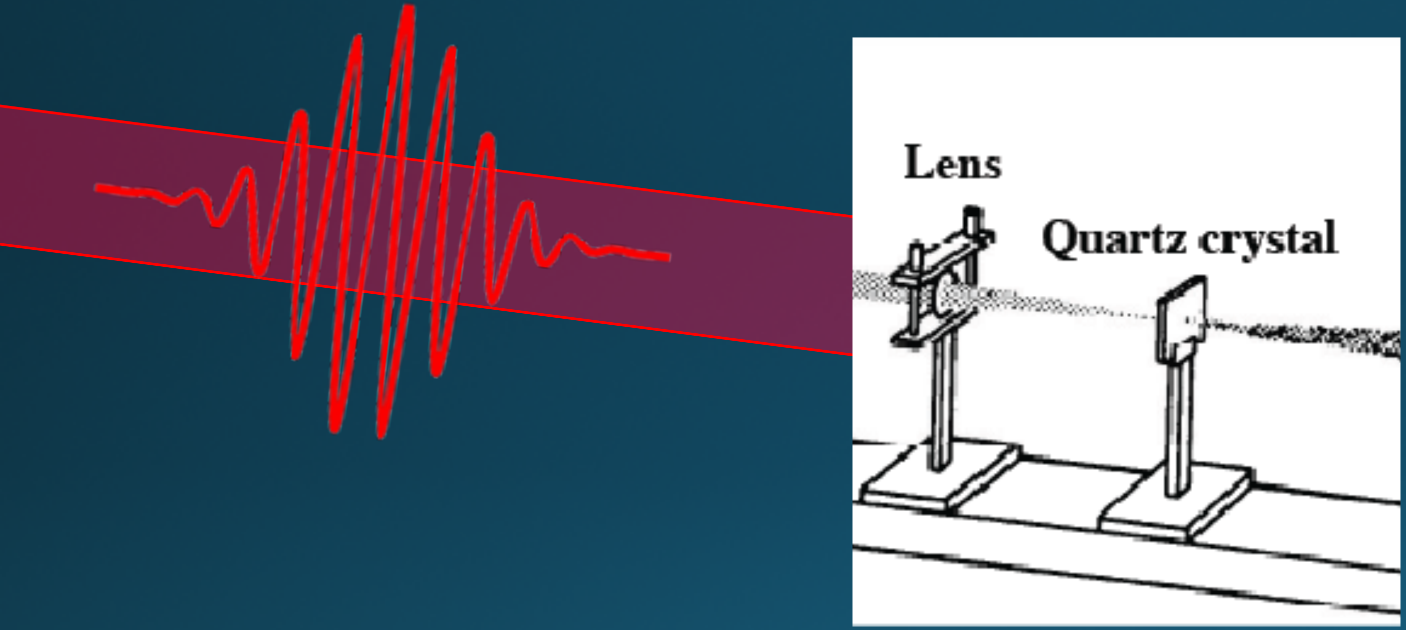

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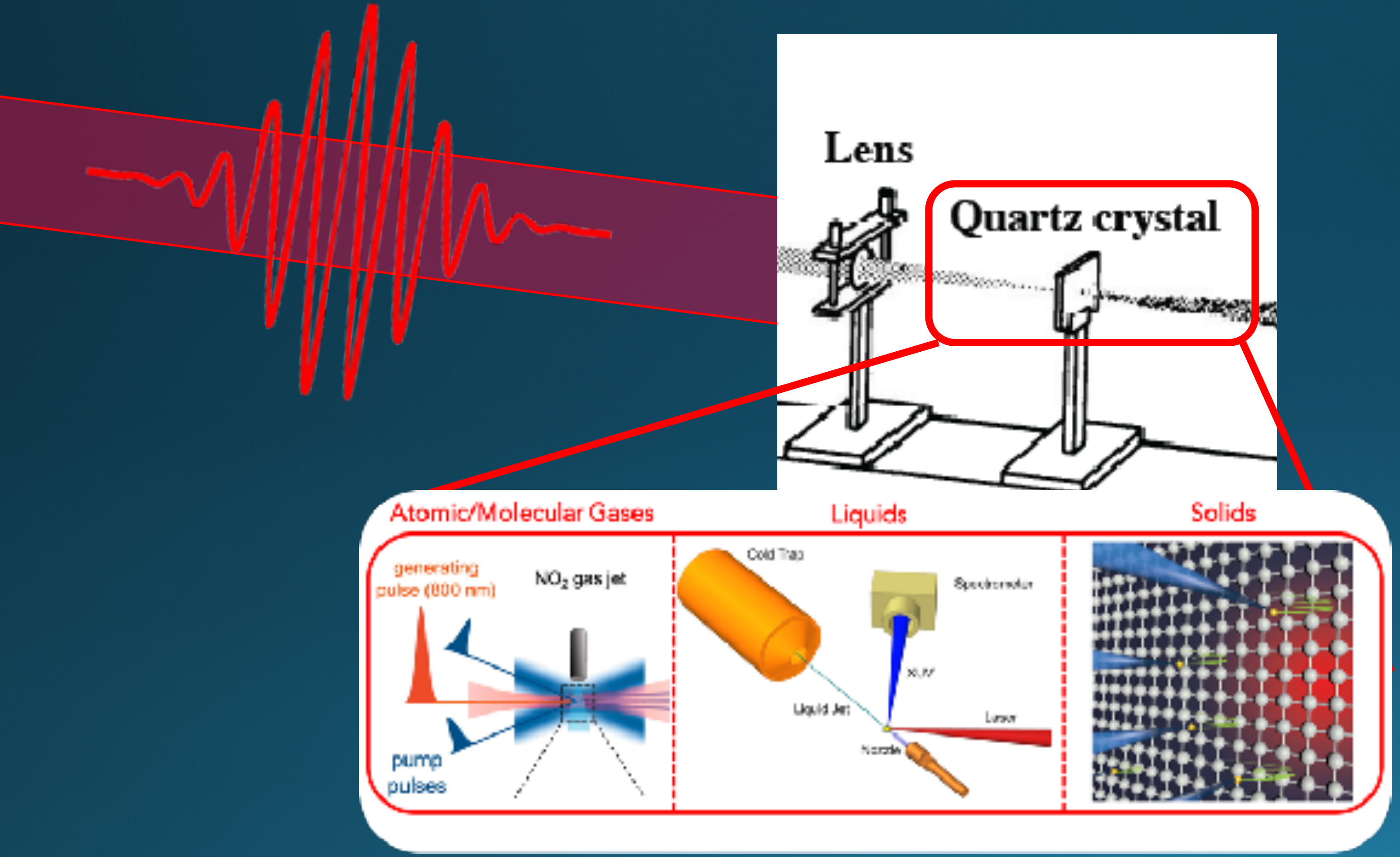

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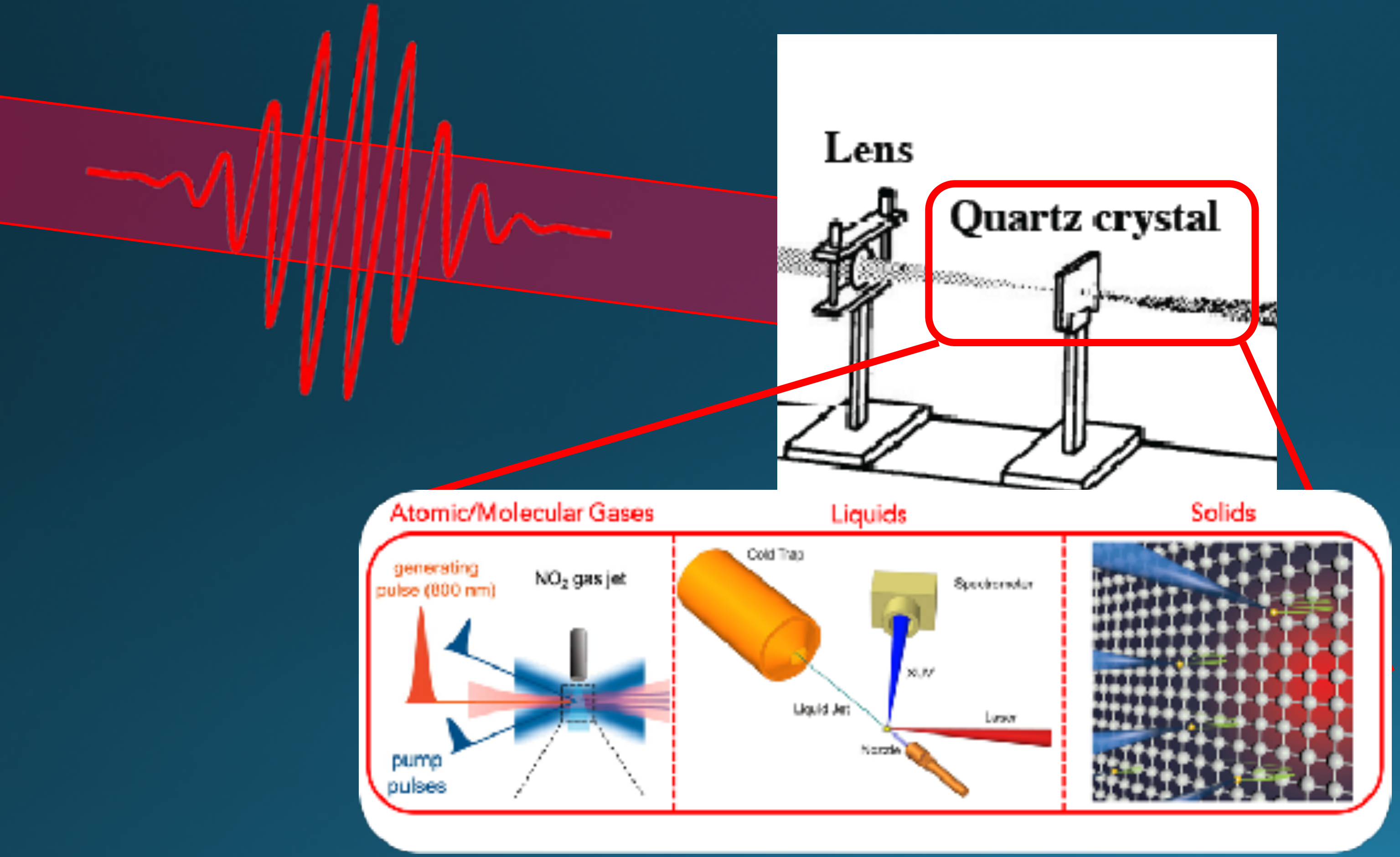
Time Domain



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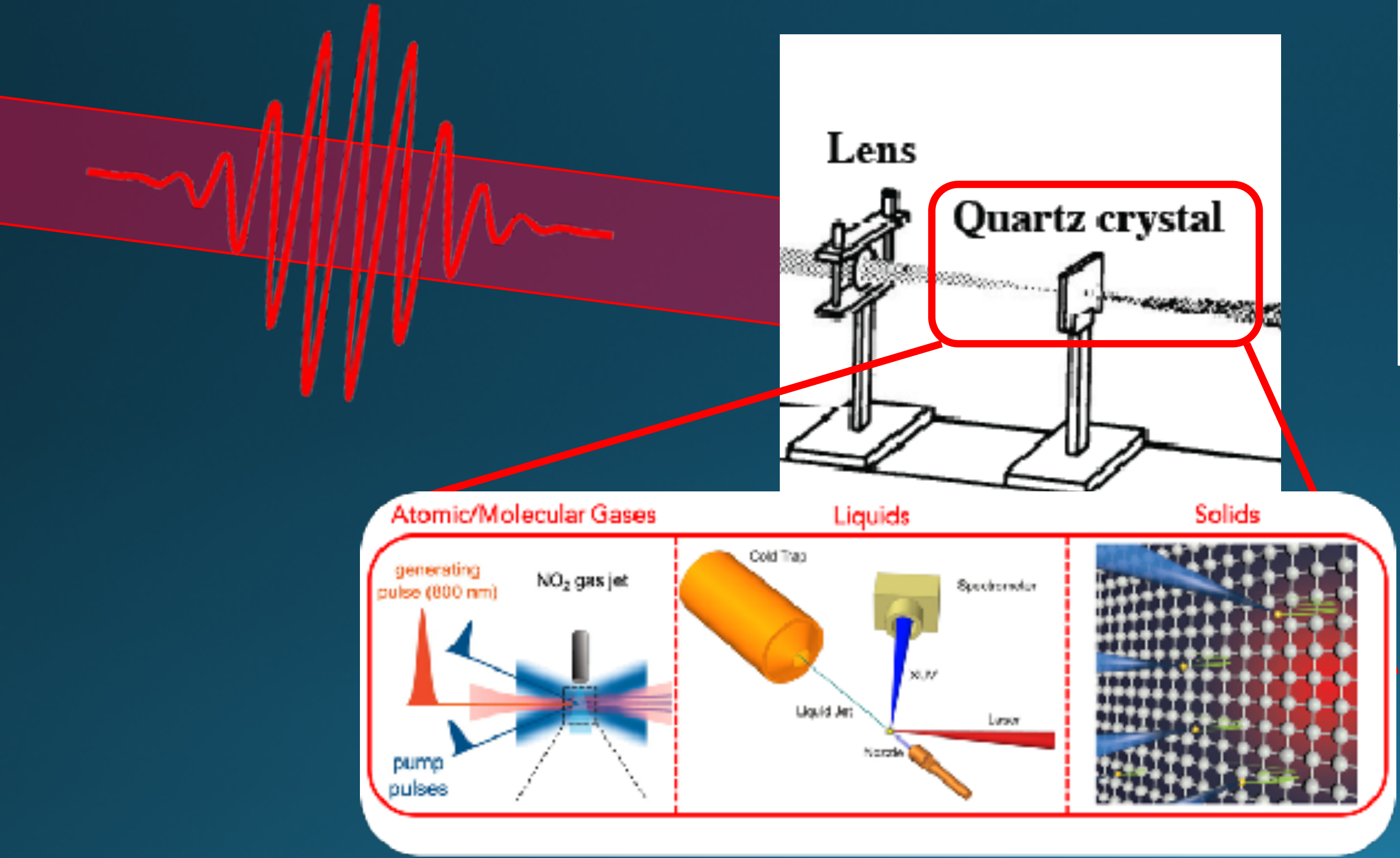

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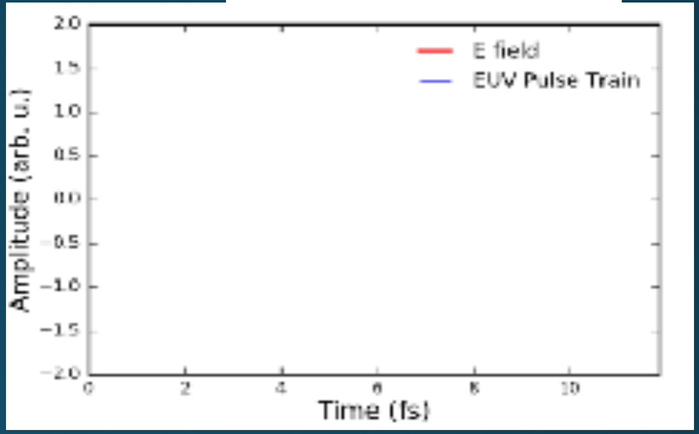

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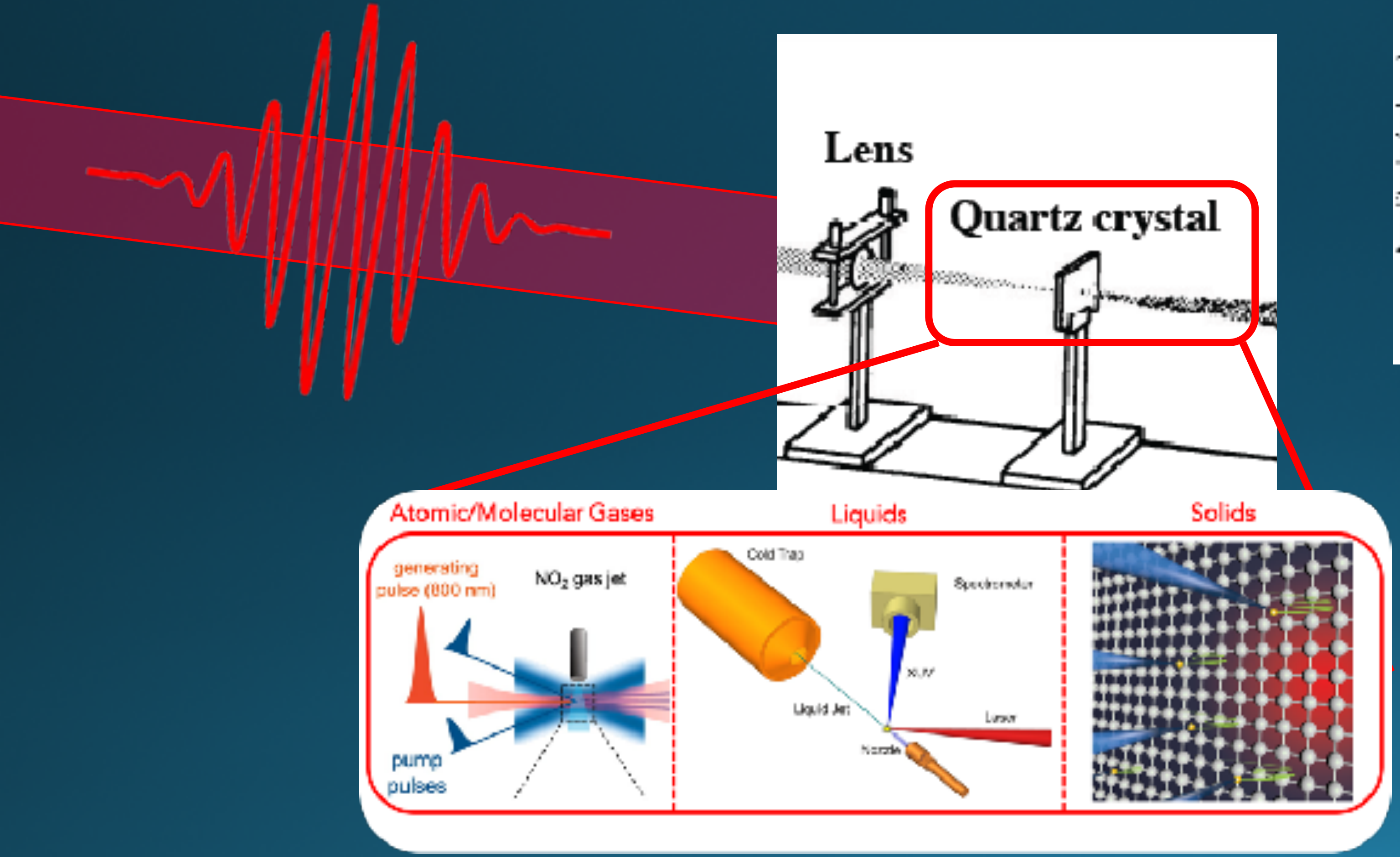

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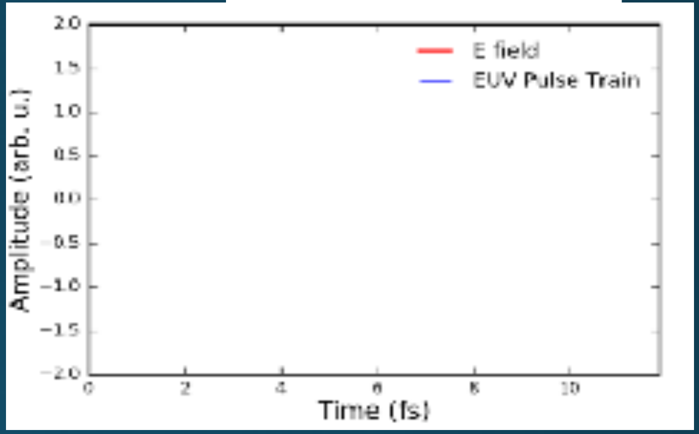
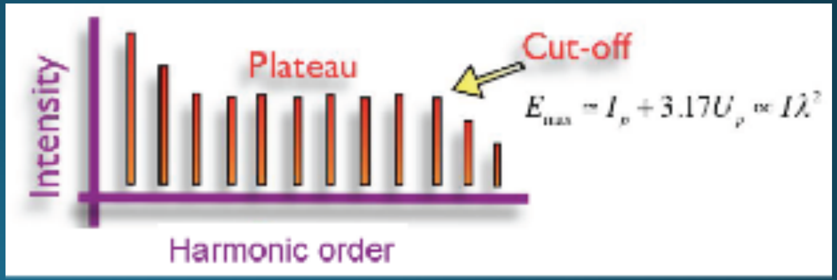
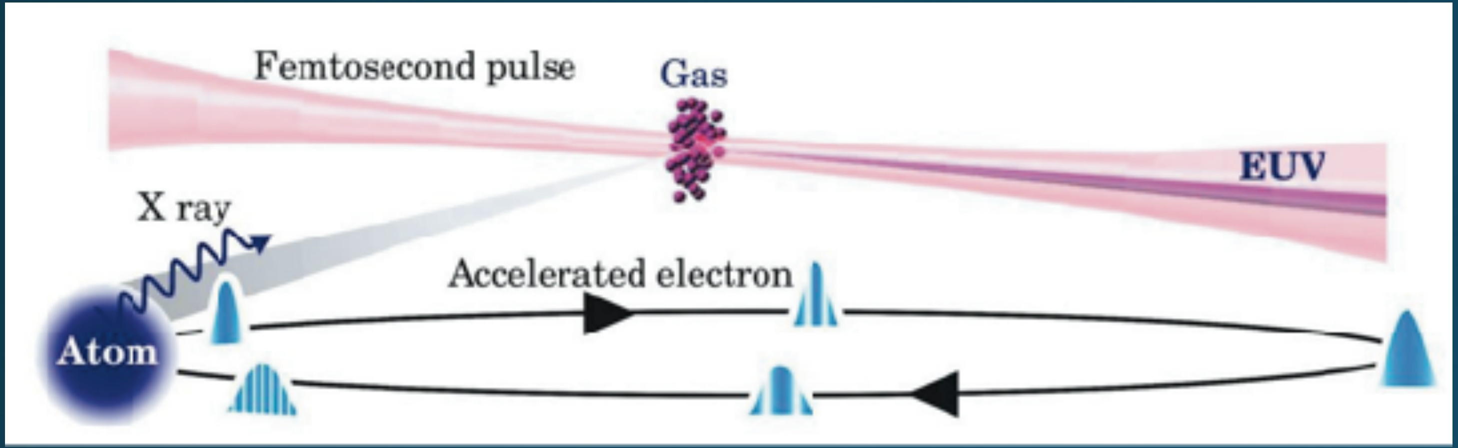


Frequency Domain



Microscopic Mechanism of HHG: Epitome of Classical Correspondence Principle

- High harmonic generation is the most extreme nonlinear process in nature.



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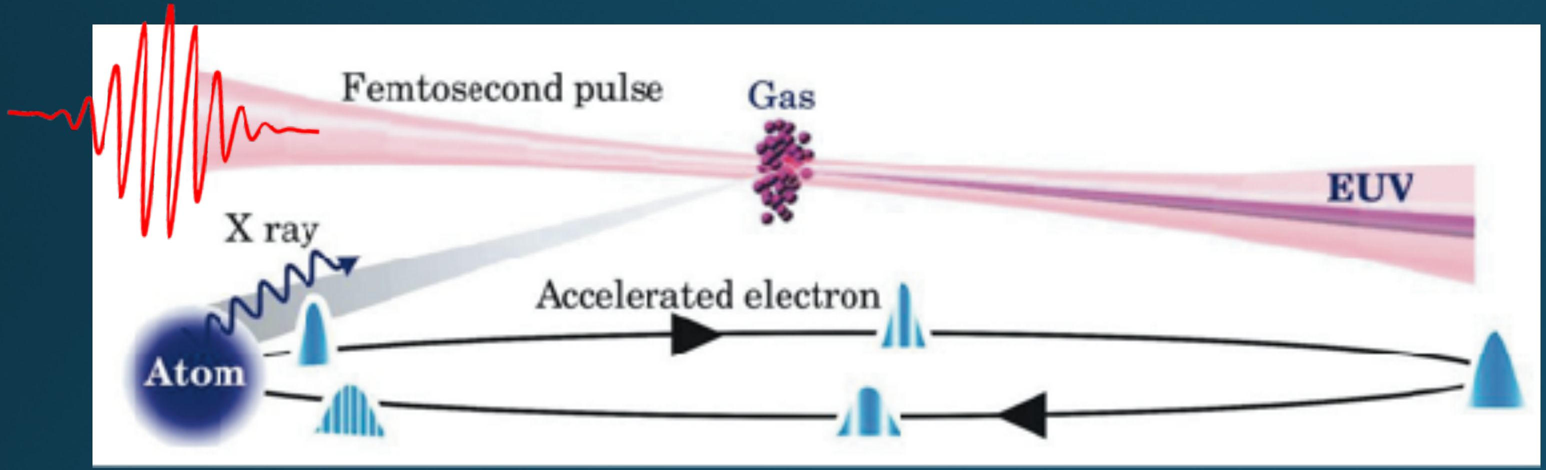

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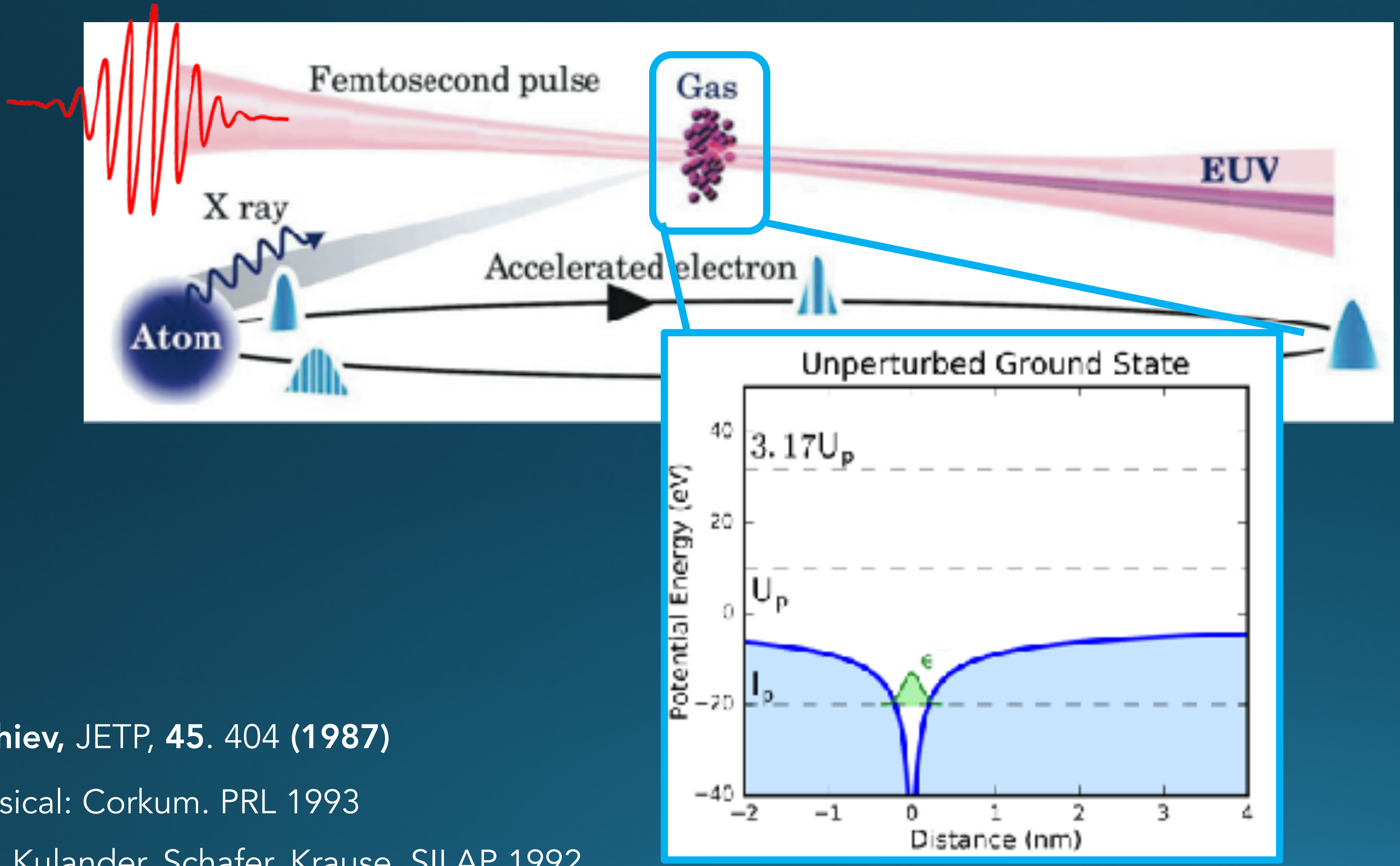

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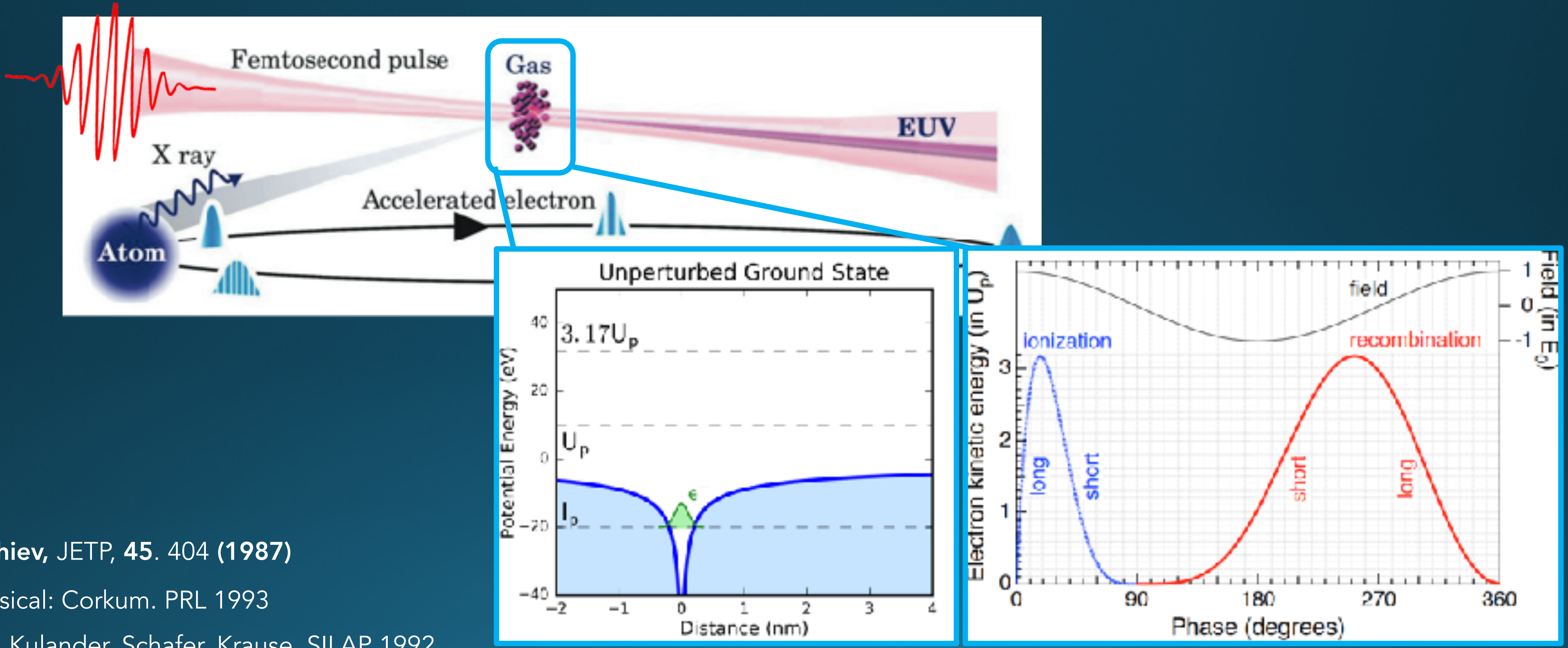

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Macroscopic HHG: Attosecond Nonlinear Optics in a K (Nut) Shell

- The generation of bright, coherent beams of X-ray light demands that we solve the currency exchange problem inherent to nonlinear optics.
- Single atom yield ~ $\lambda^{-6.5}$

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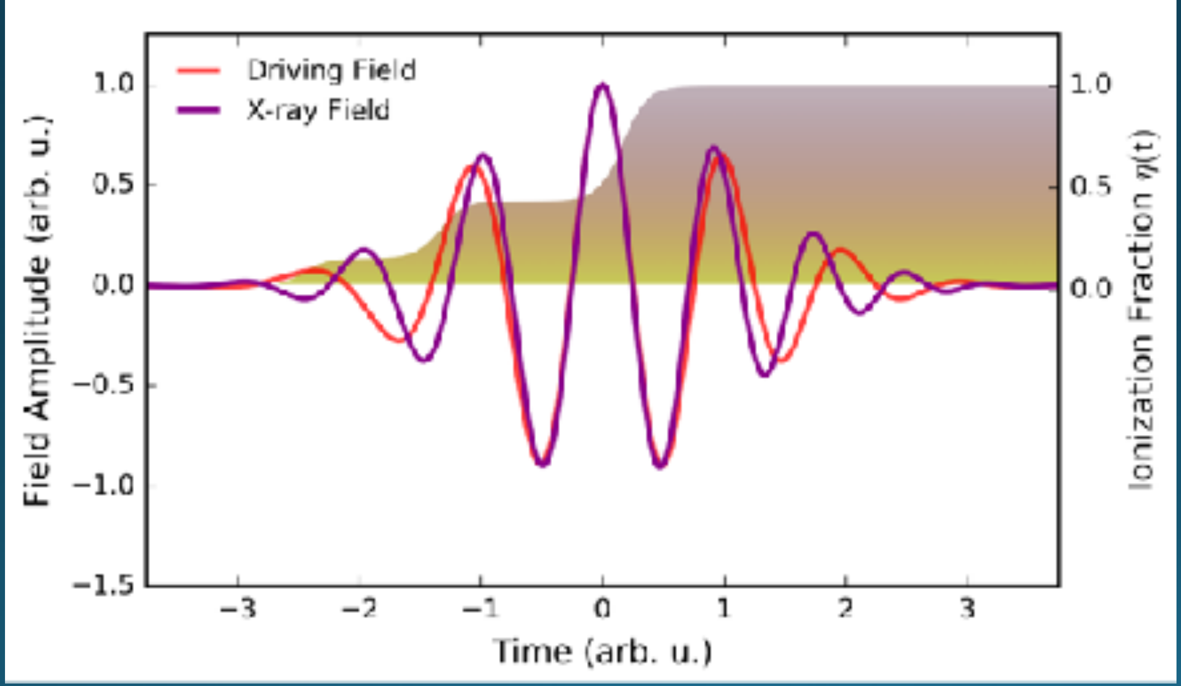

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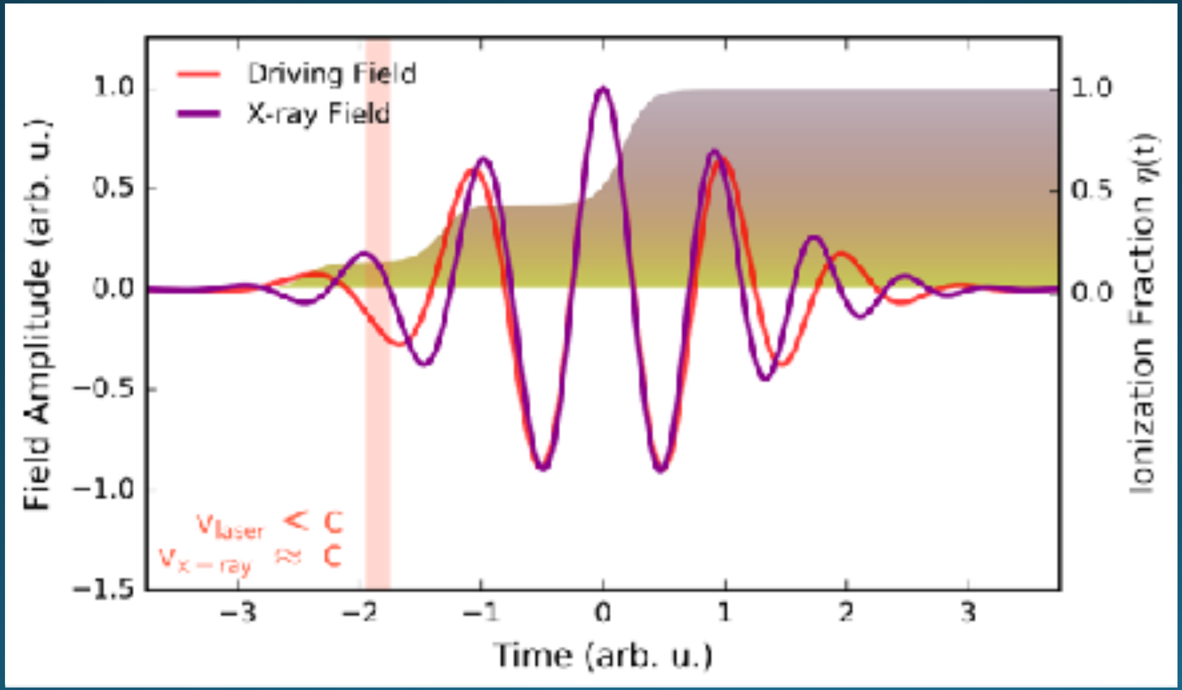

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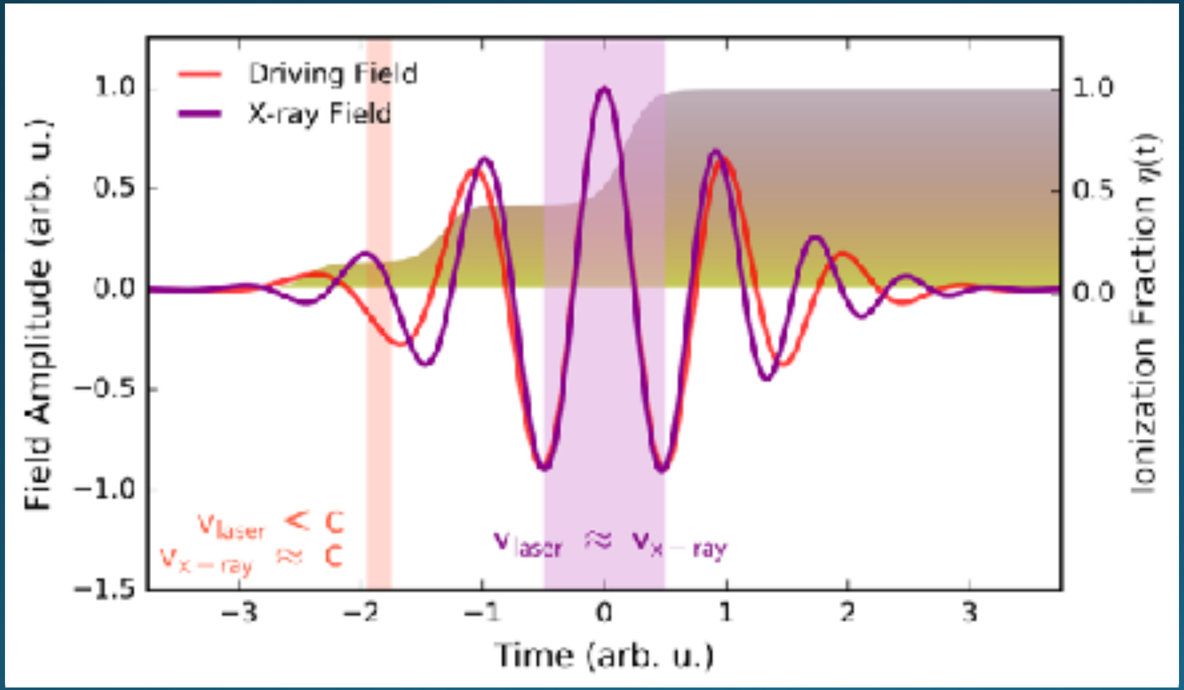

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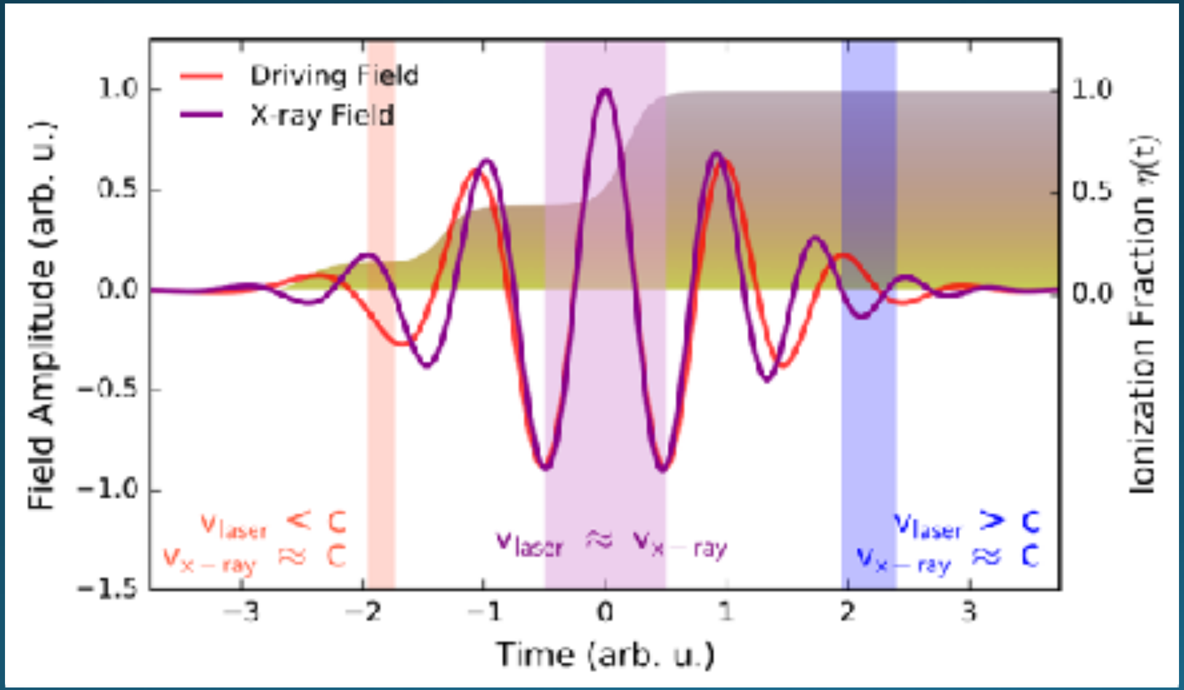

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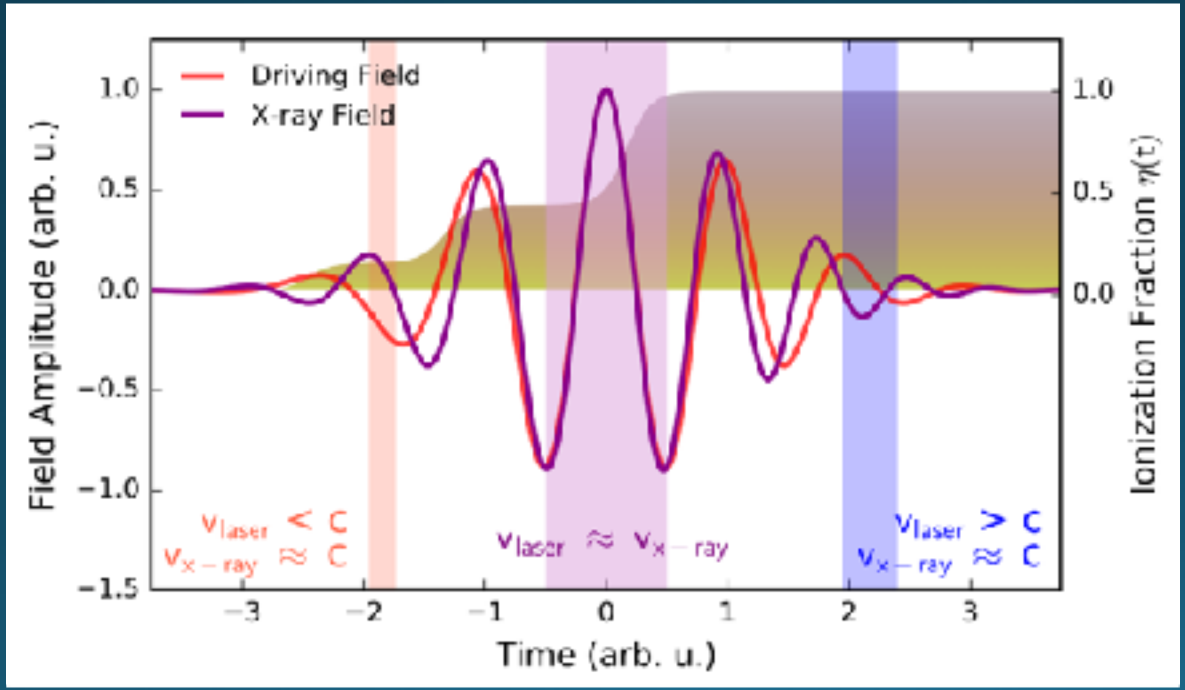

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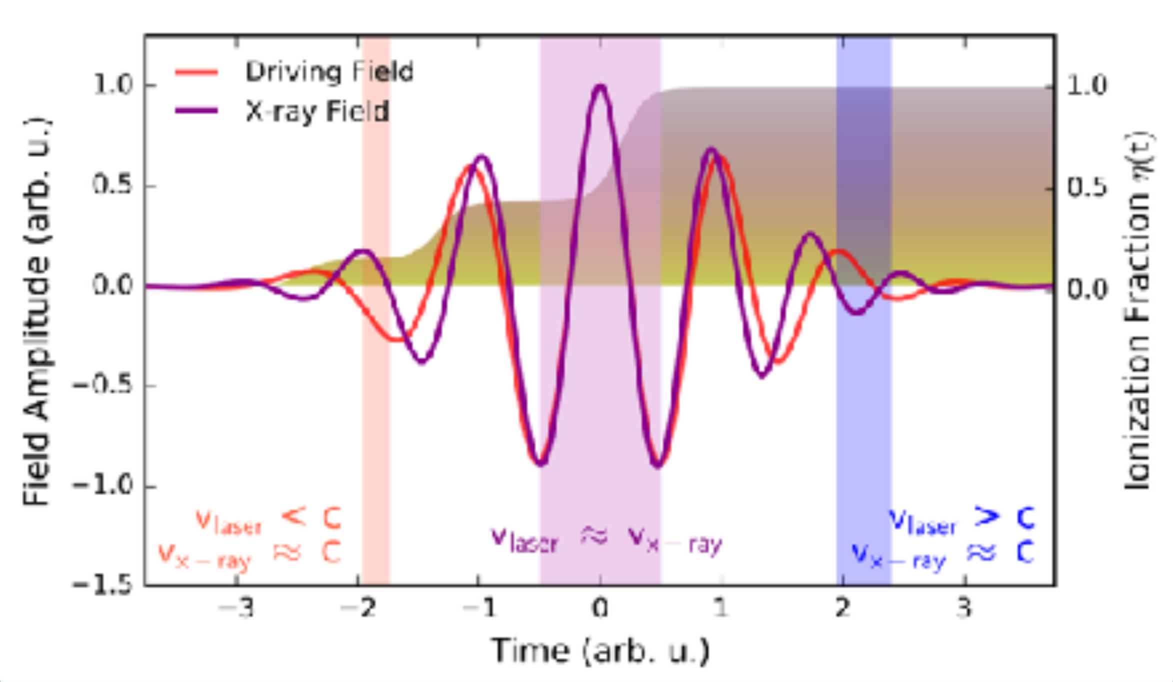

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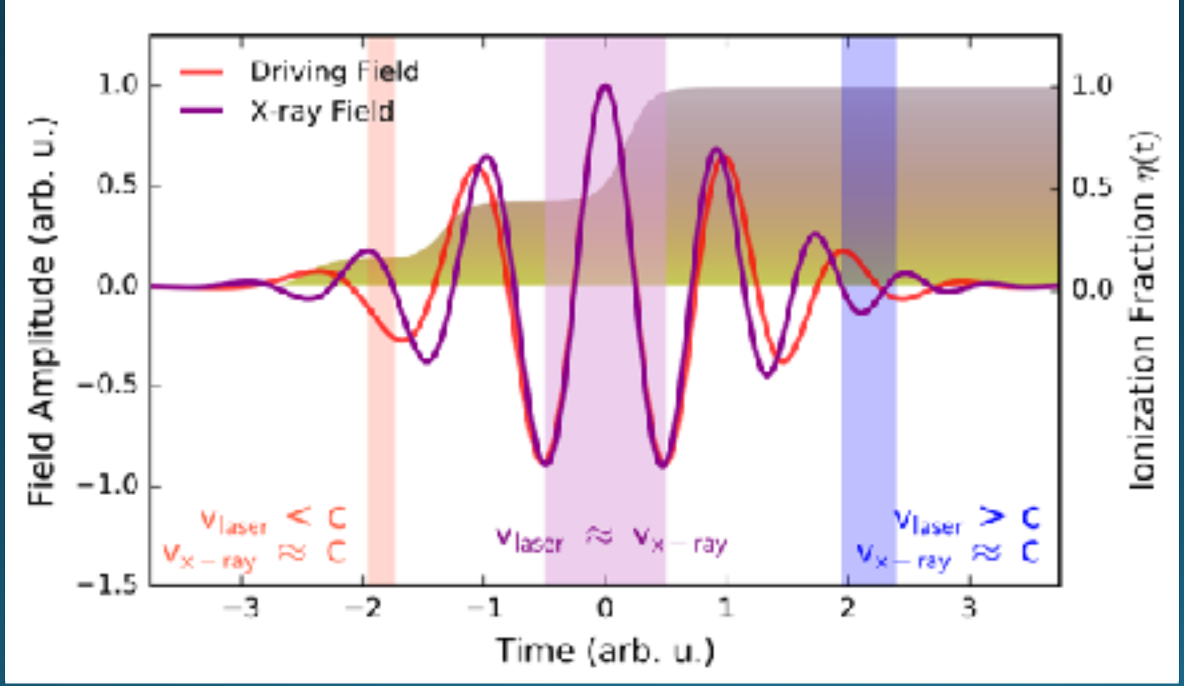

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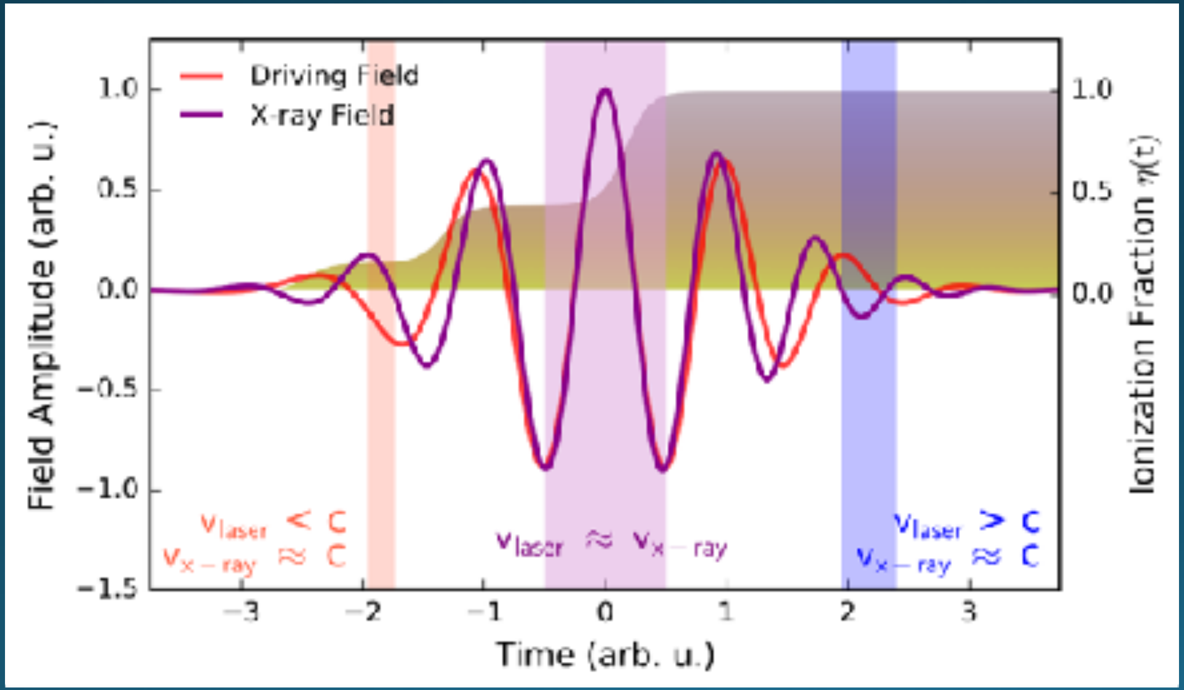

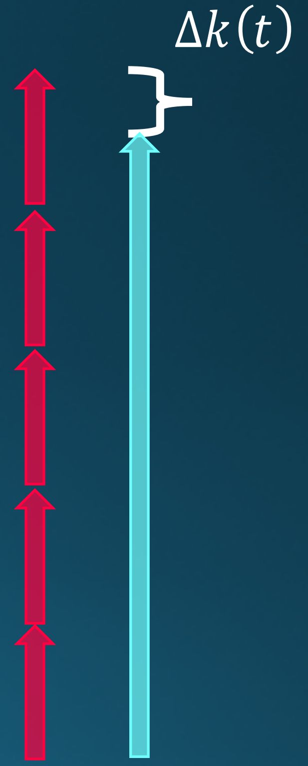

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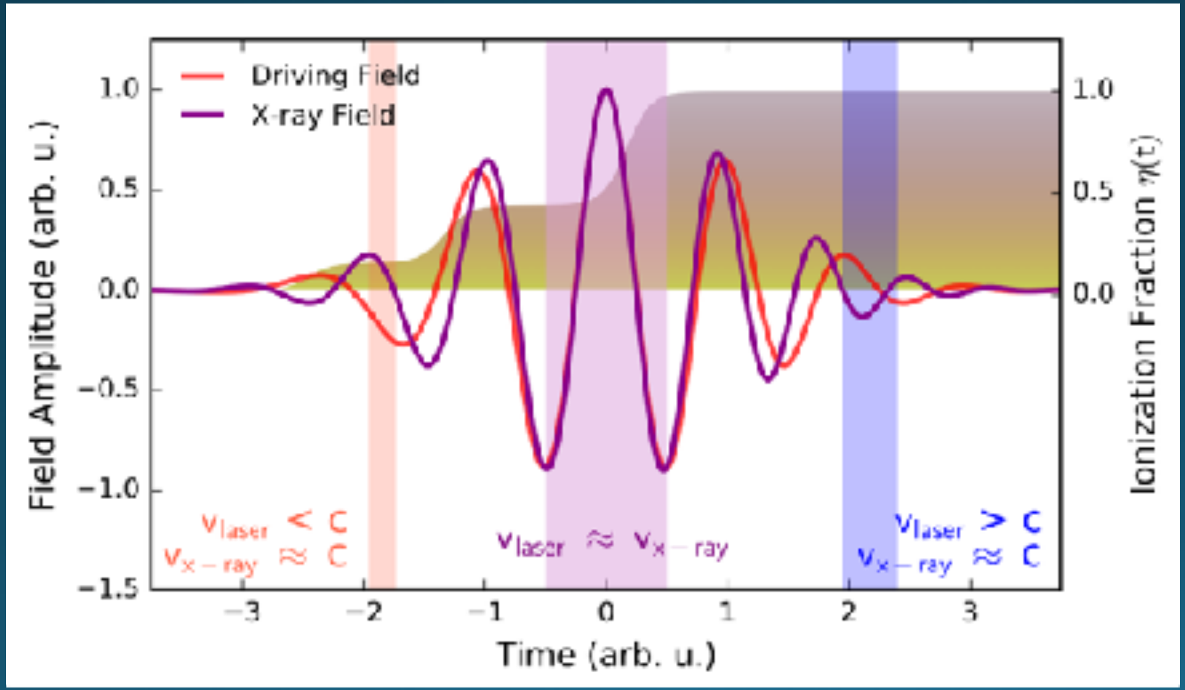

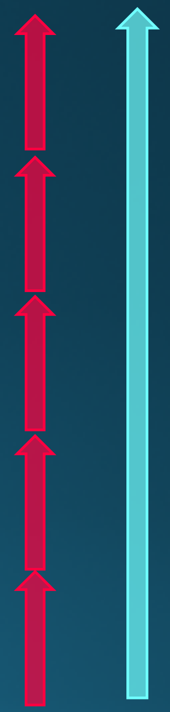

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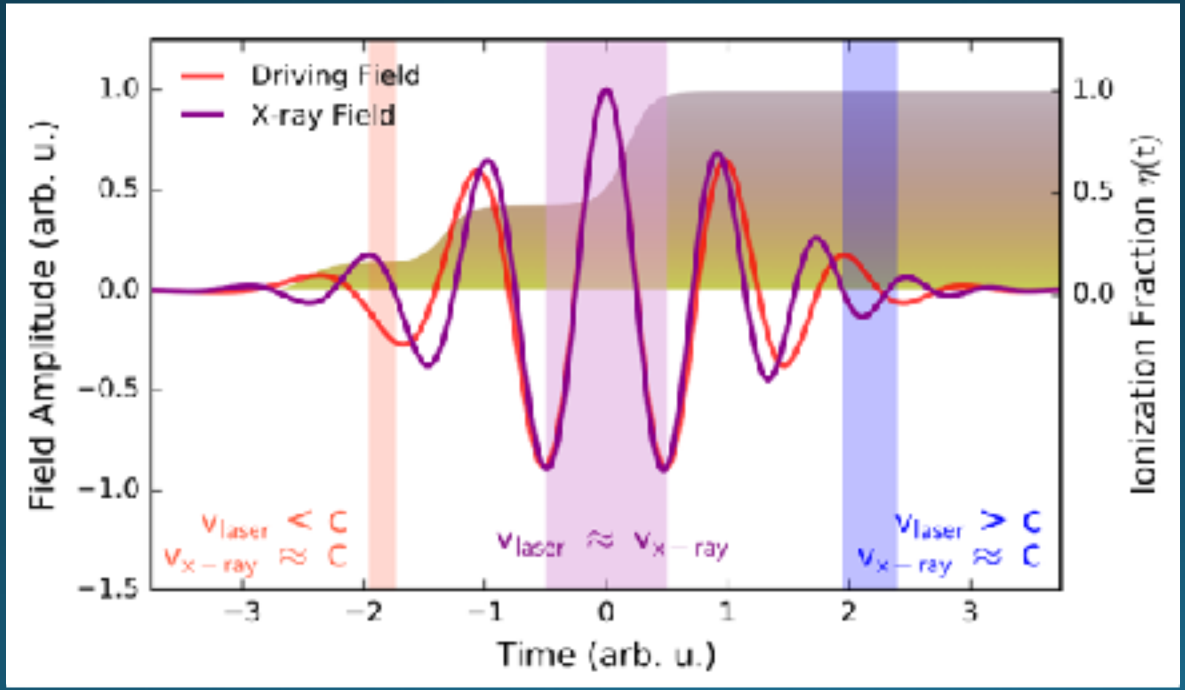

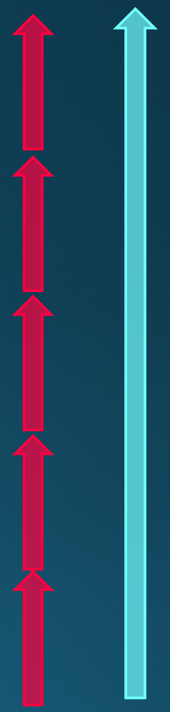

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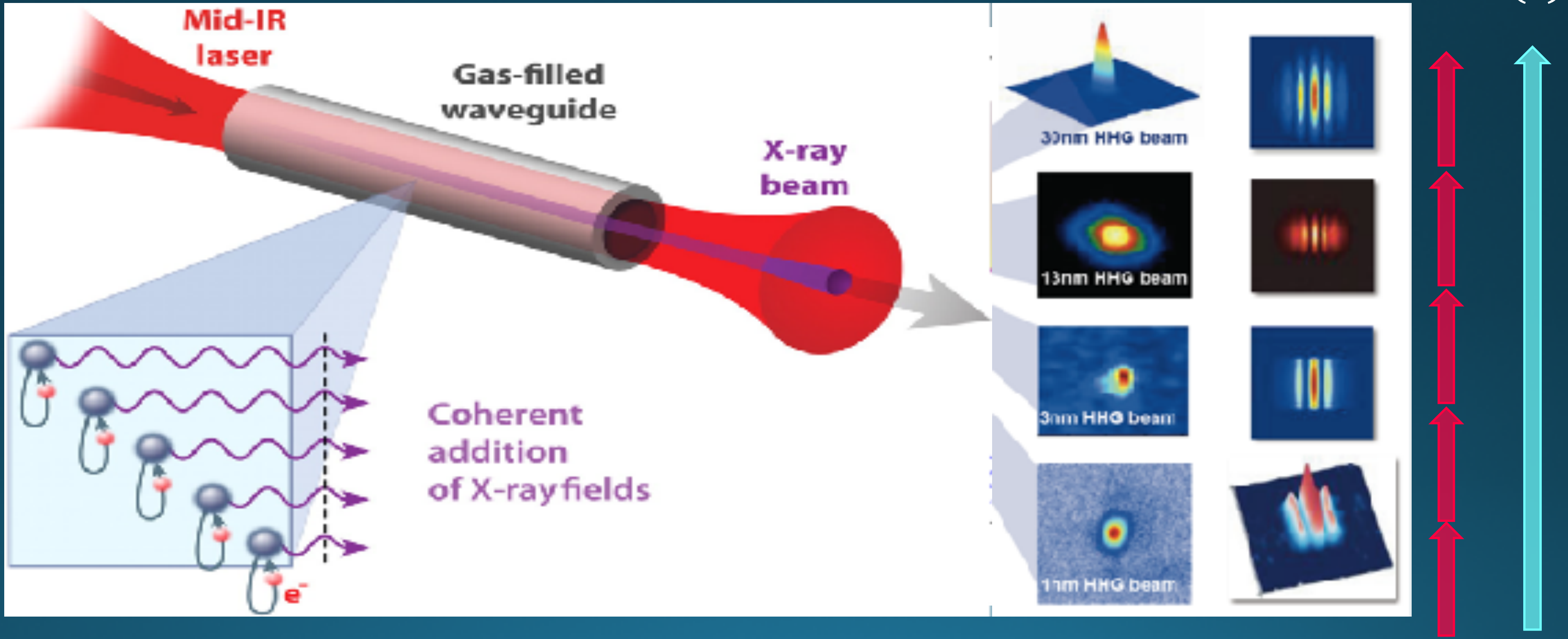

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Dancing to the Quantum Tune: Using Macroscopic Control to Direct HHG Emission

Courtesy Carlos Hernandez-Garcia, Universidad de Salamanca

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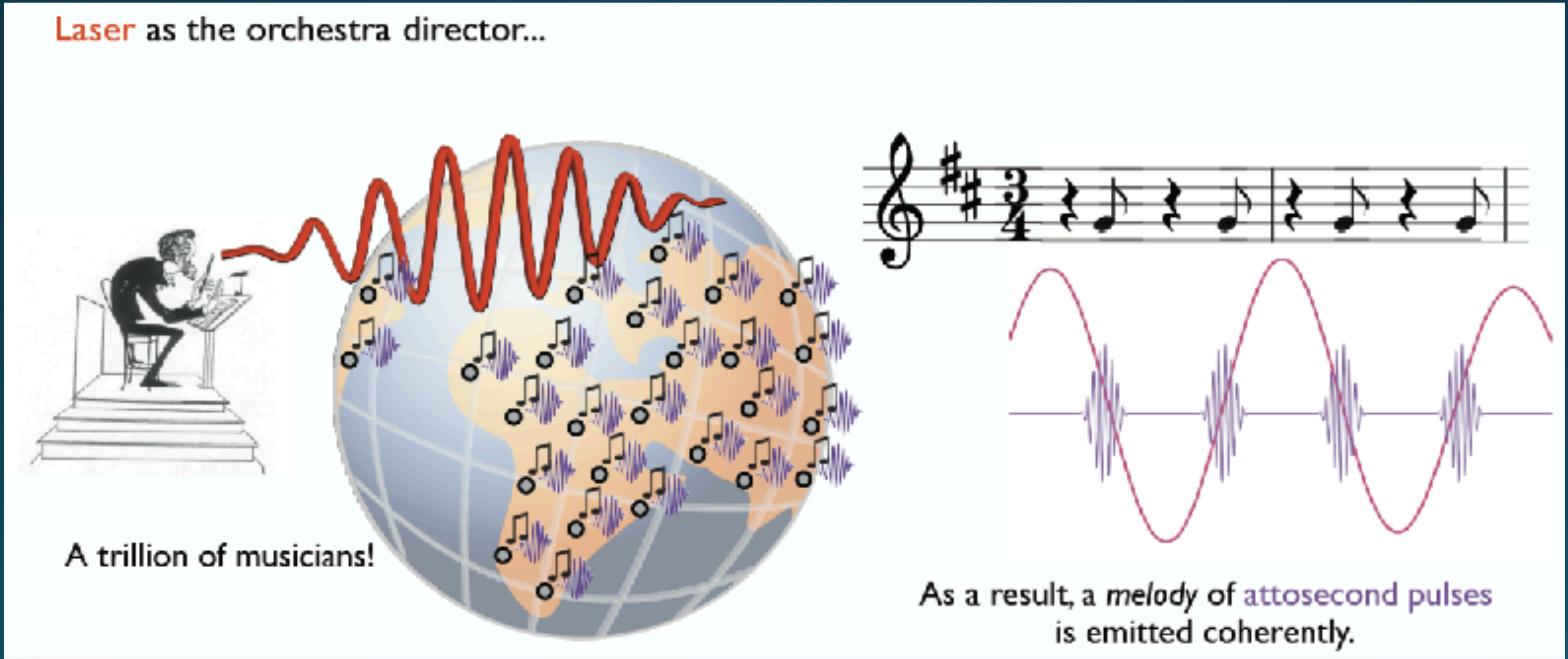

Custom Tailored EUV and X-ray Light From a Table-Top: Exquisite Control Over the Entire Up-Conversion Process

Chen, PNAS, 111, 2014
Hernandez-Garcia, Opt Exp, 106, 2017
Popmintchev, Science, 6086, 2012
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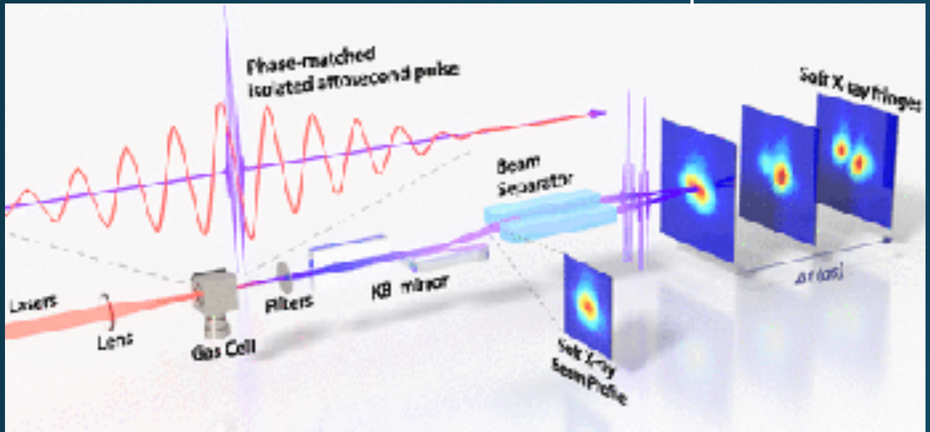
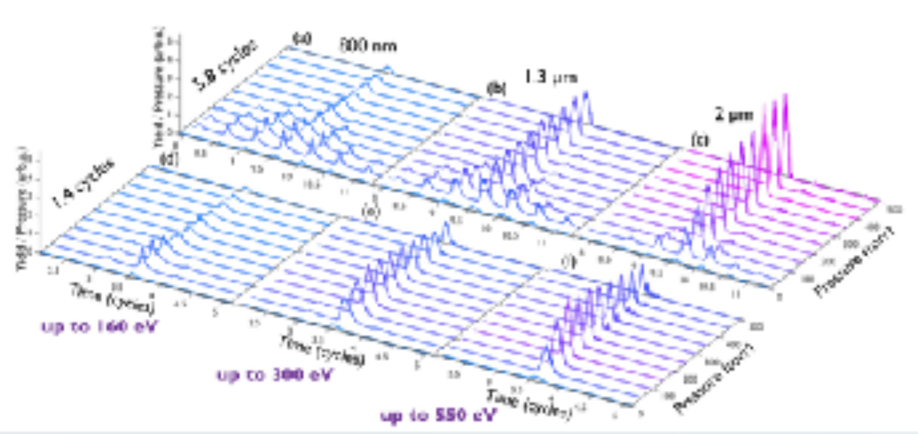
Phase-matched isolated as pulses





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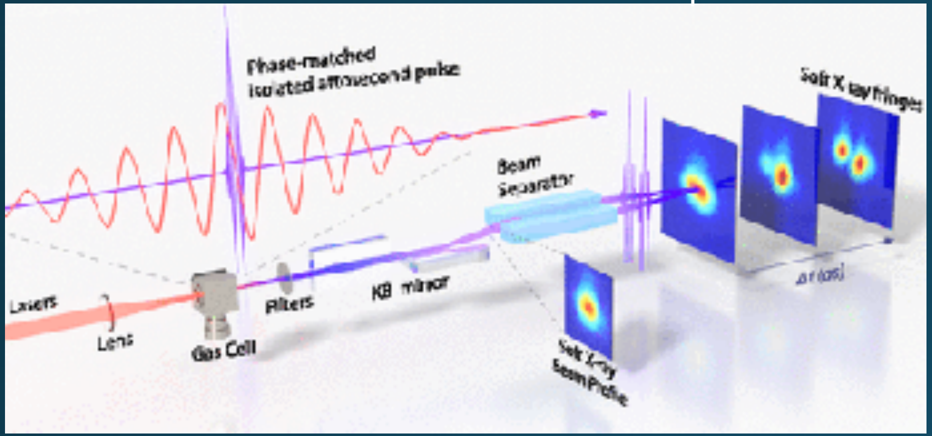
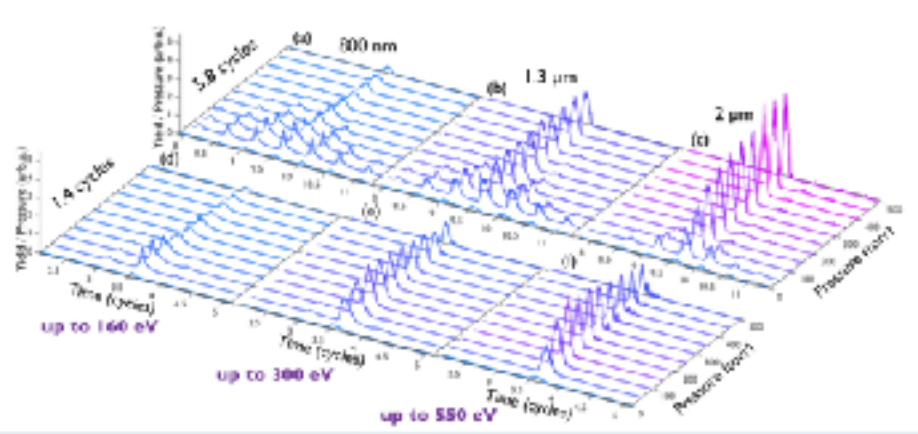
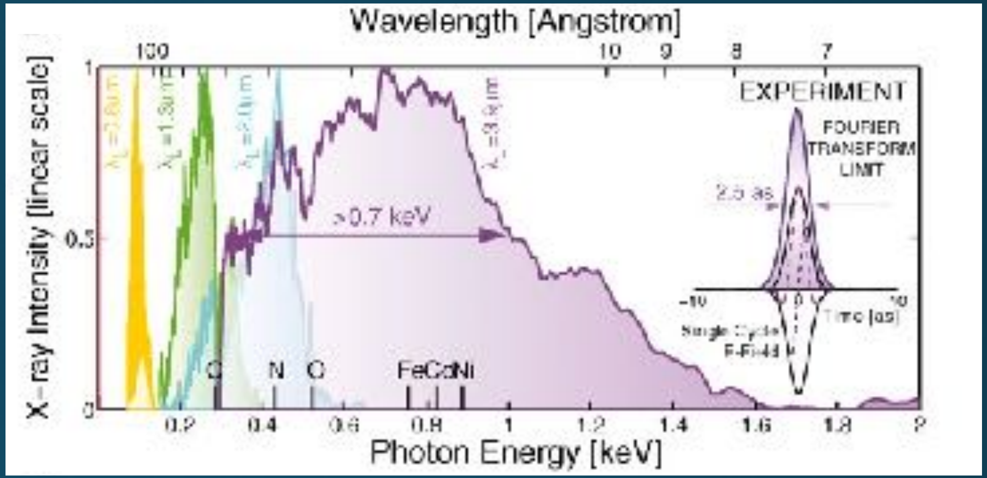




Coherent, zeptosecond x-rays



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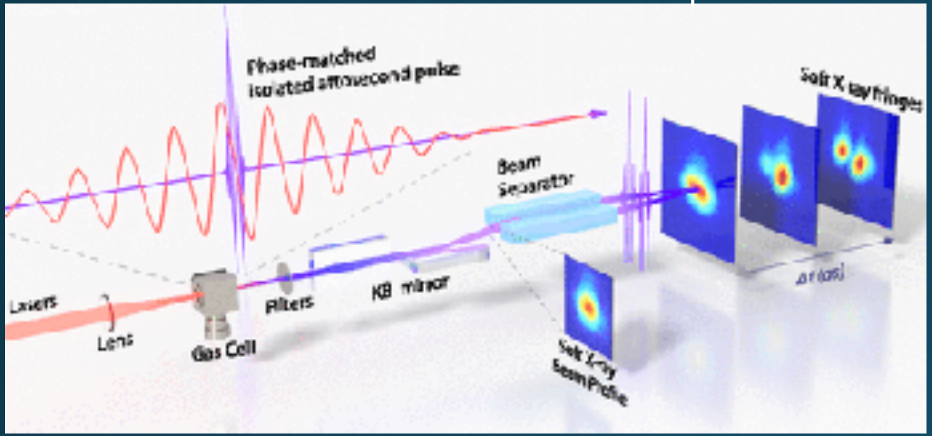

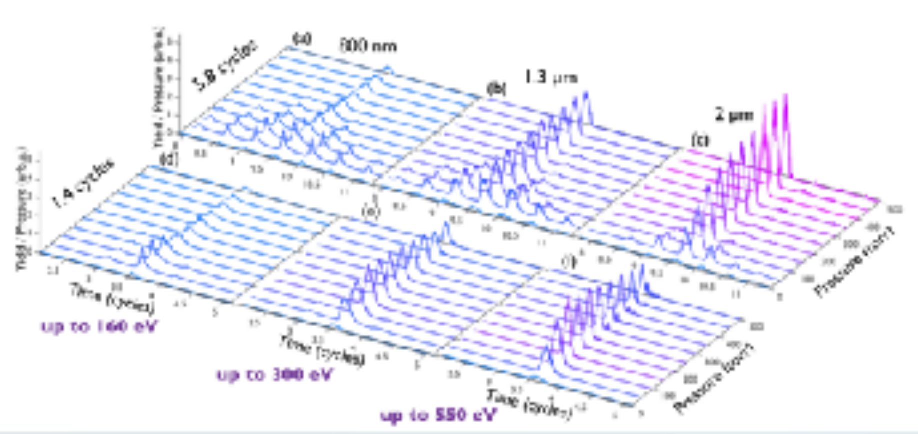

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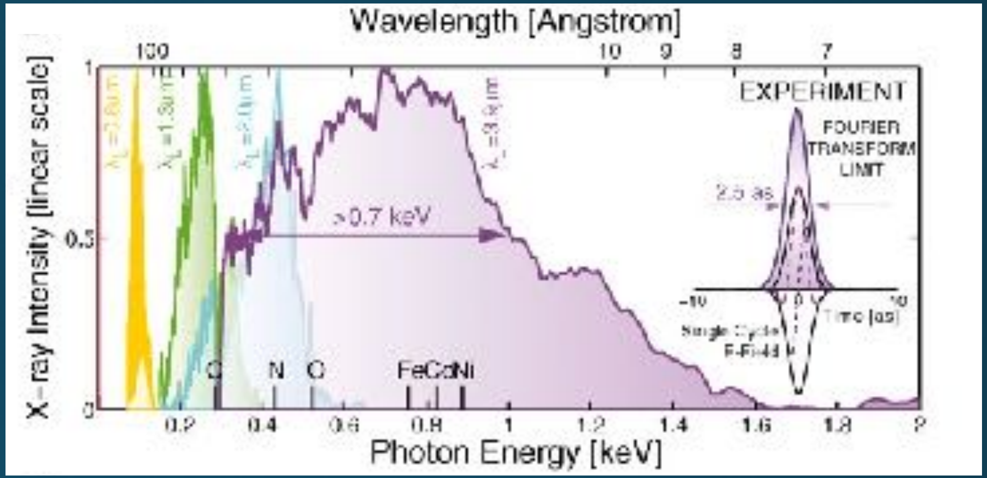
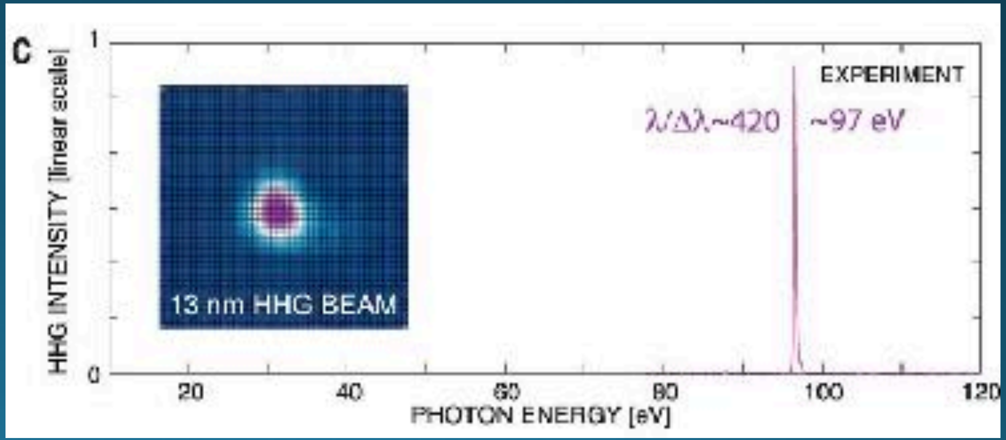


Bright, isolated harmonics



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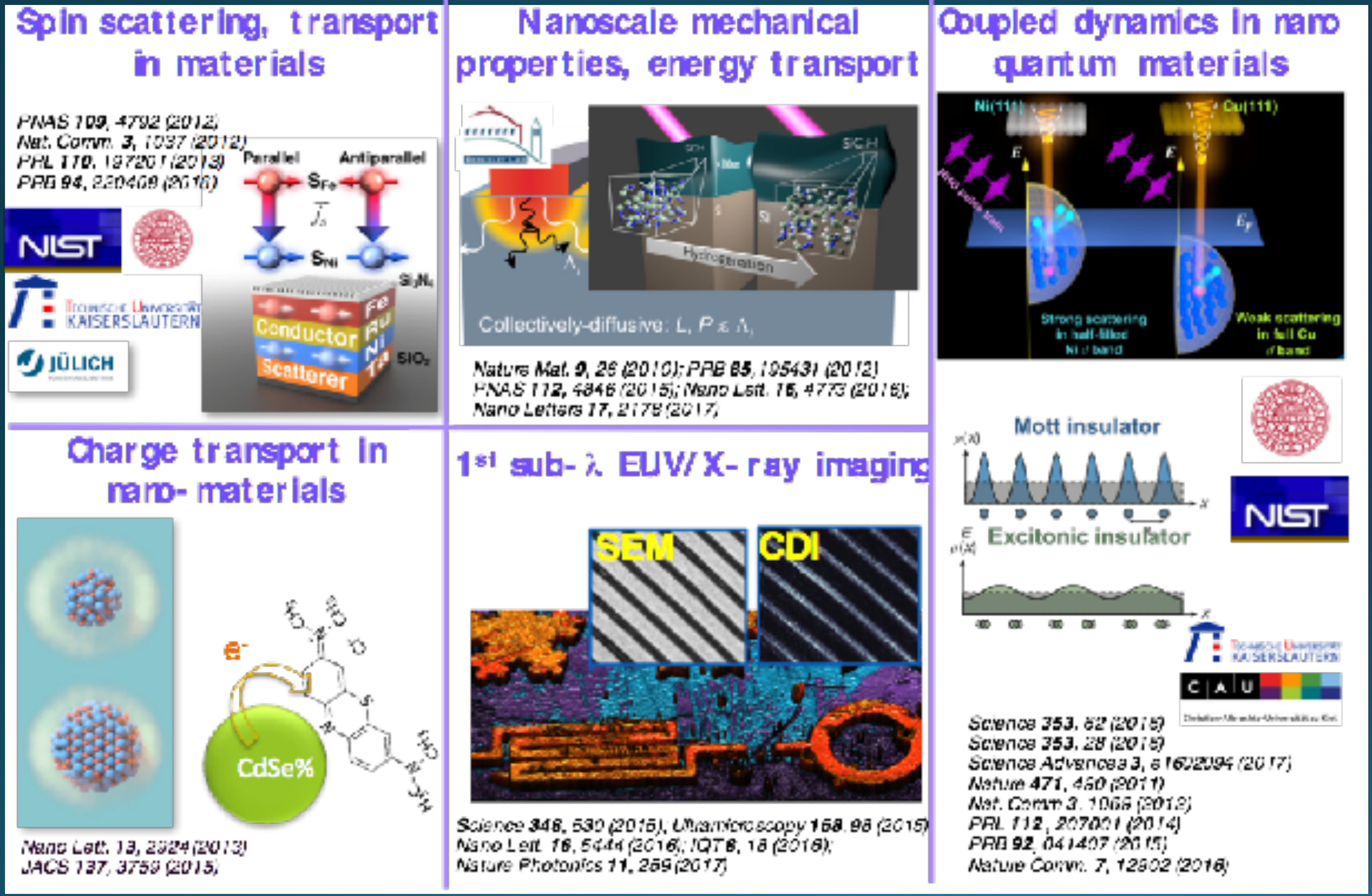
Tailored HHG Waveforms: Uncovering New Material Science Nano to Atto



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

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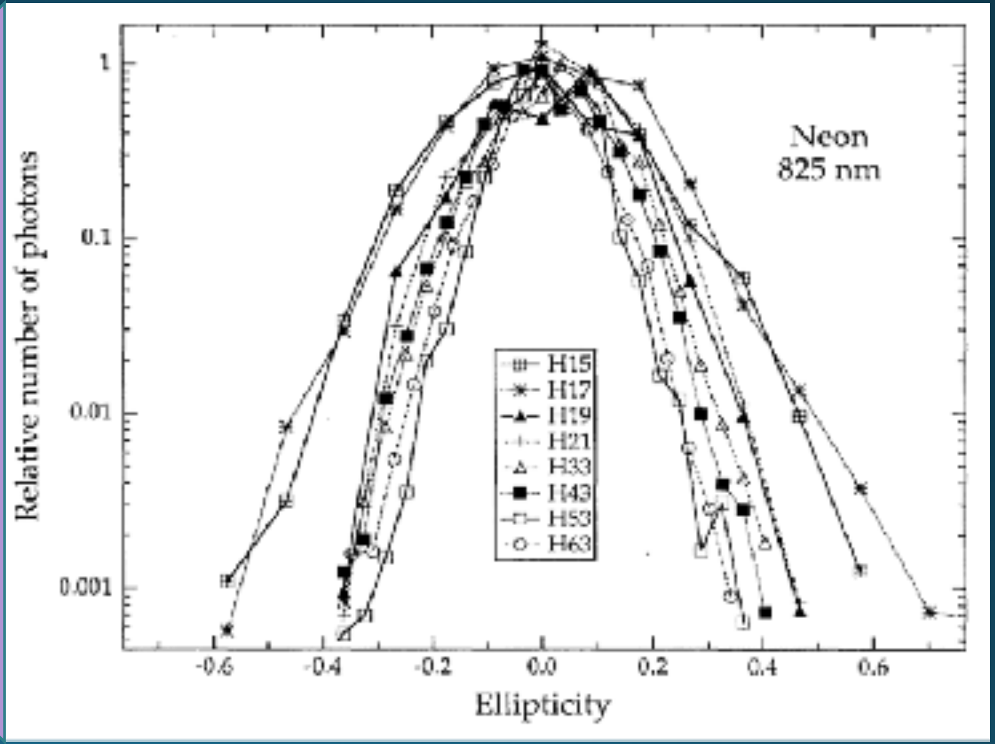

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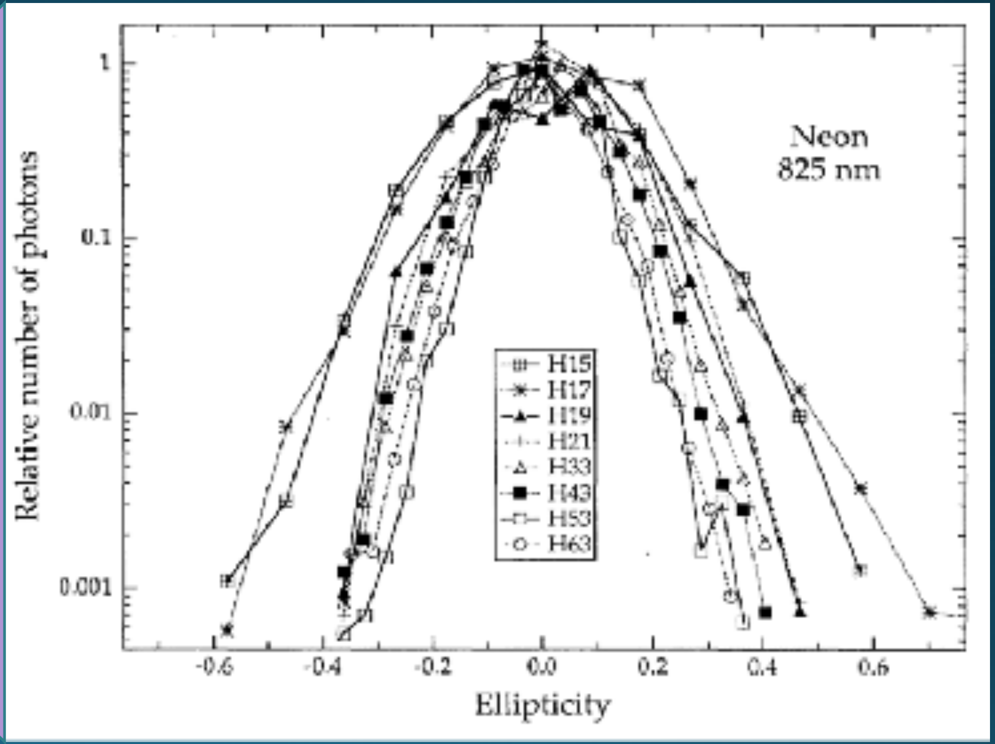

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Reshaping the Frontier of Attosecond Science

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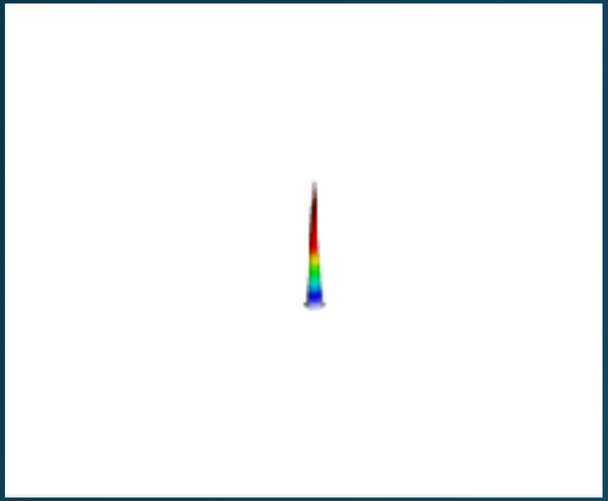

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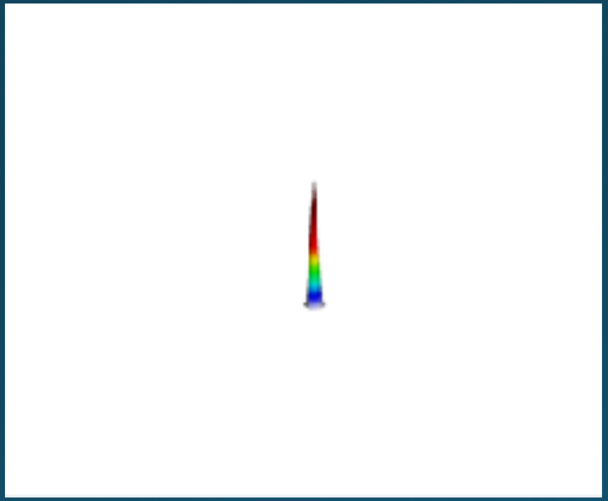
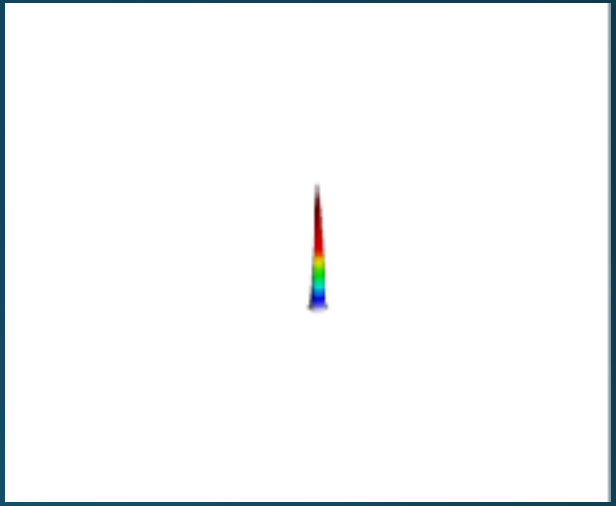


Two-Color Counter-Rotating



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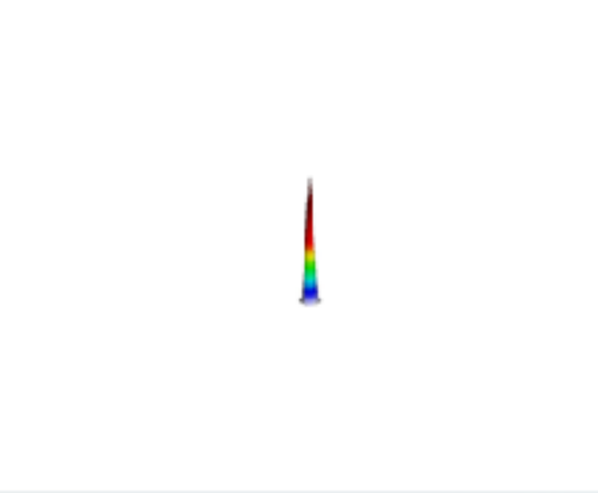

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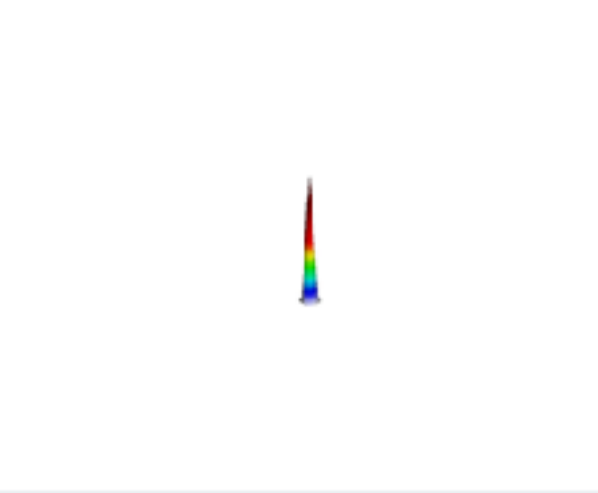

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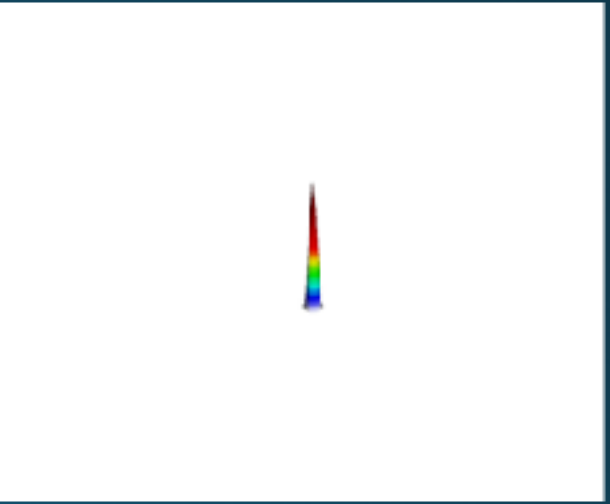

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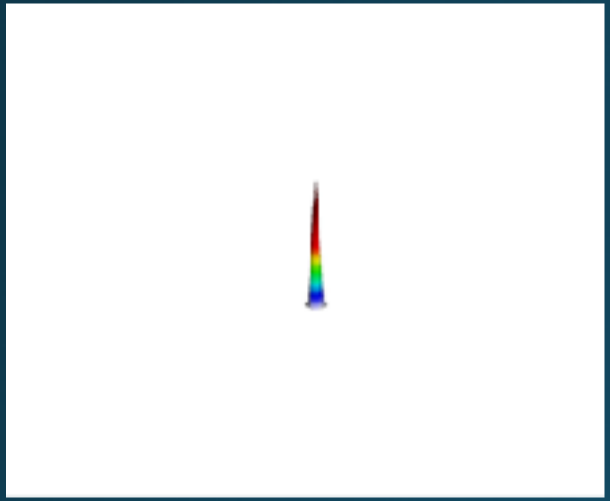

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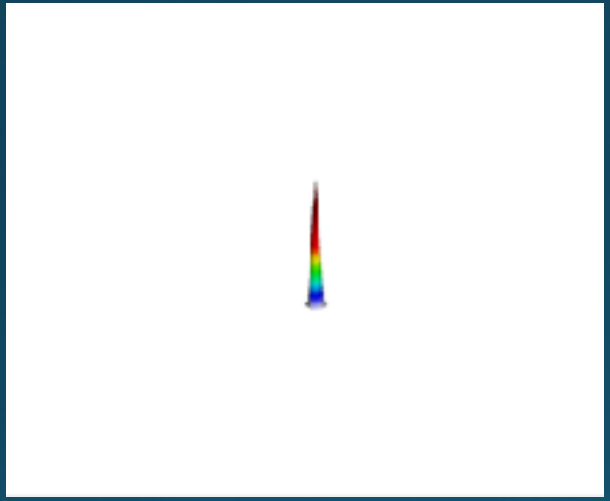

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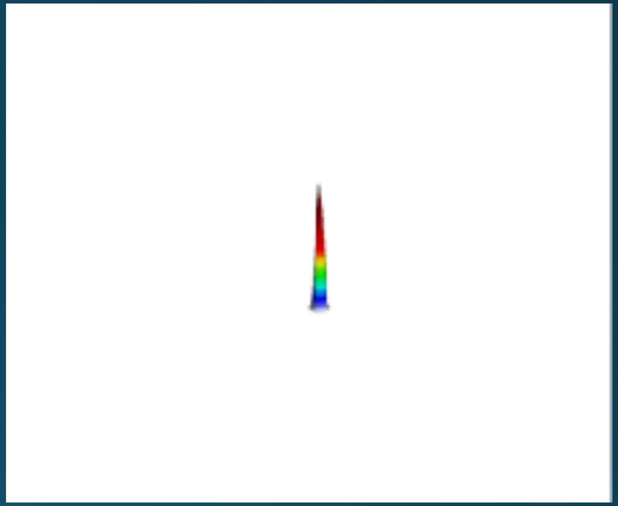

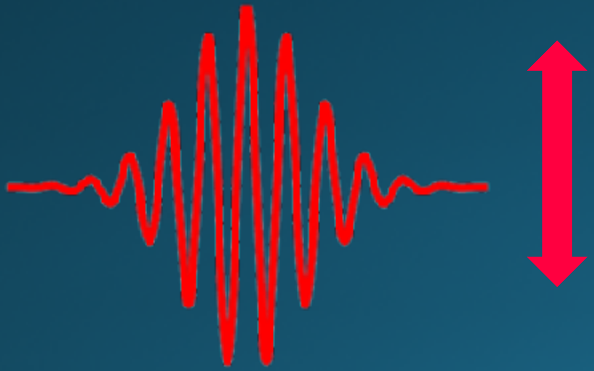

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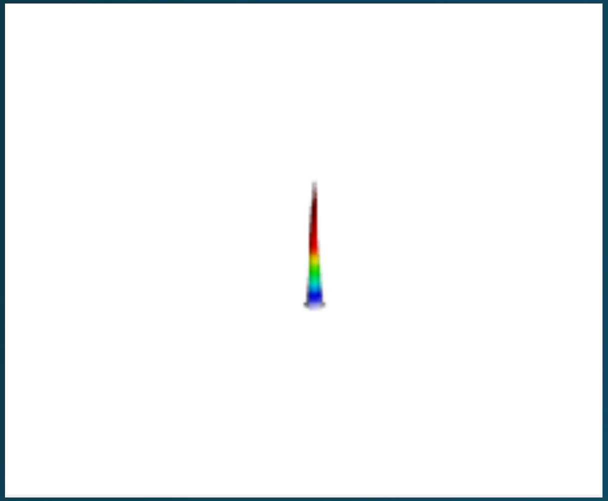

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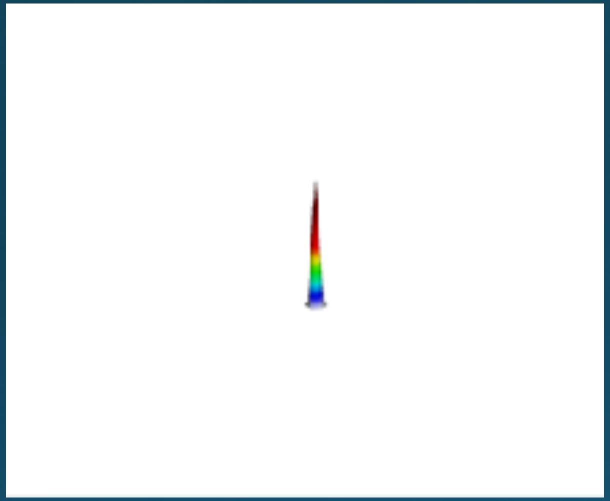

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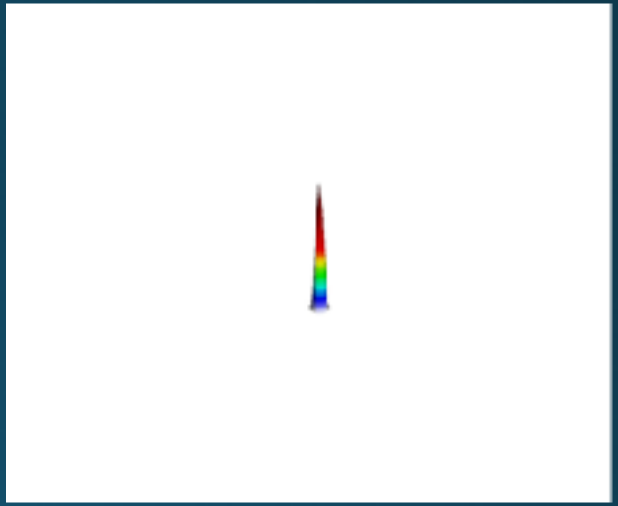

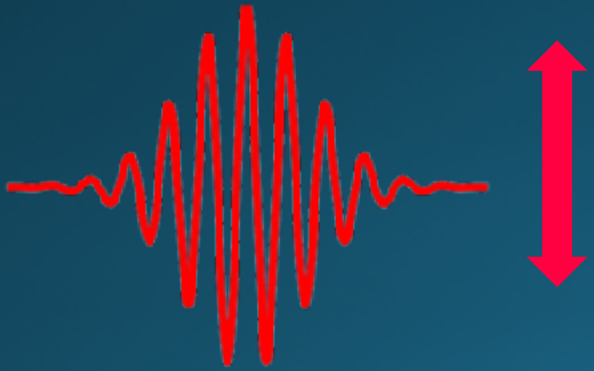

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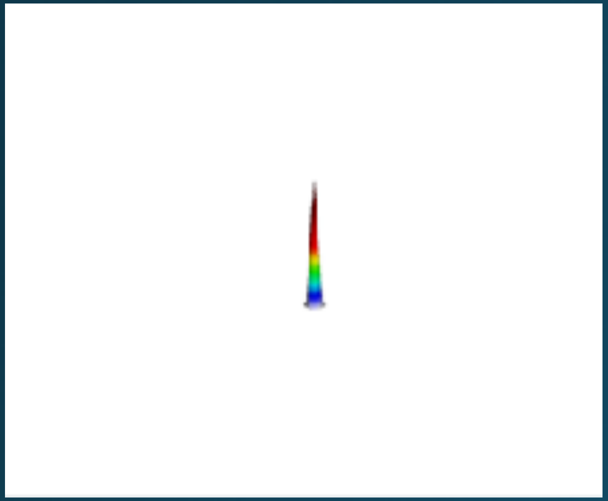

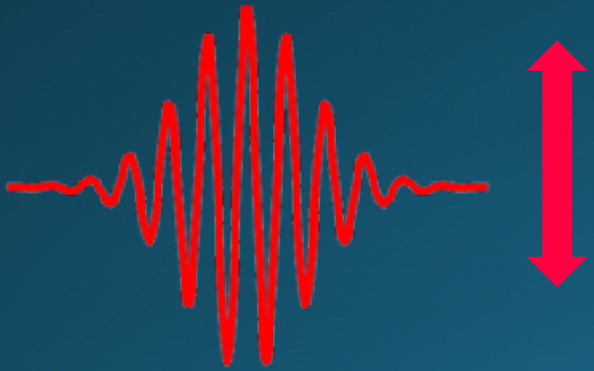

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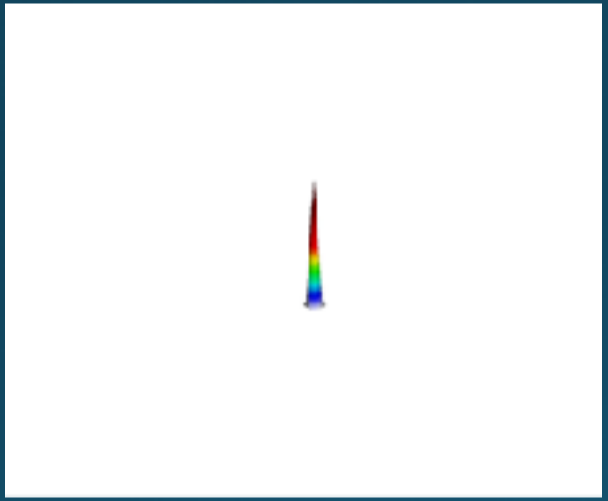

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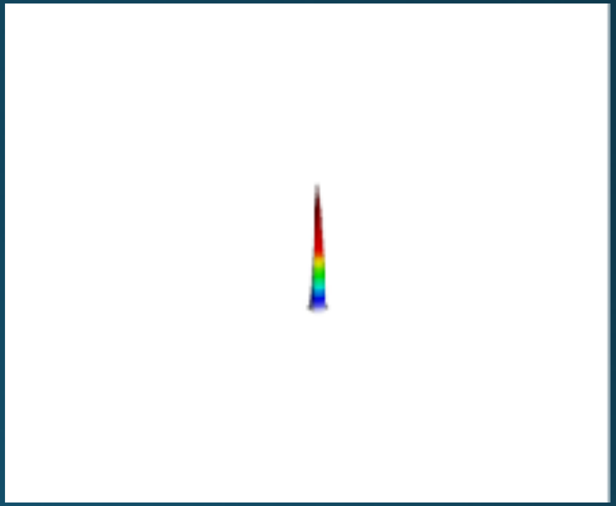

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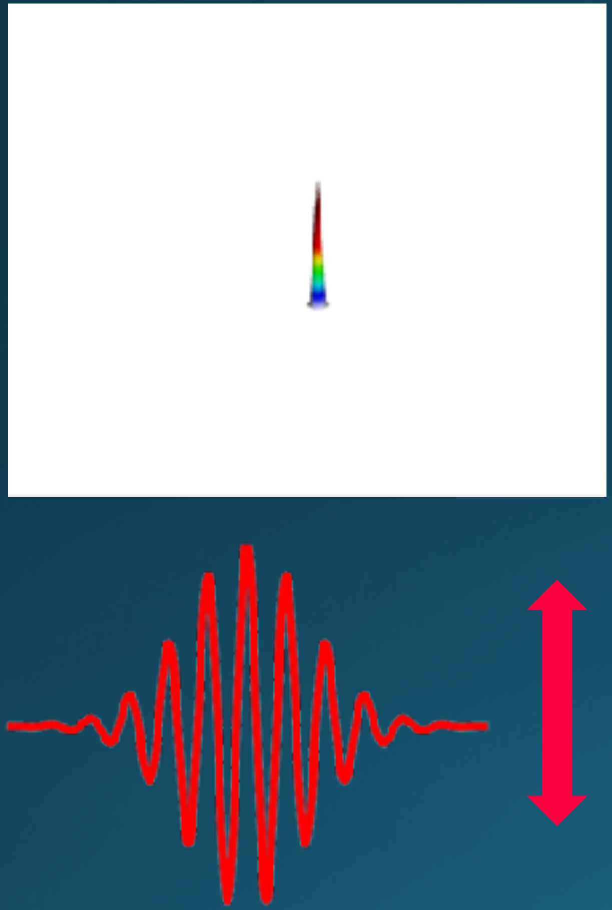

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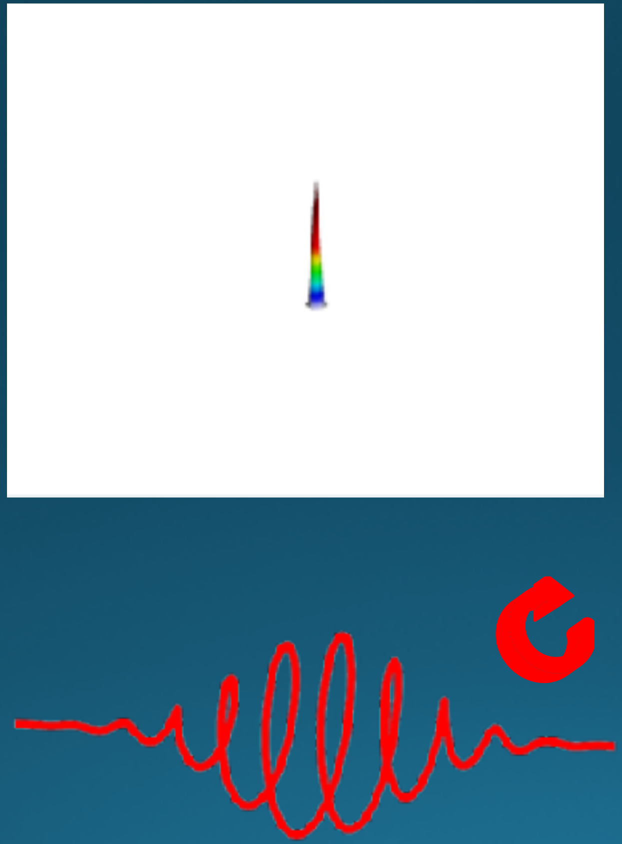

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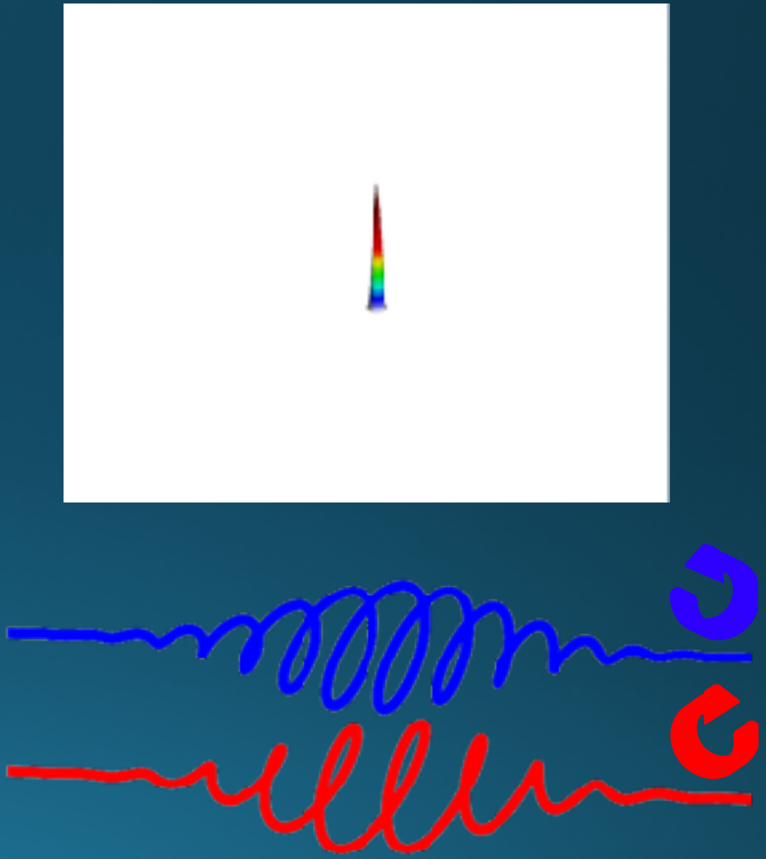

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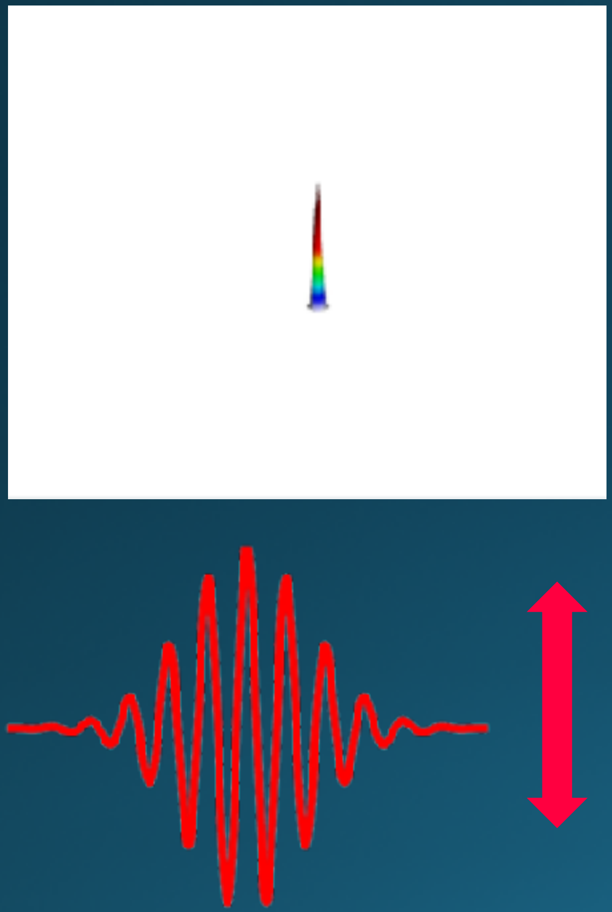

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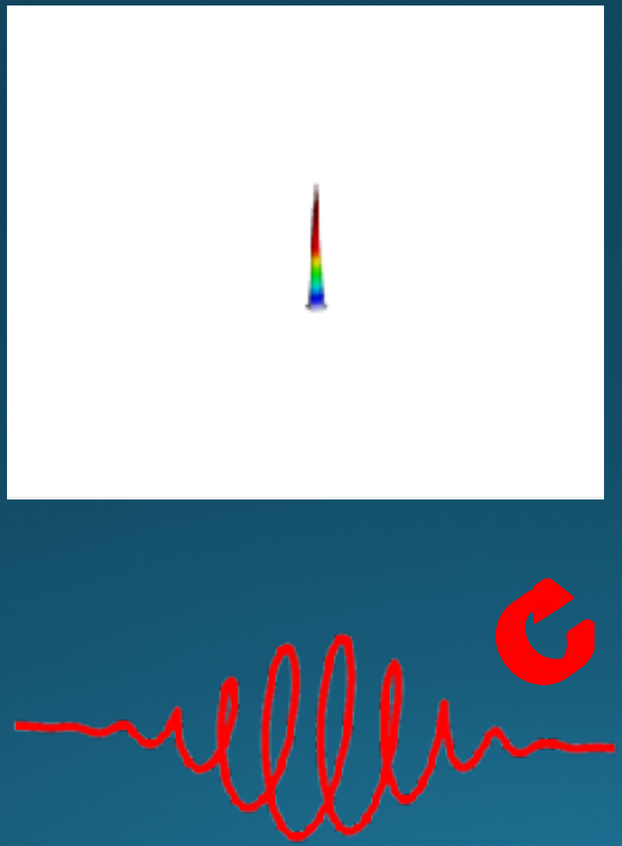

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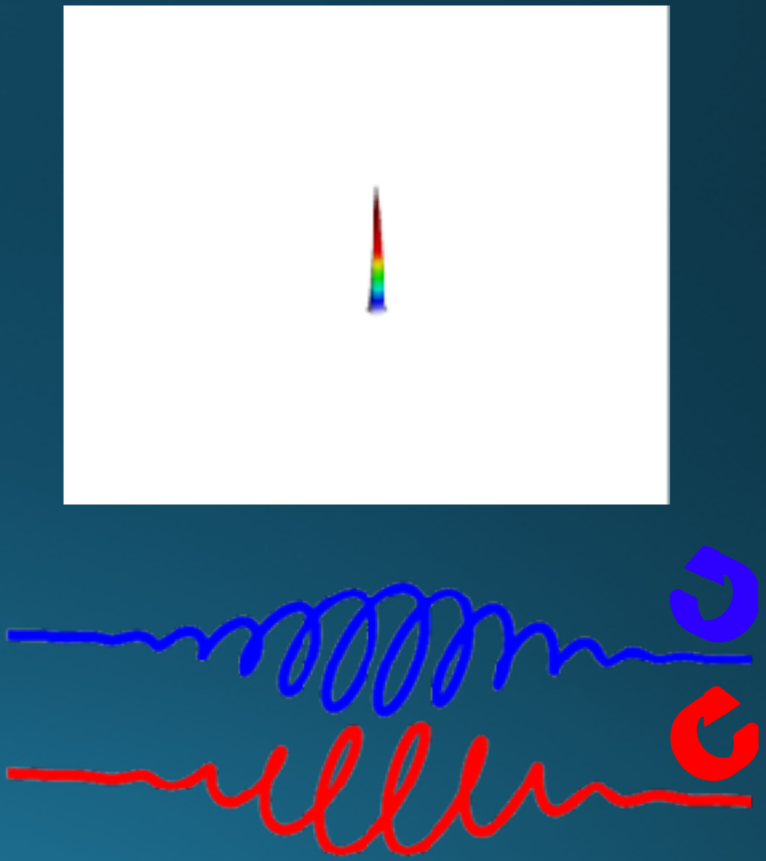

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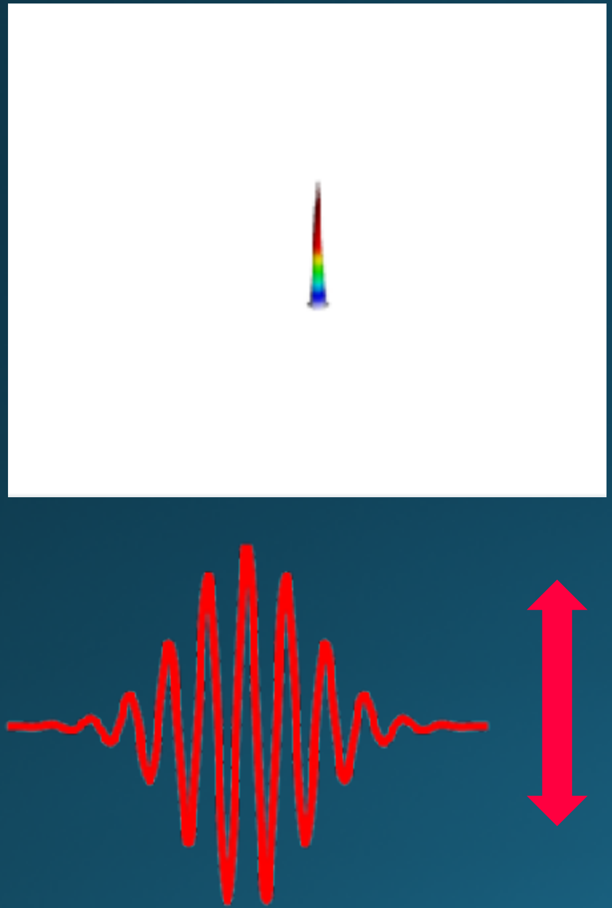

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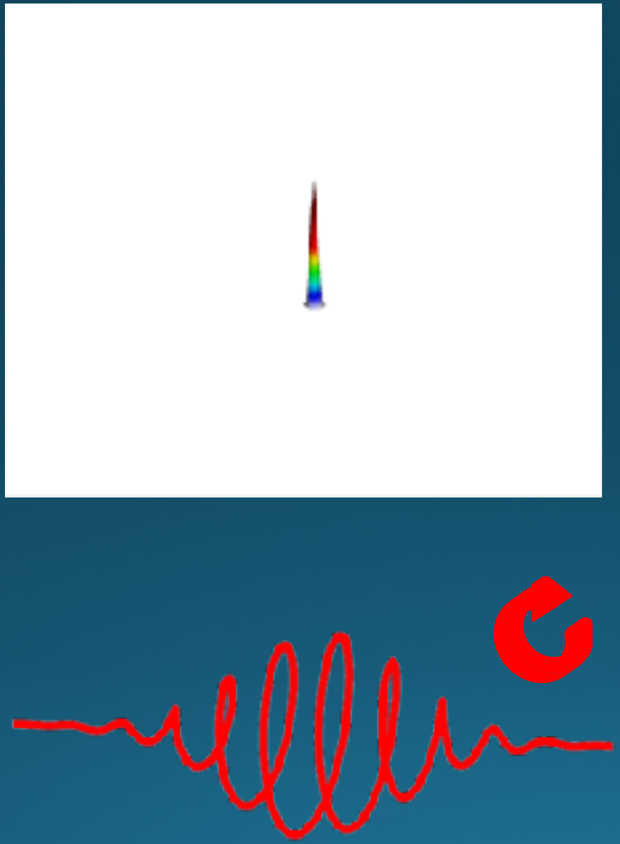

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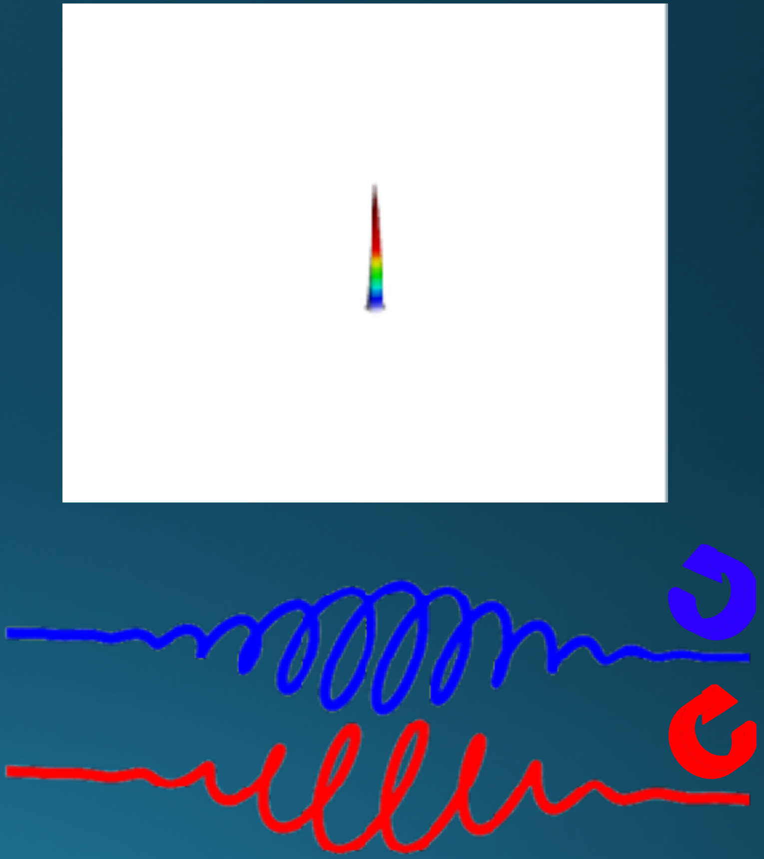

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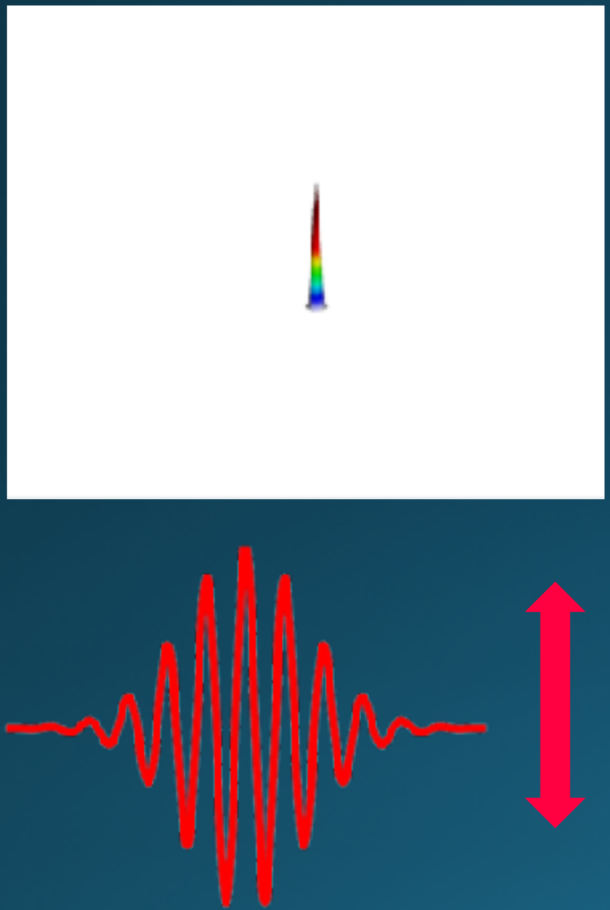

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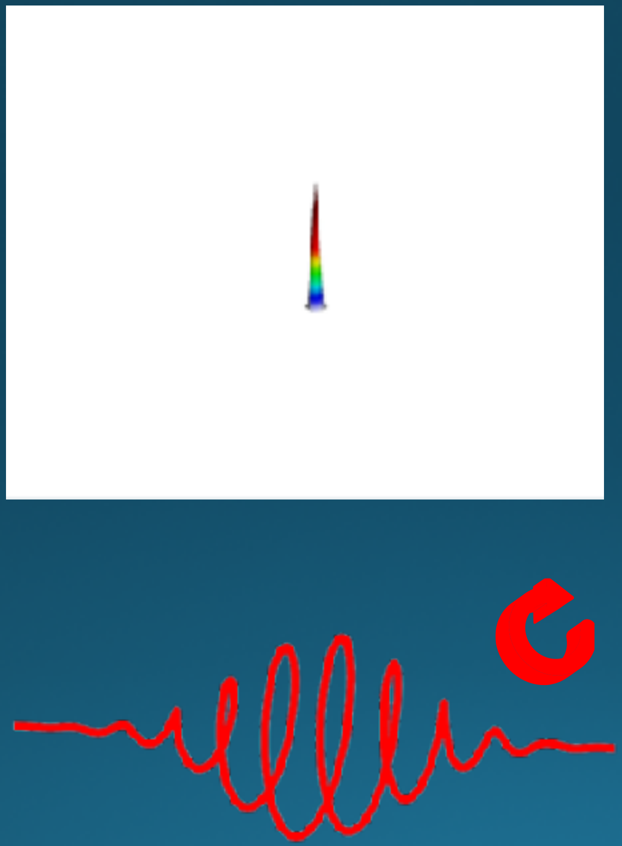

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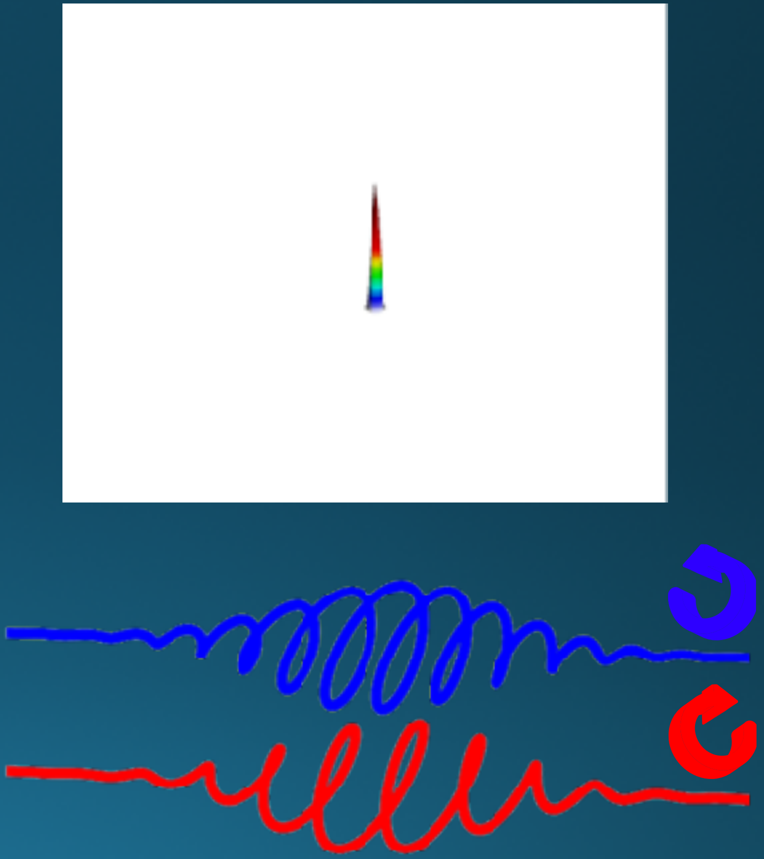
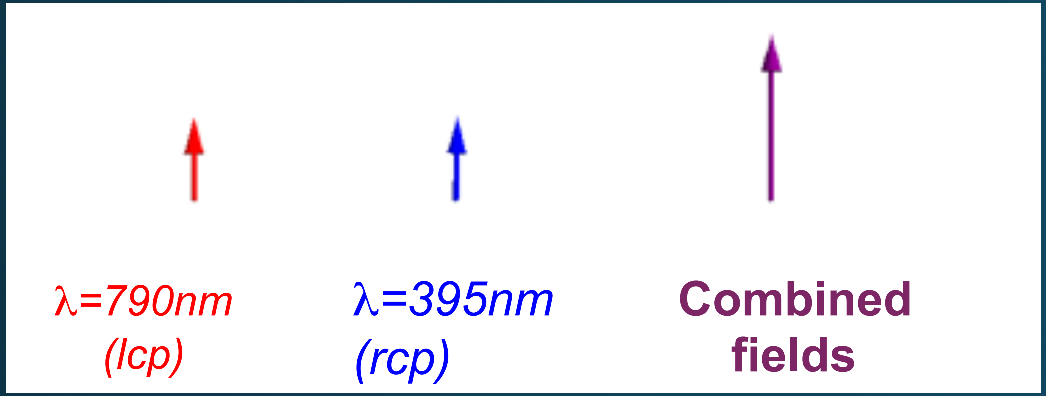


Circularly Polarized High Harmonic Generation (CPHHG): The Attosecond(s) From the Past!



Eichmann, Phys. Rev. A. 51, 1995
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Many, many, more

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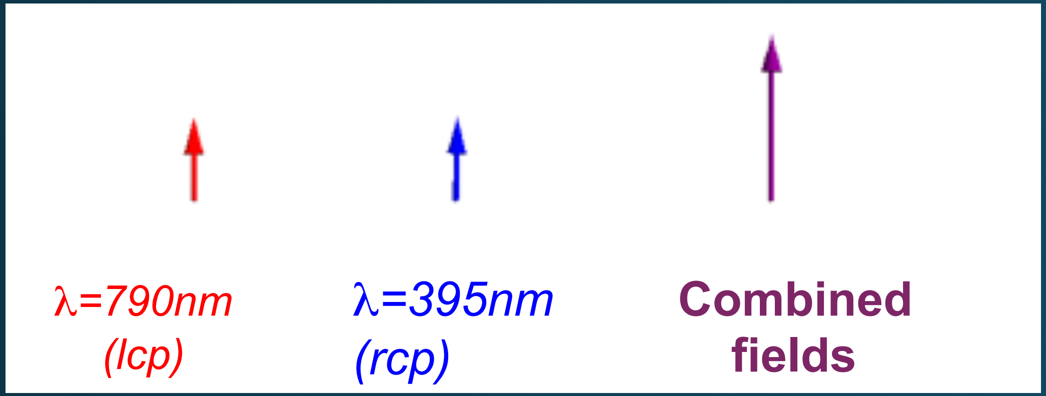

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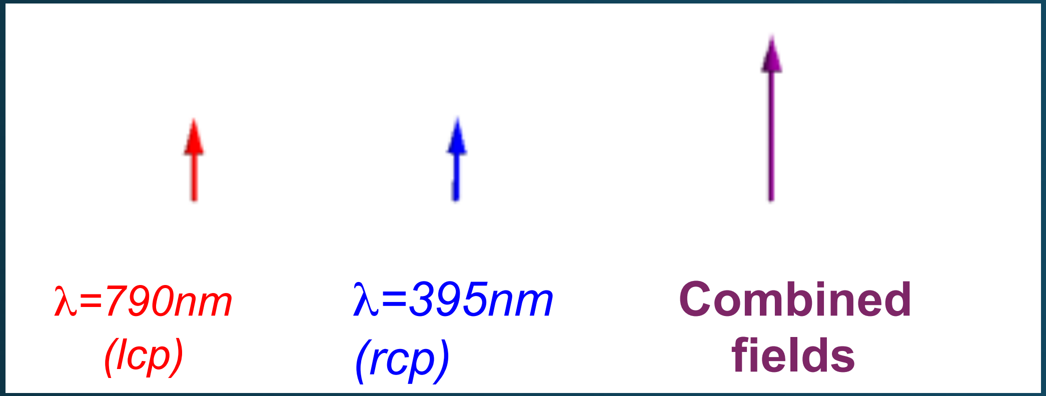

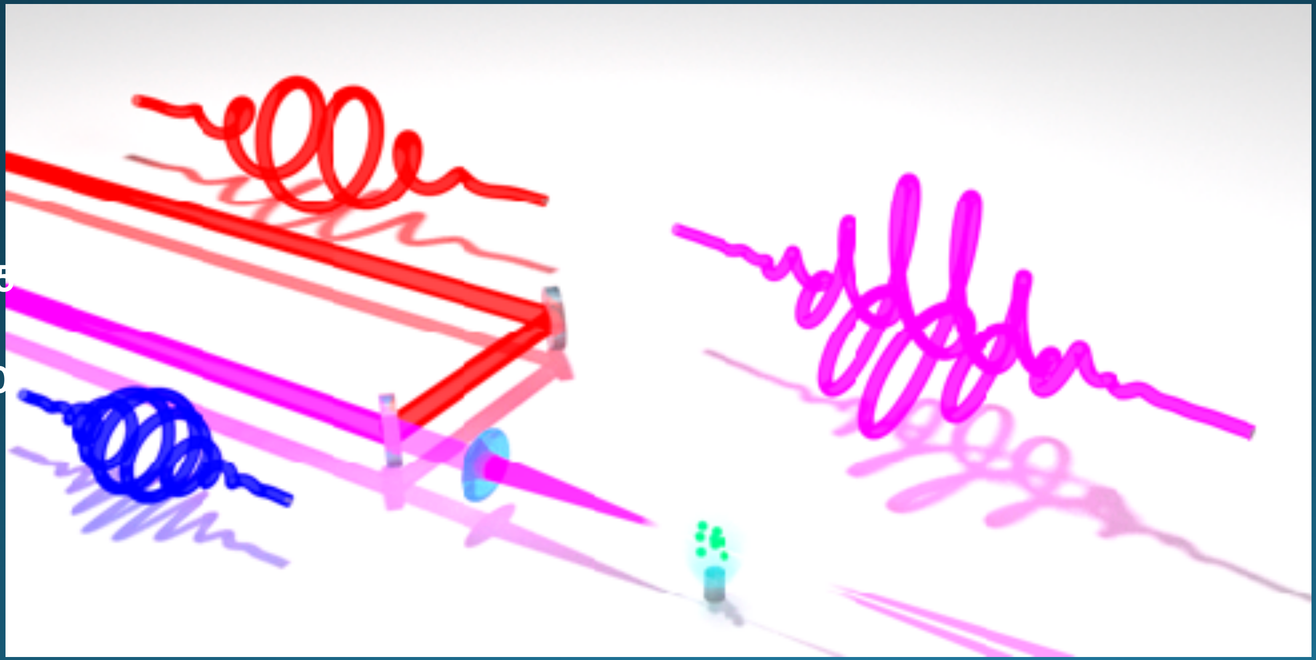

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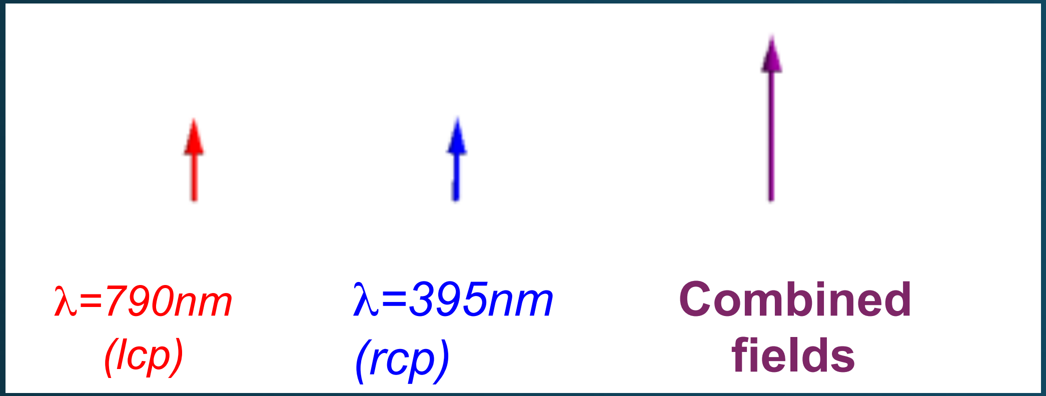

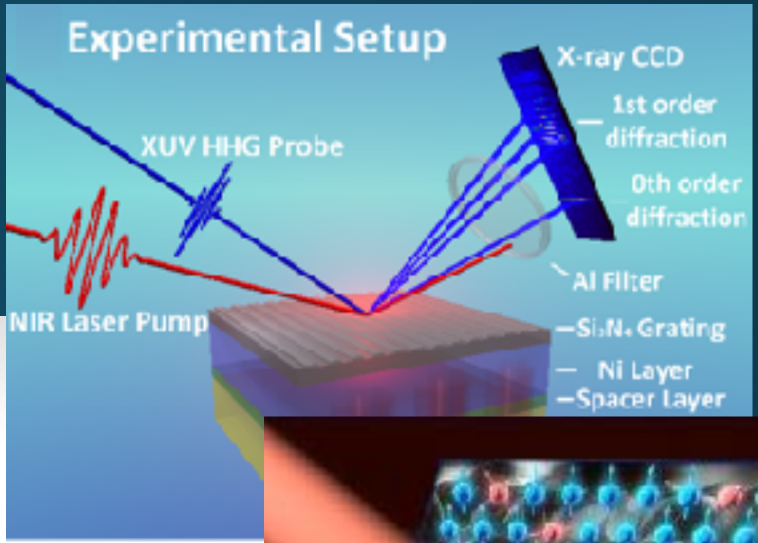

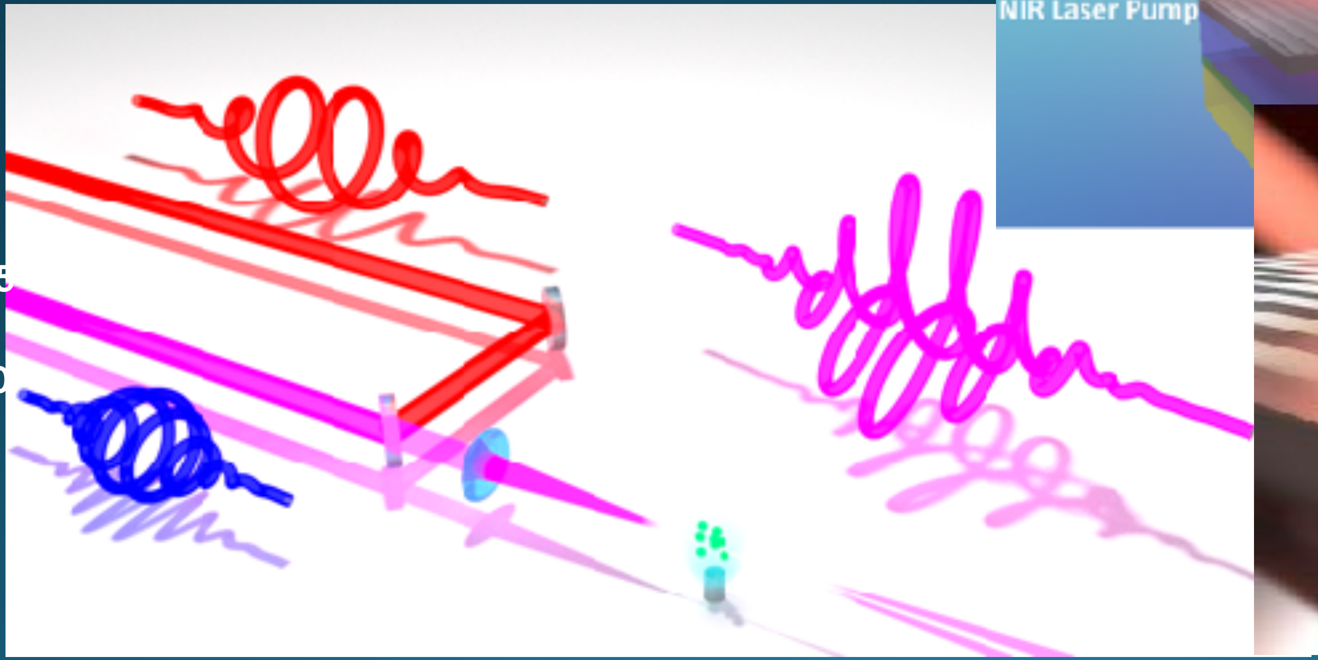

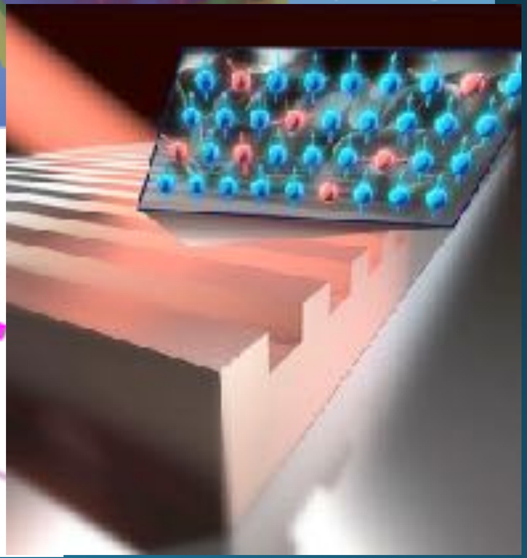

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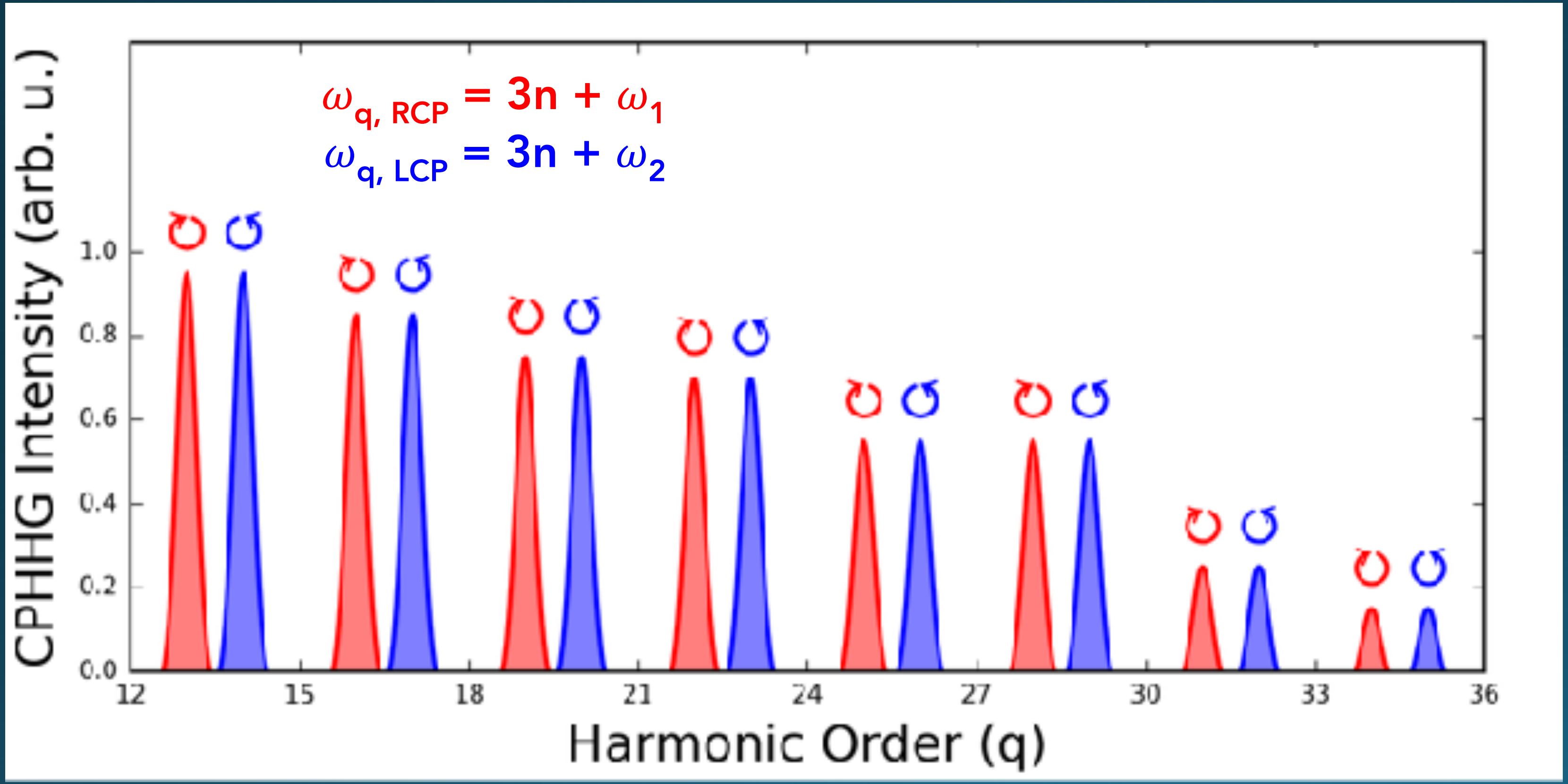
Circularly Polarized Harmonics, You Say? Well, Surely We Have Circular Attosecond Pulses!

- Spin angular momentum conservation = unique spectral and temporal features!



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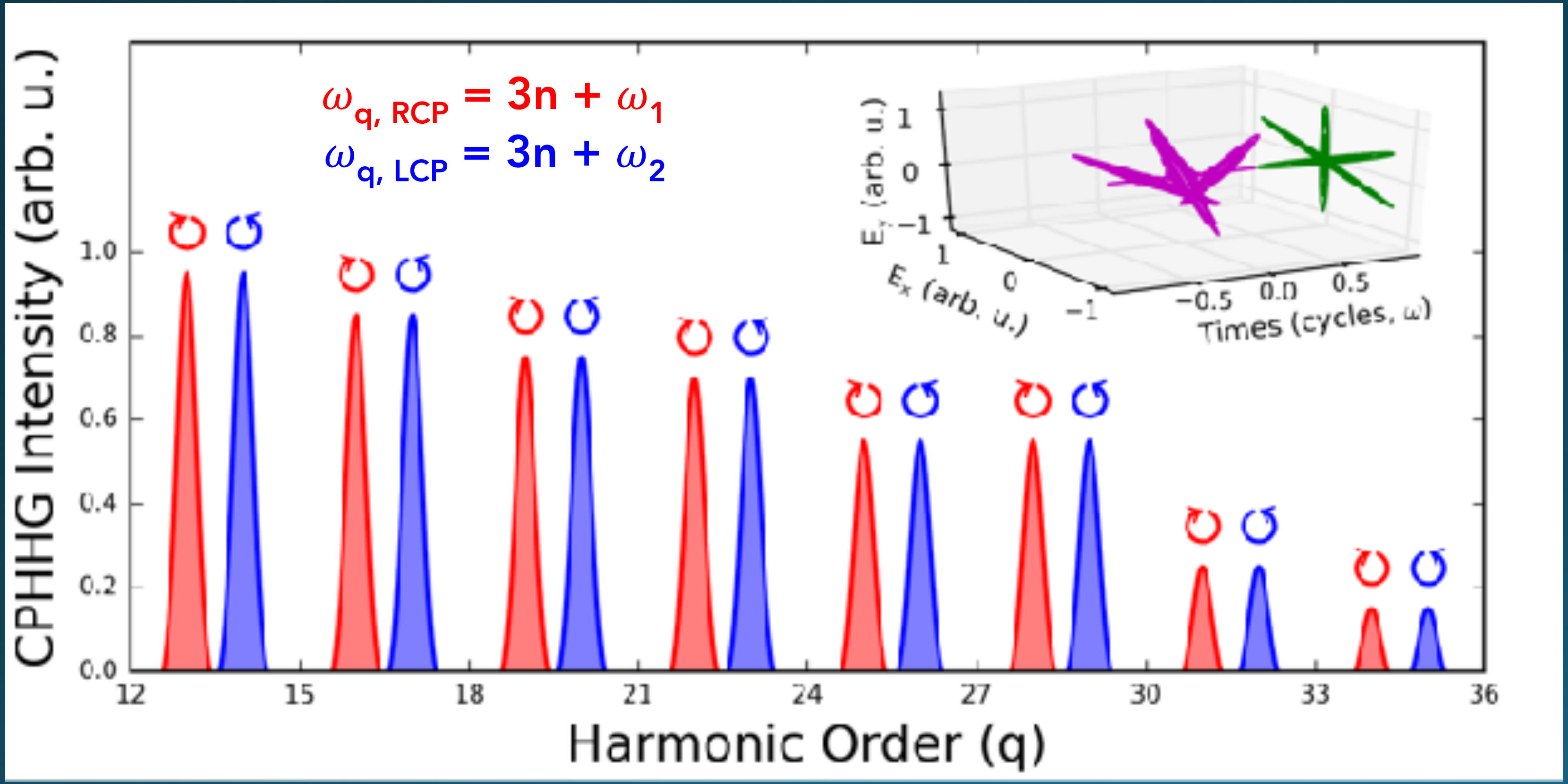

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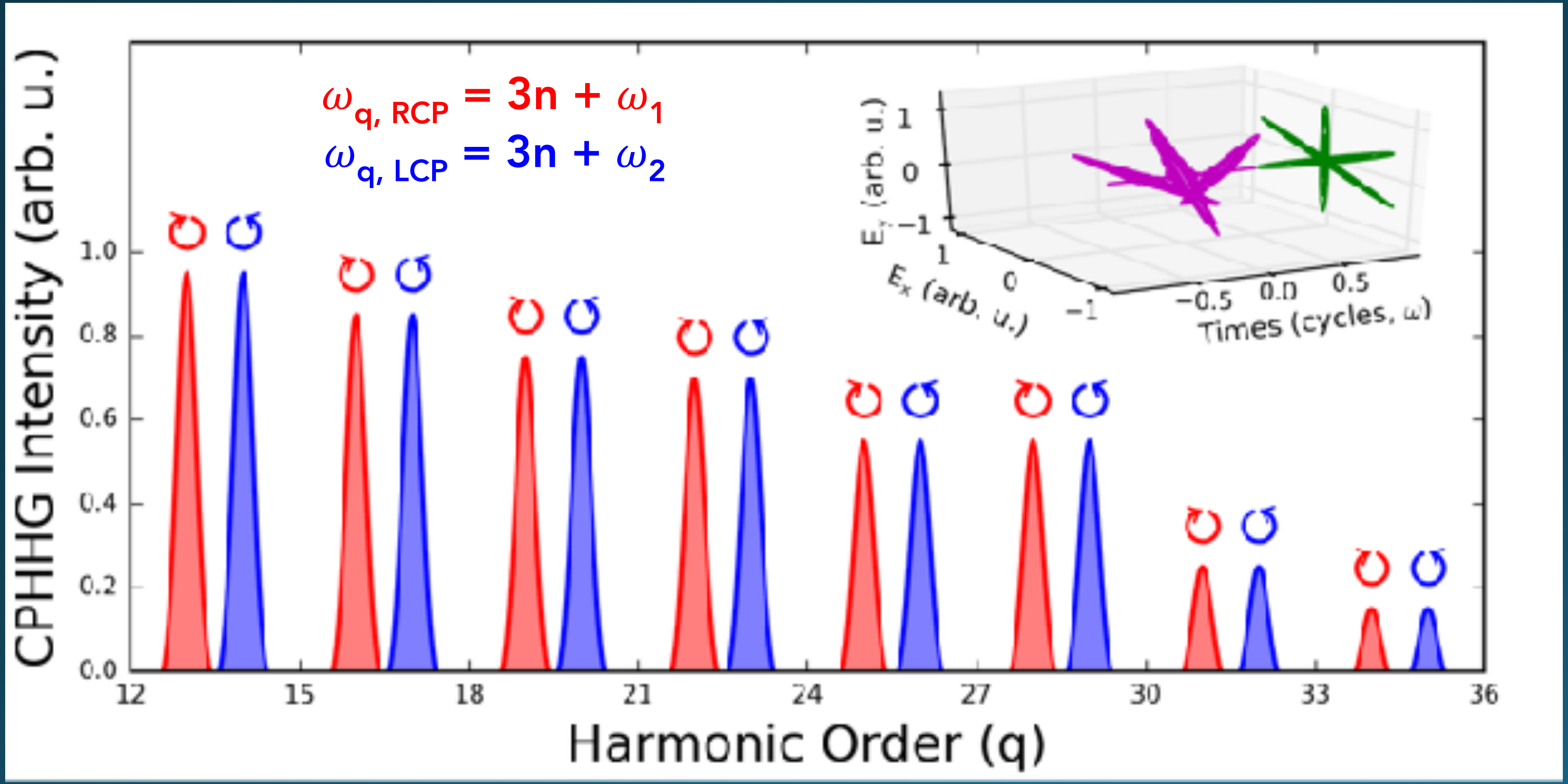

Linearly polarized APTs!
And don't call me Shirley

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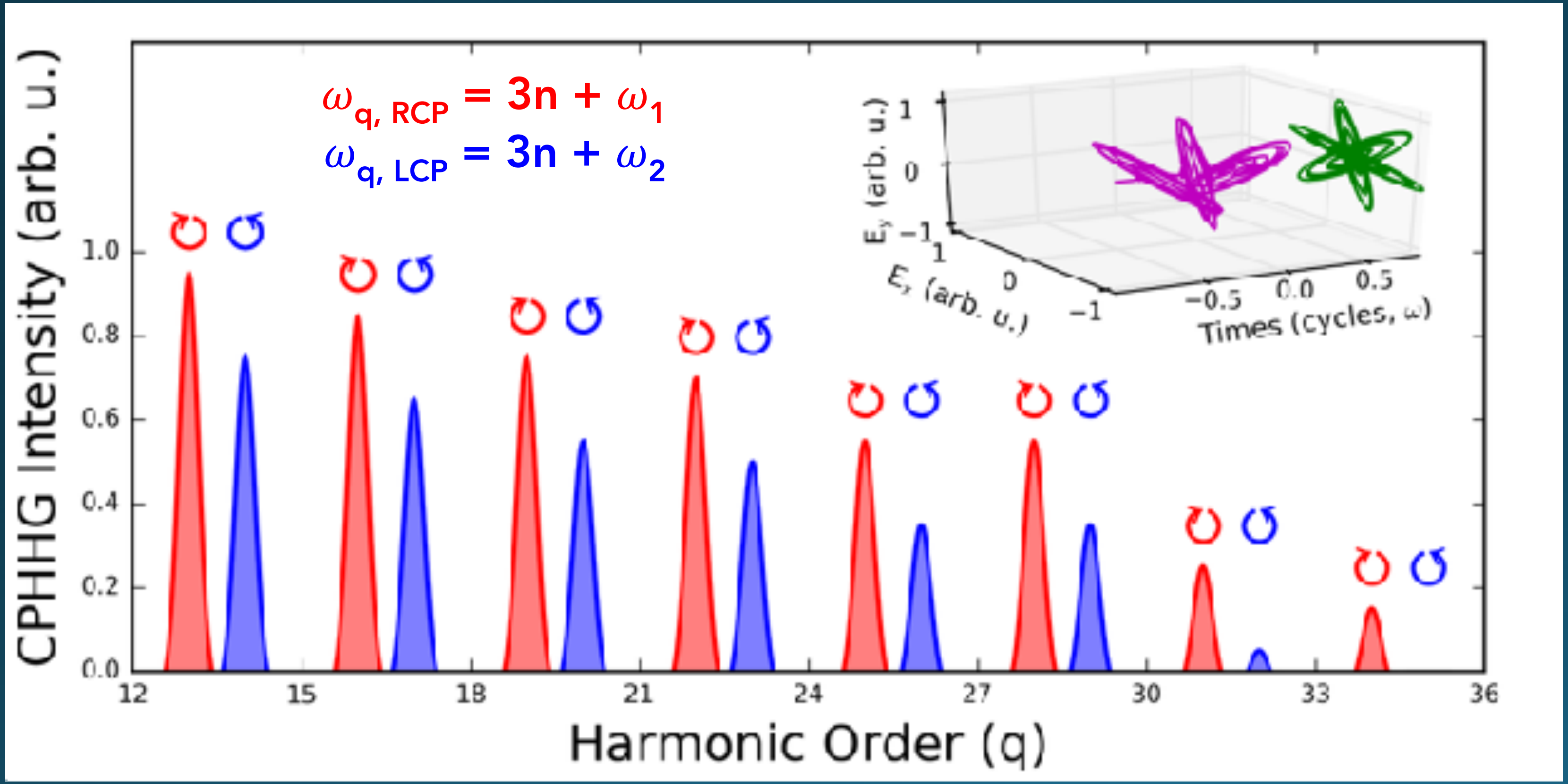

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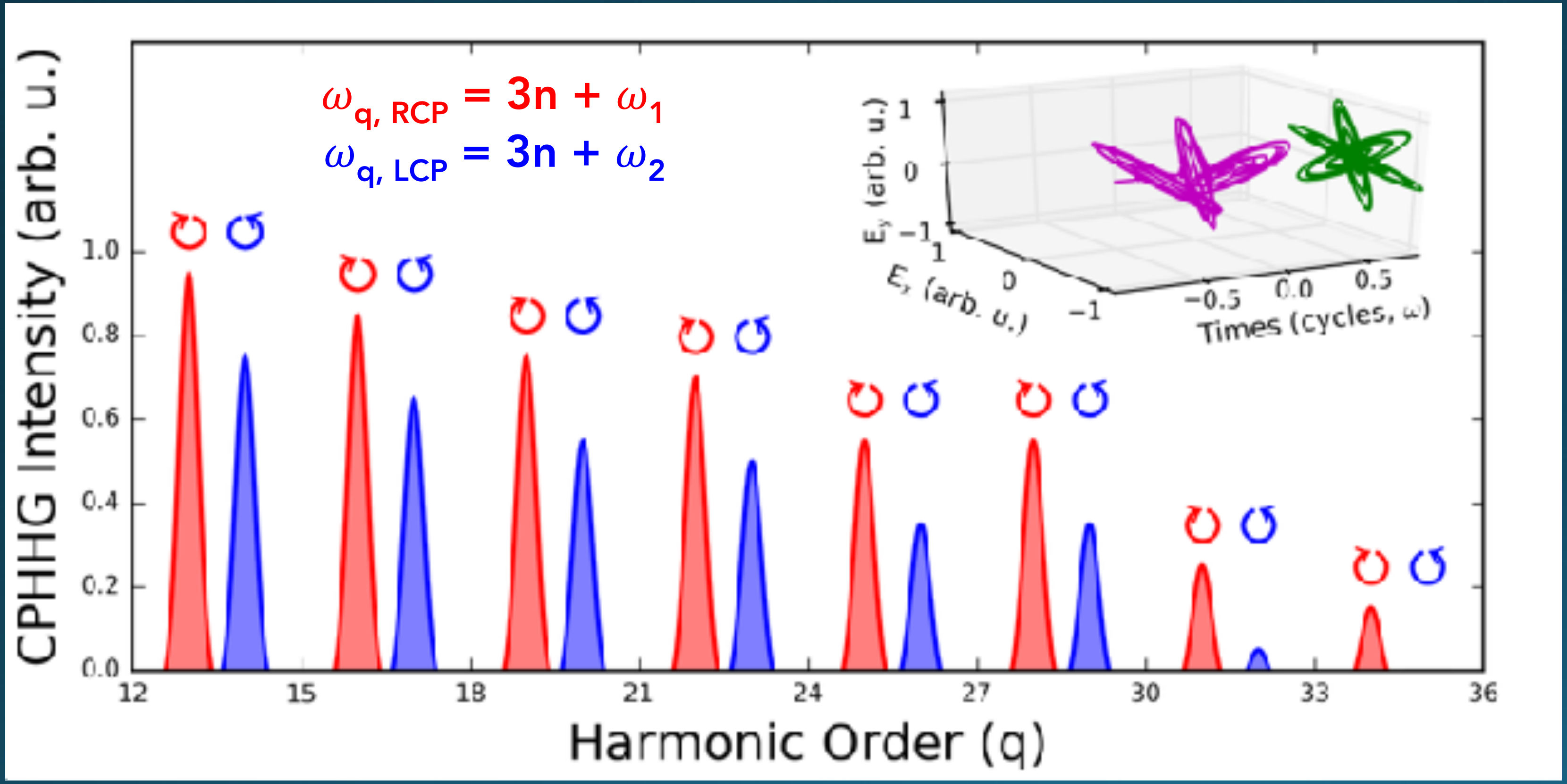
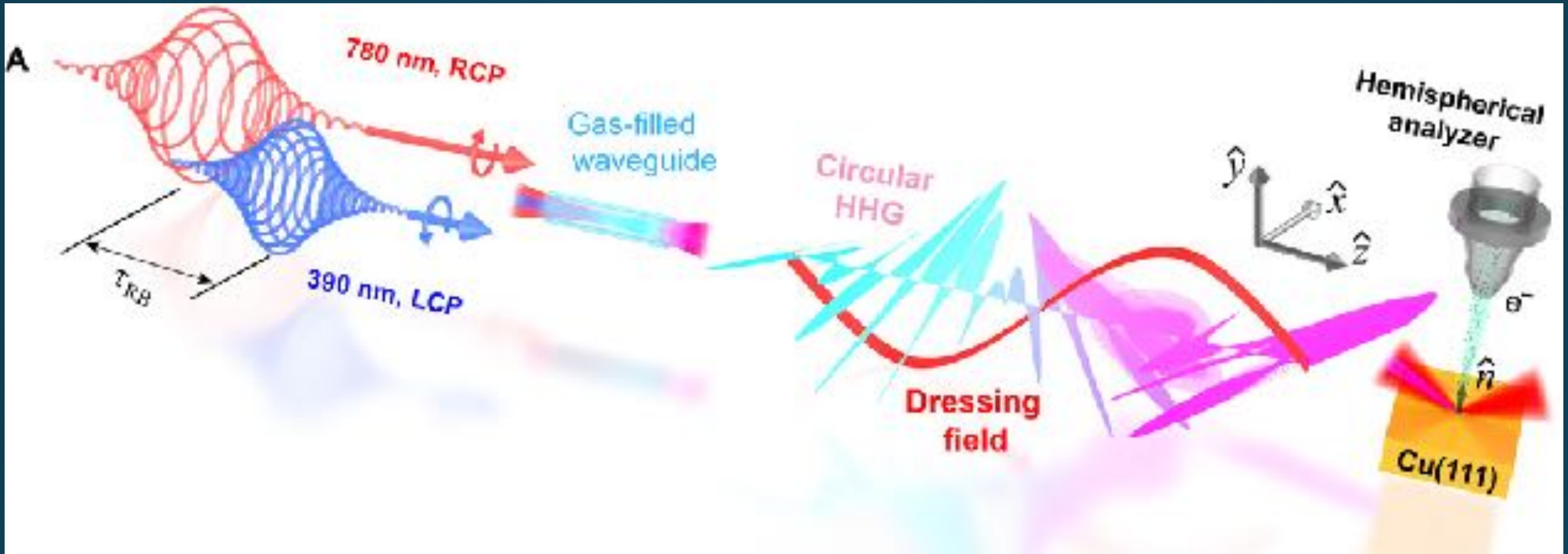


Well I'll be Shirley…

Atto-ARPES Metrology of CPHHG Waveforms: "Peeking" Inside the Attosecond Twists



Chen, Sci. Adv. 2, 2016

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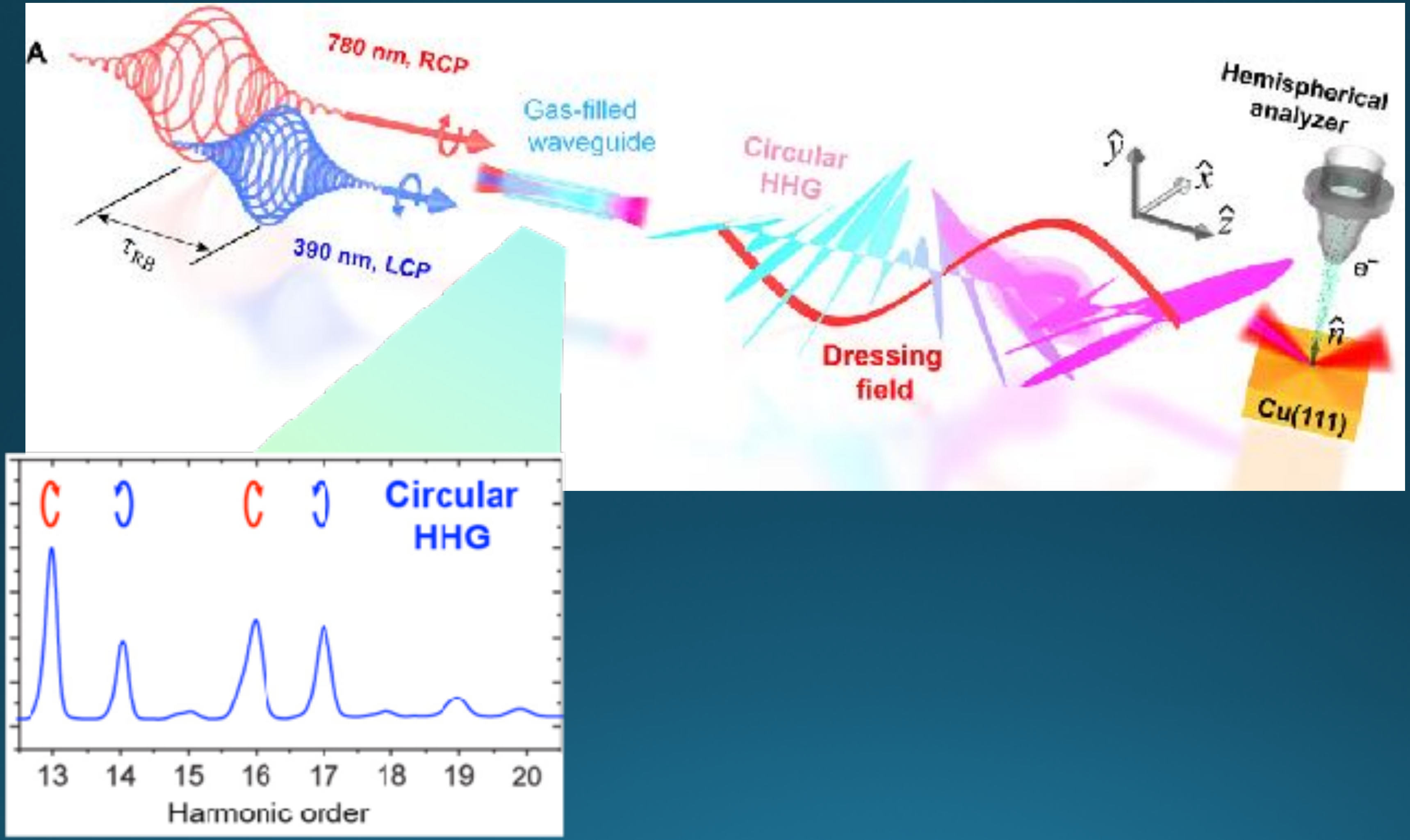

Chen, Sci. Adv. 2, 2016

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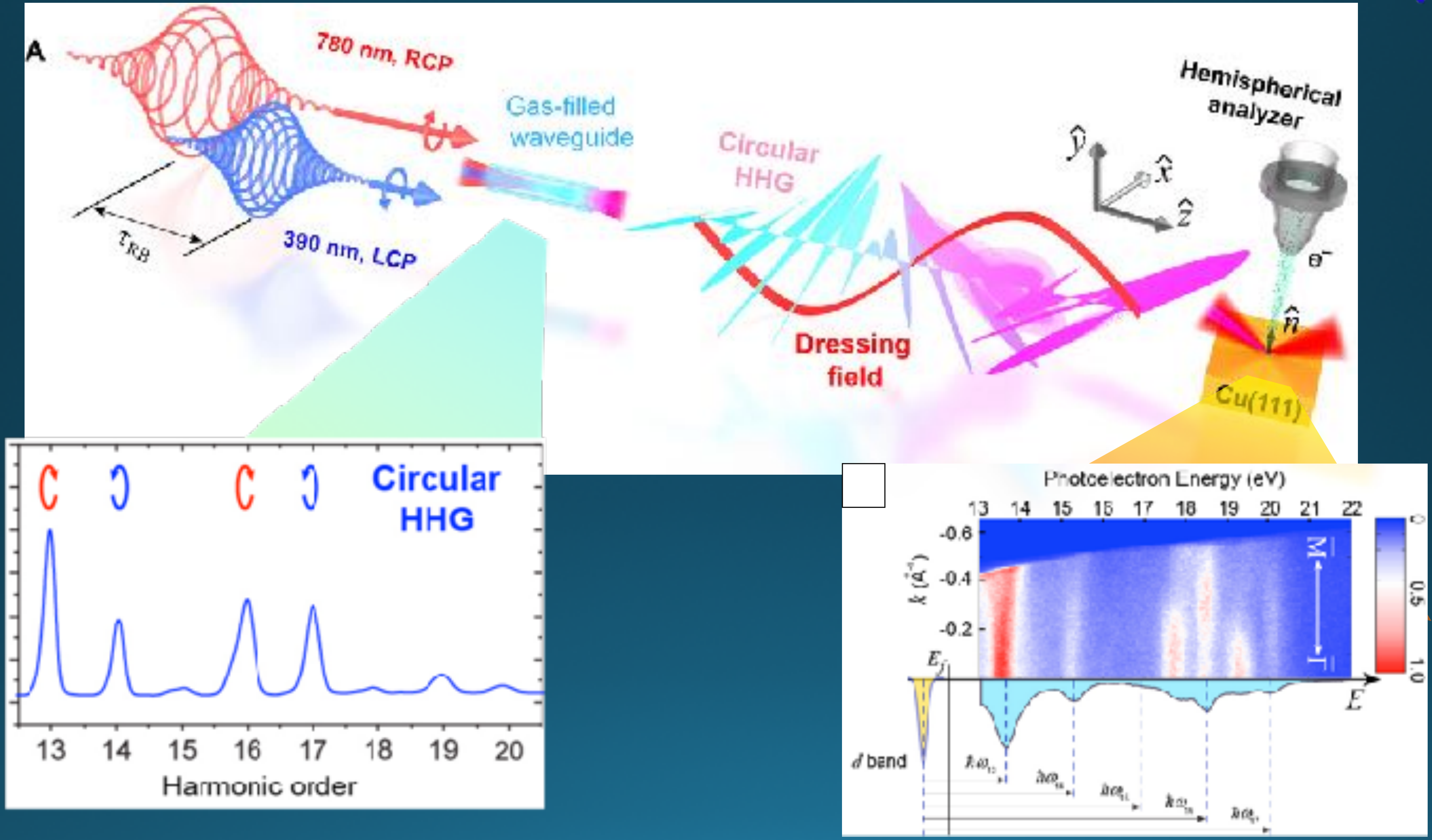

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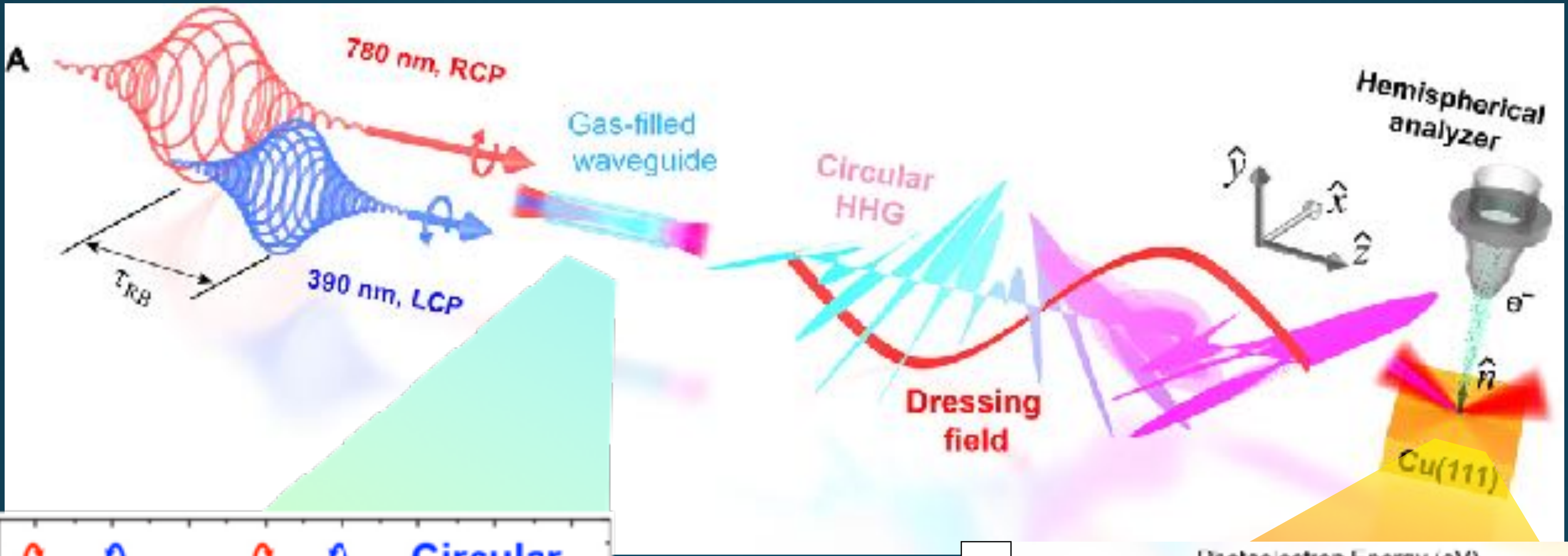
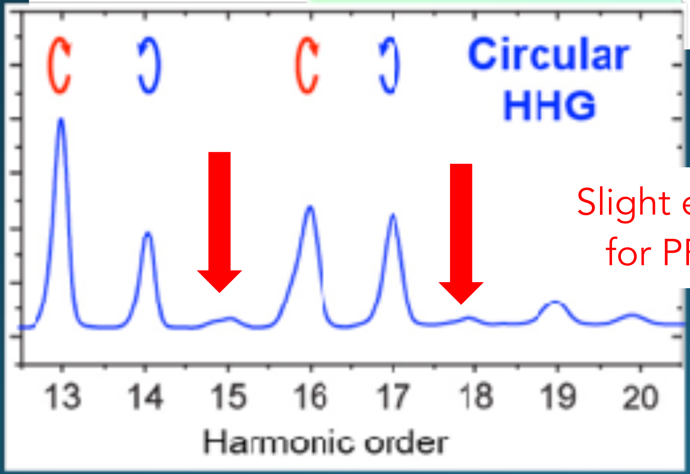
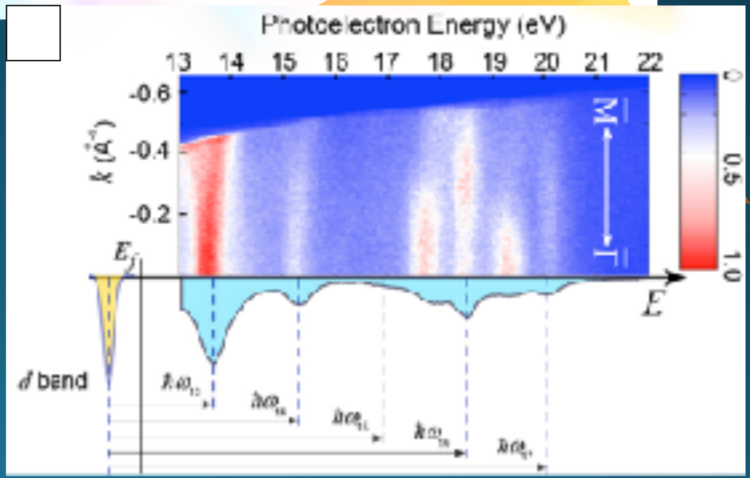




Slight ellipticty for PROOF



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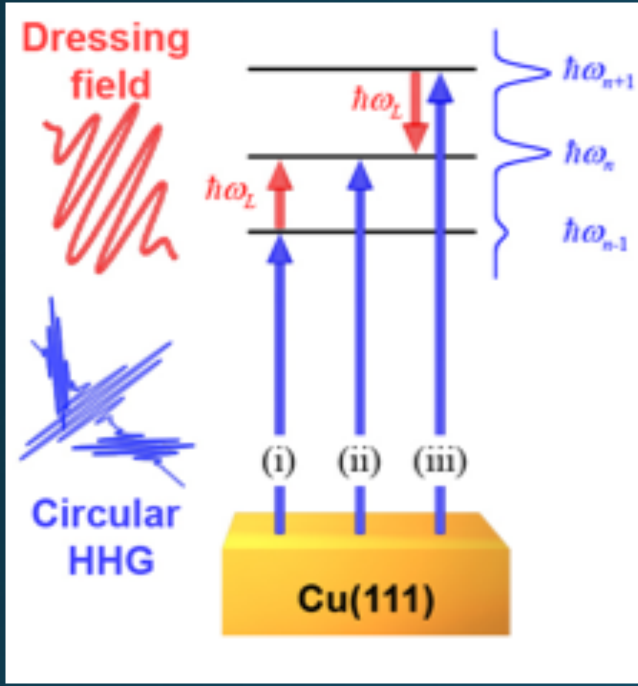
Atto-ARPES Metrology at JILA:
Complete Reconstruction of the Most Complex Light Field to Date!



Video: Courtesy Dan Hickstein

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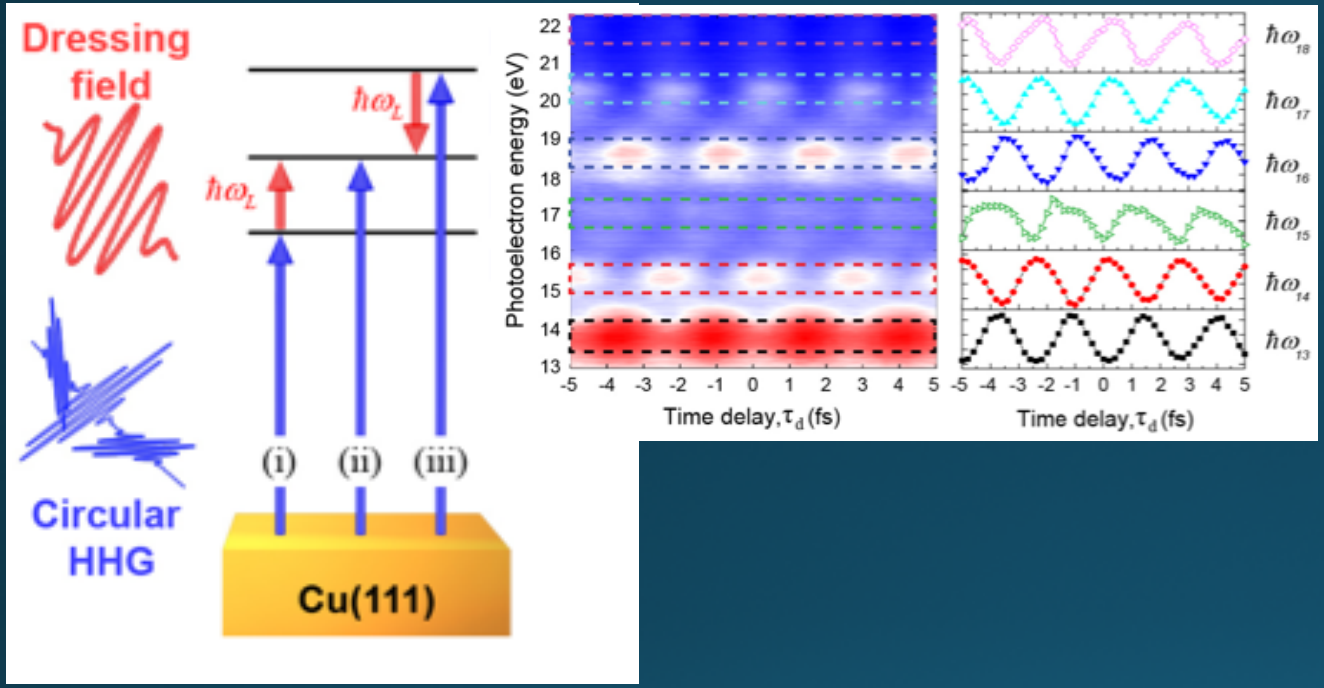

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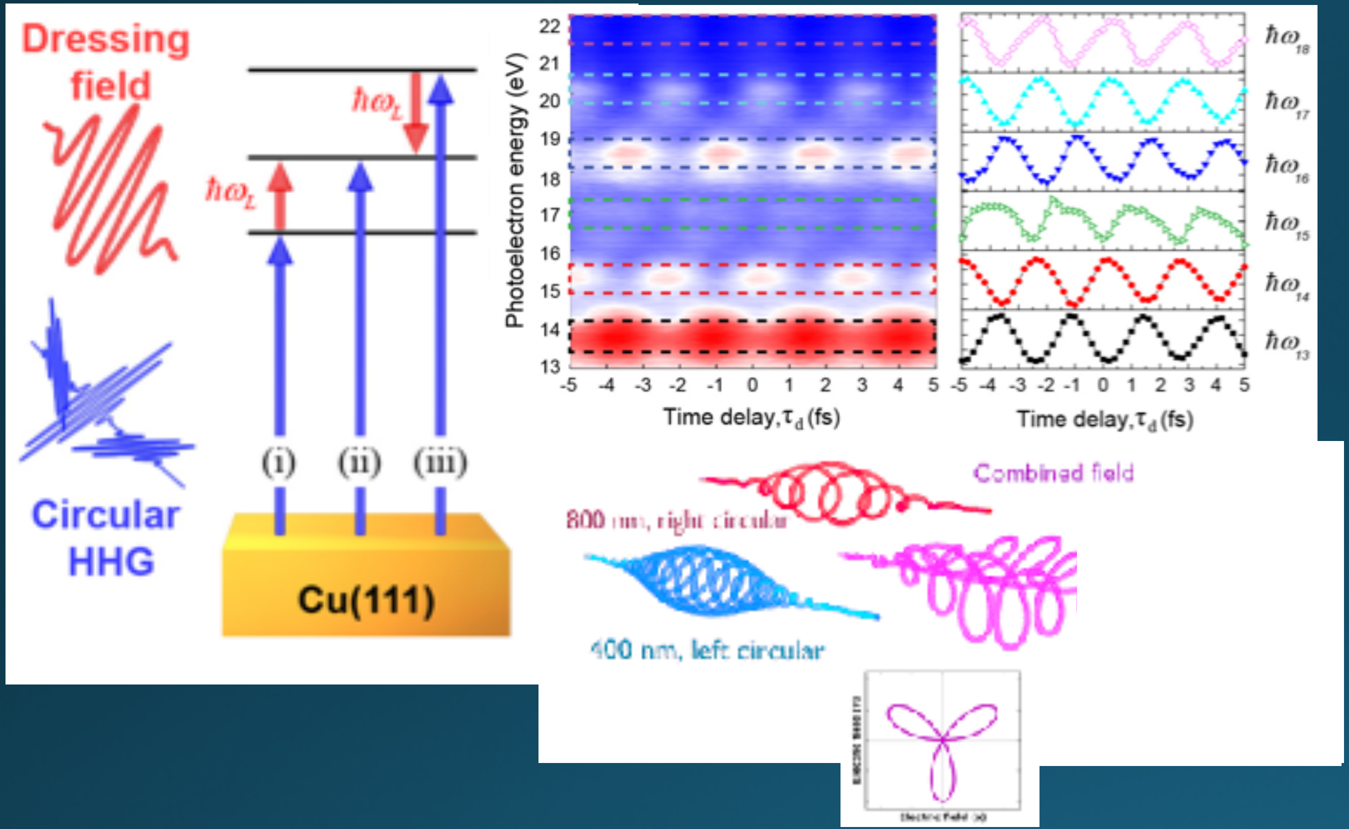

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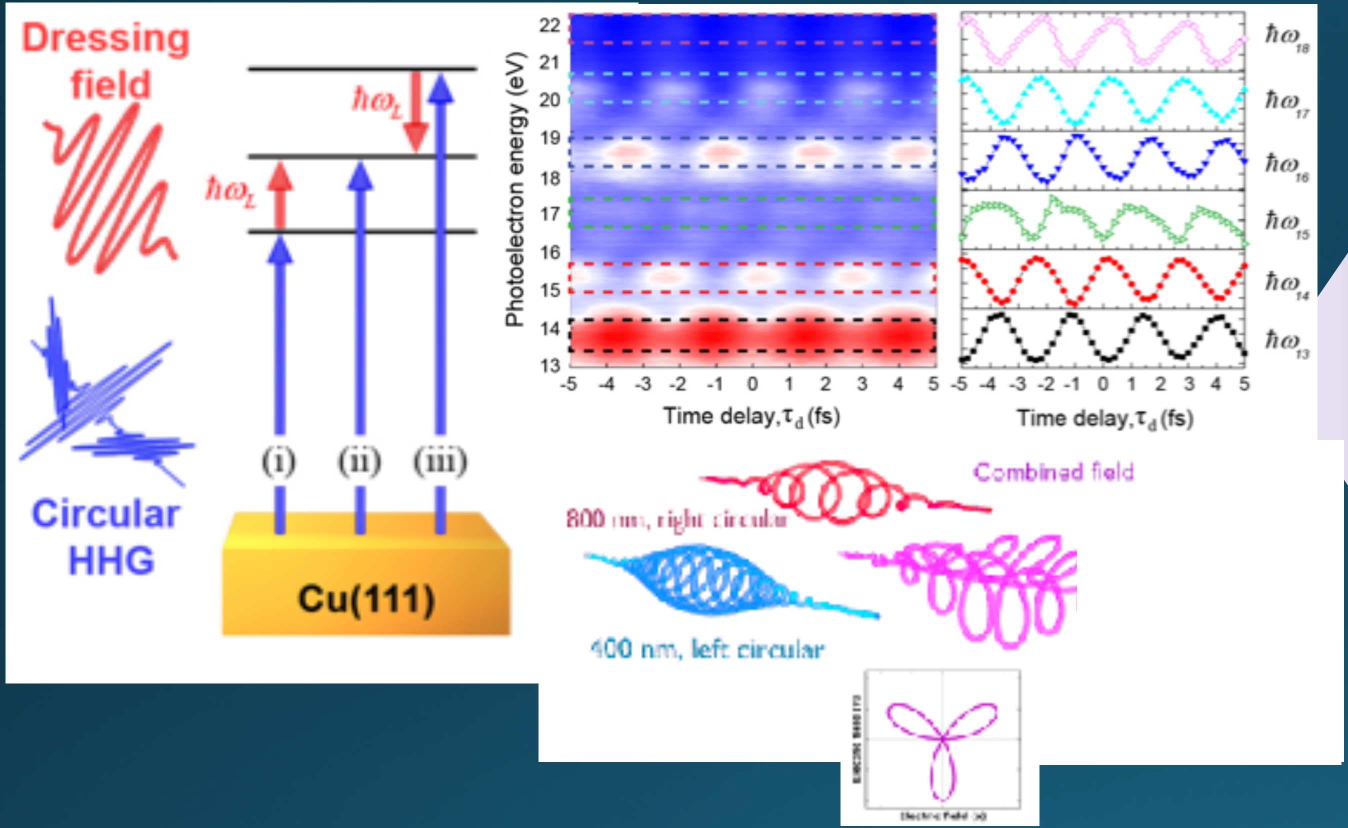

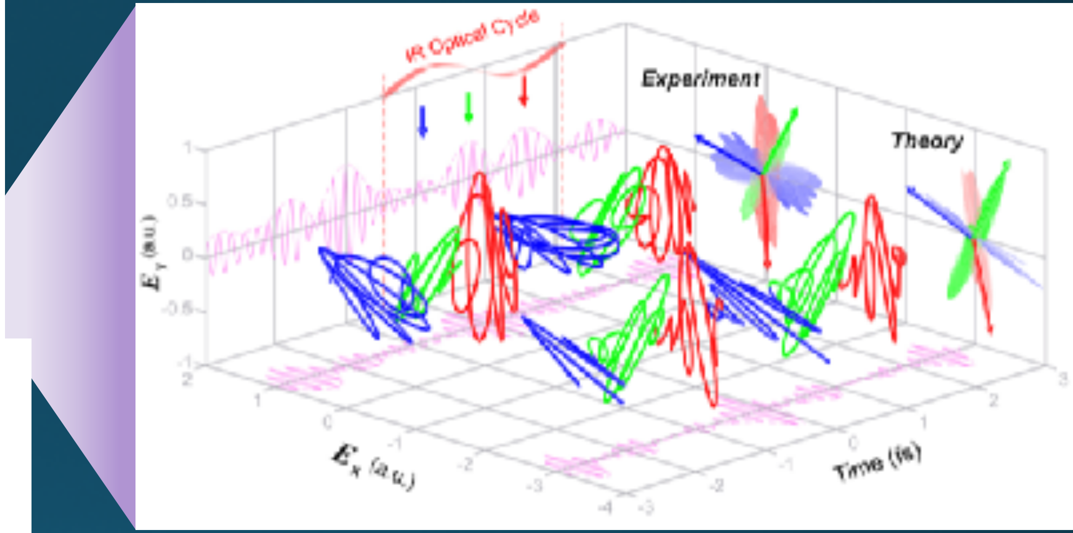

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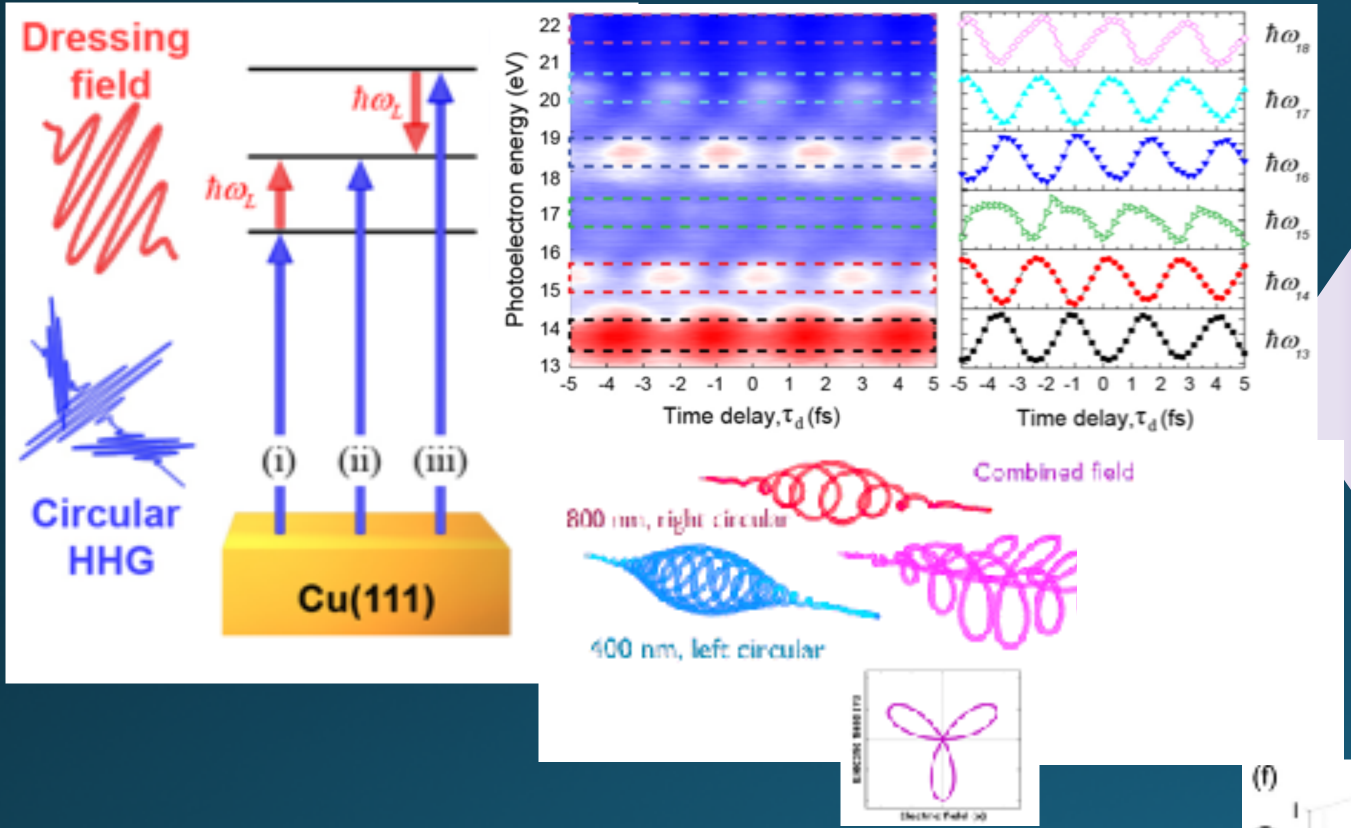

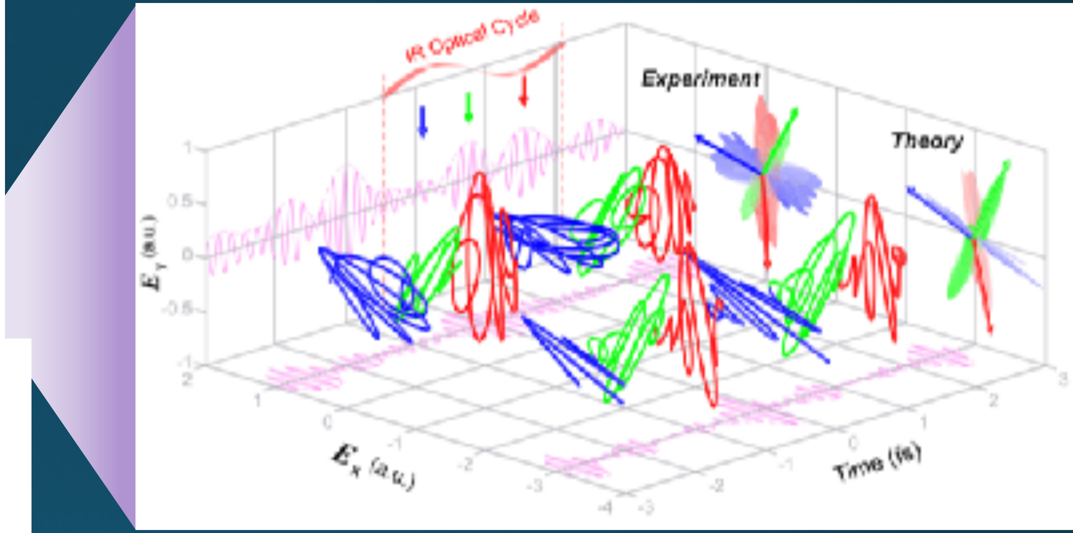

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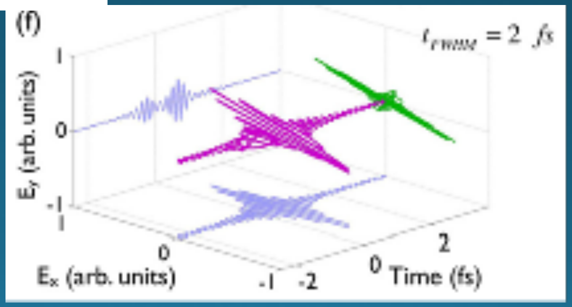
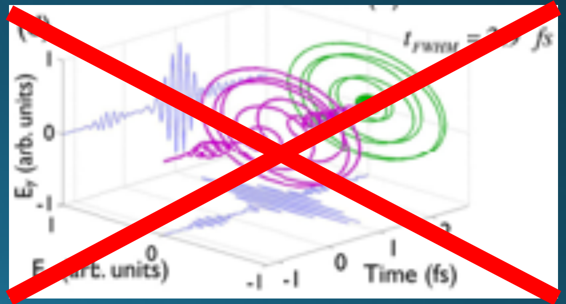
Isolated Attosecond Pulses?





Circularly Polarized High Harmonic Generation (CPHHG): An Ideal Probe of Chiral Systems

Baykusheva, PRL, 116, 2016
Fan, PNAS, 112, 2015

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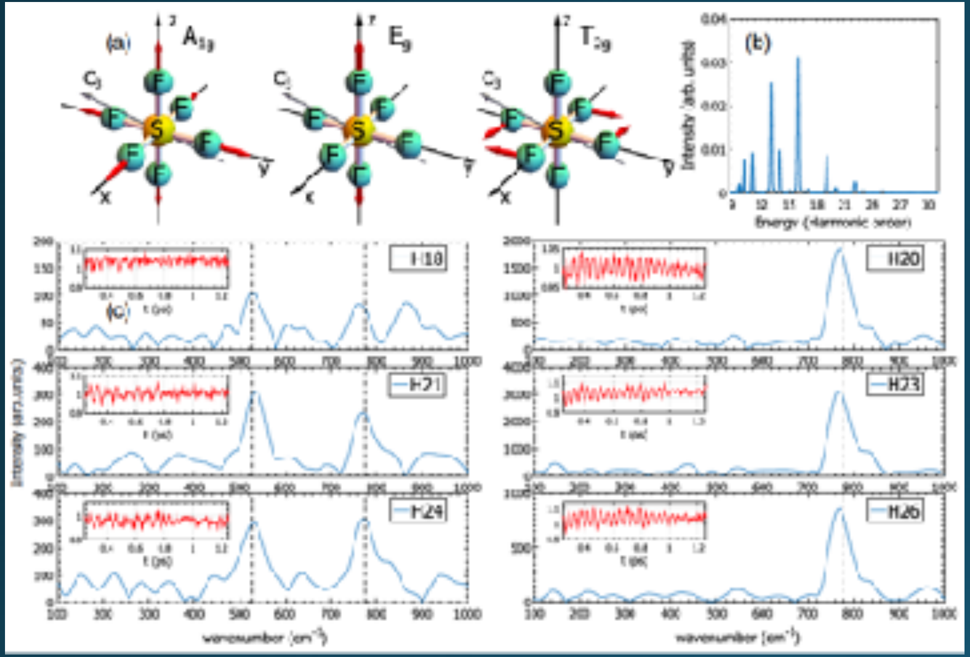
Dynamic Molecular Motions and Symmetries



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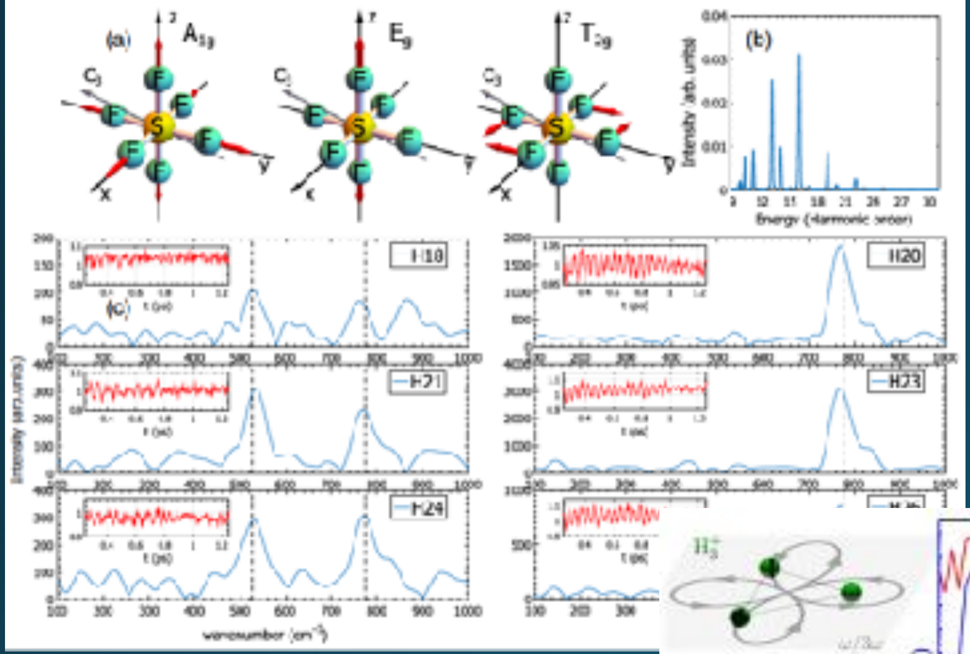

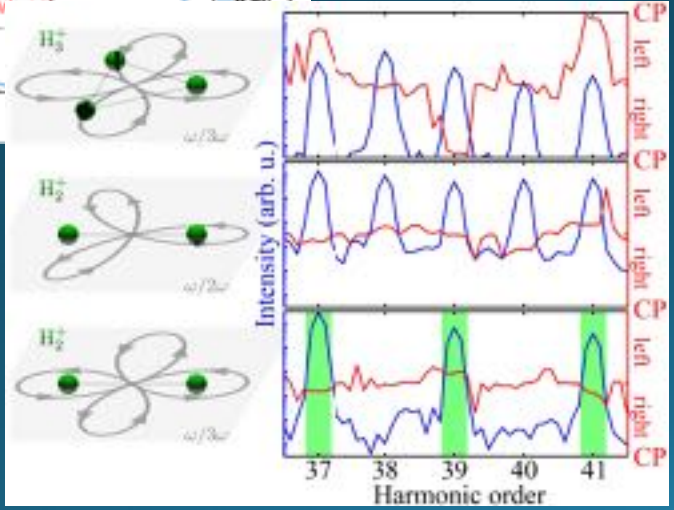

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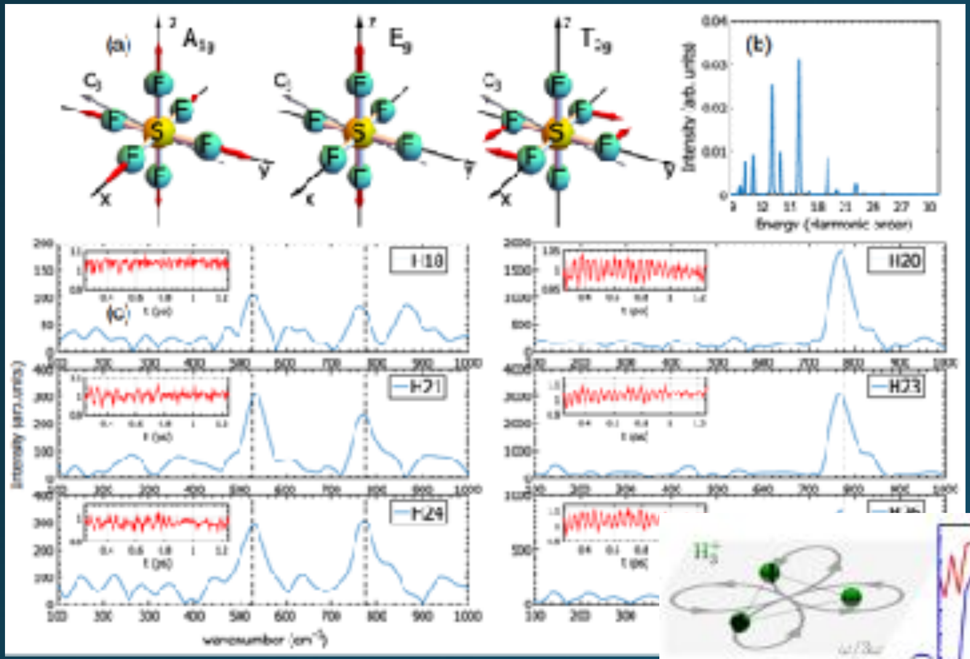
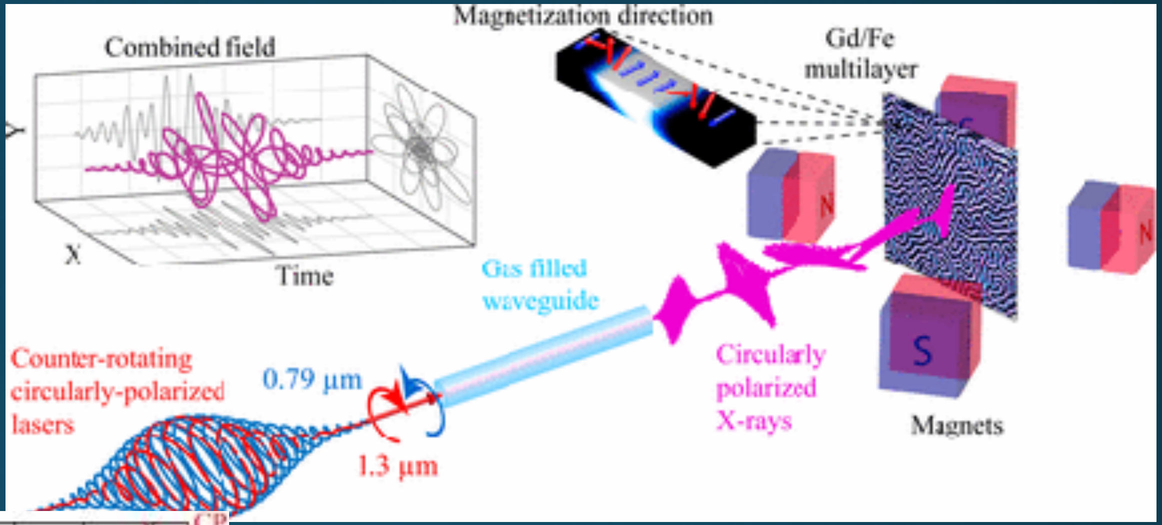
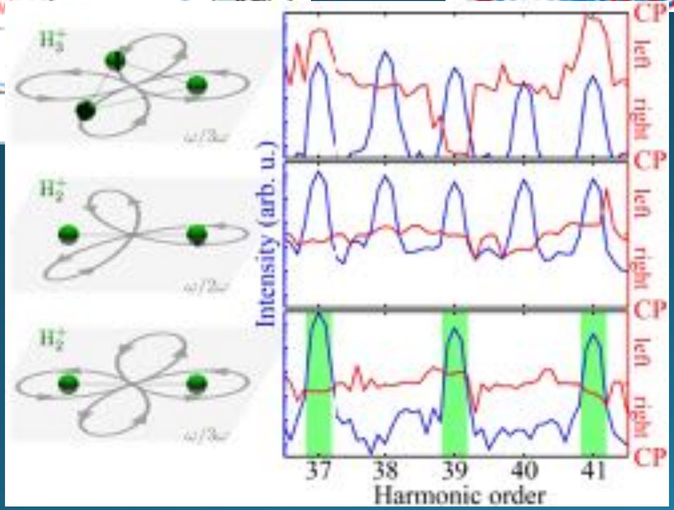


M edge Magneto-Optical Spectroscopy





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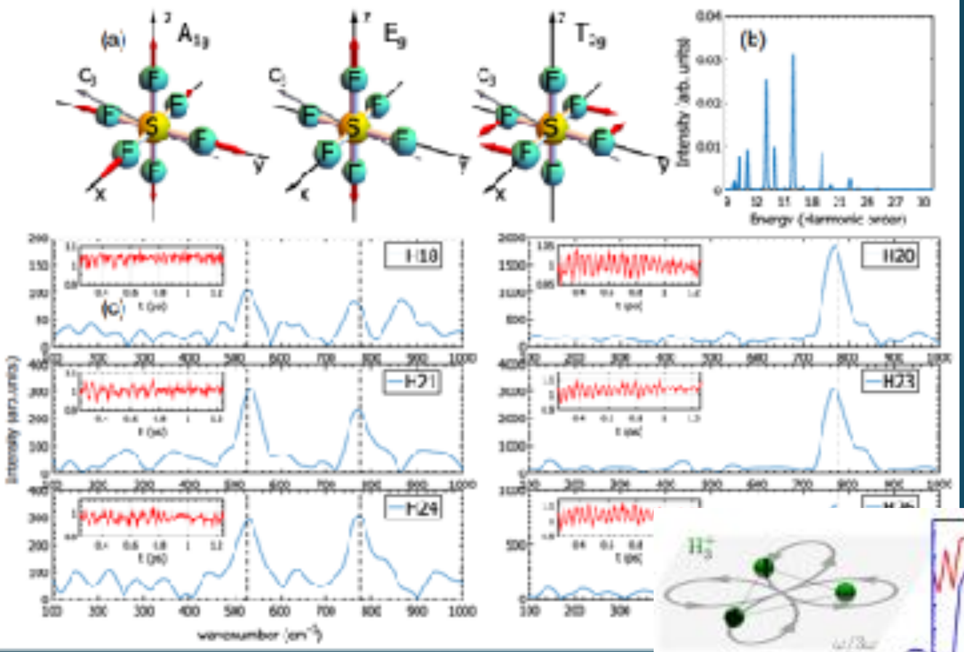

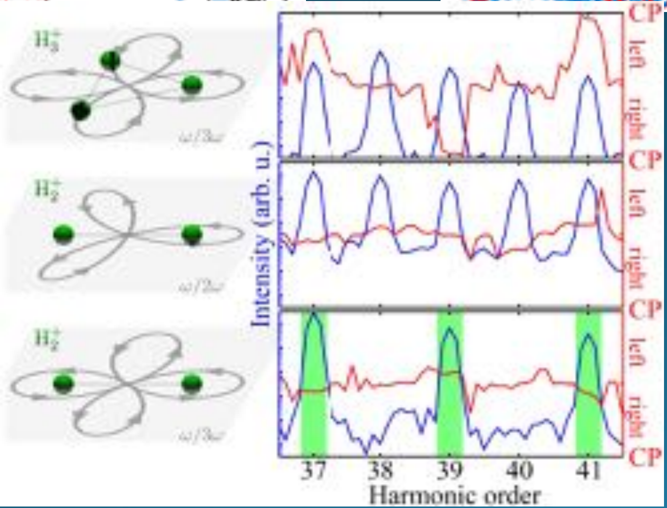

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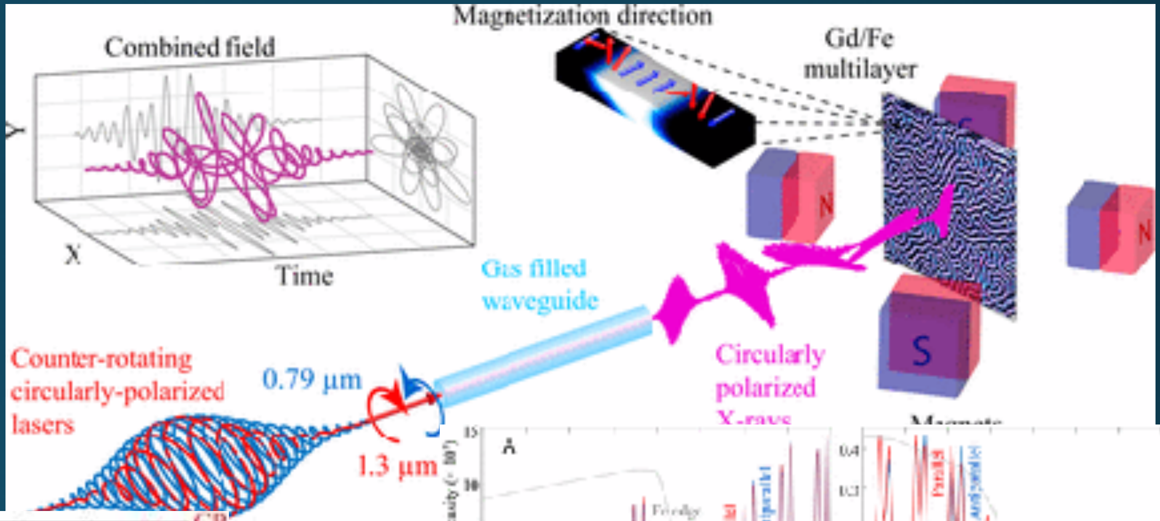

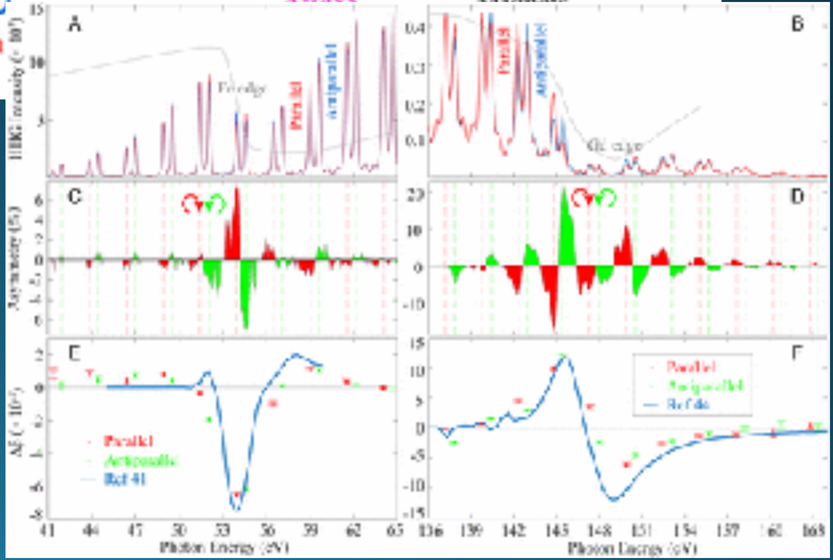

Controlling the Temporal Ellipticity of CPHHG: Towards Circularly Polarized Attosecond Pulse(s)

Medisauskas, PRL, 115, 2015
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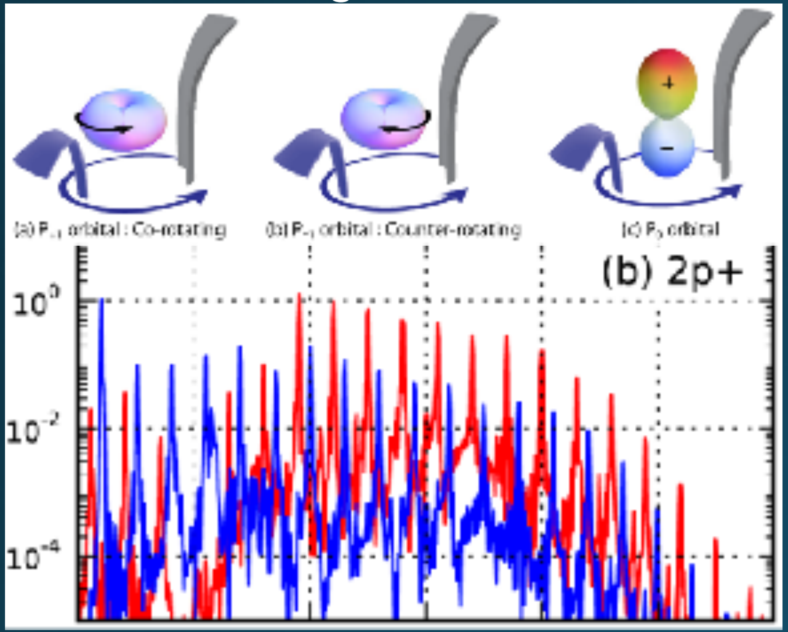
Single Atom



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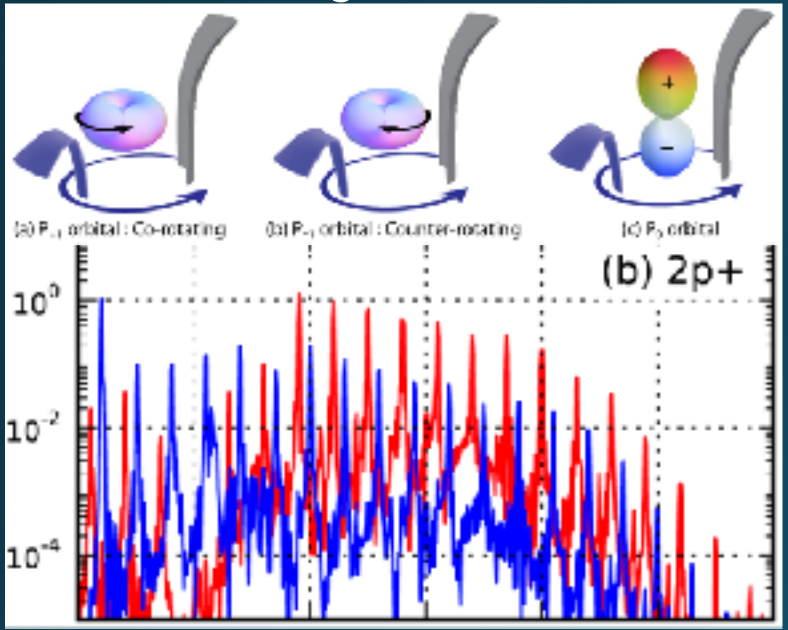

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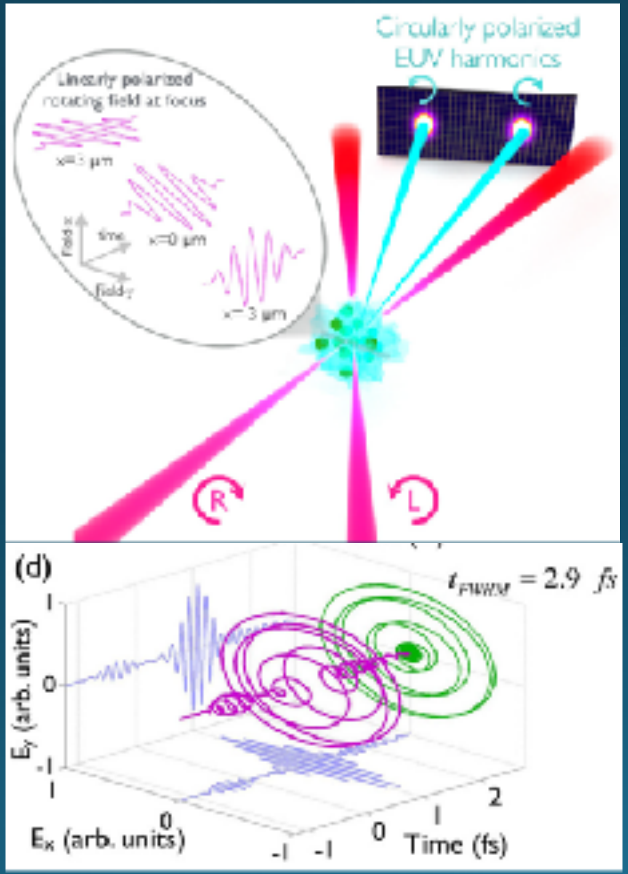
Wavefront Control



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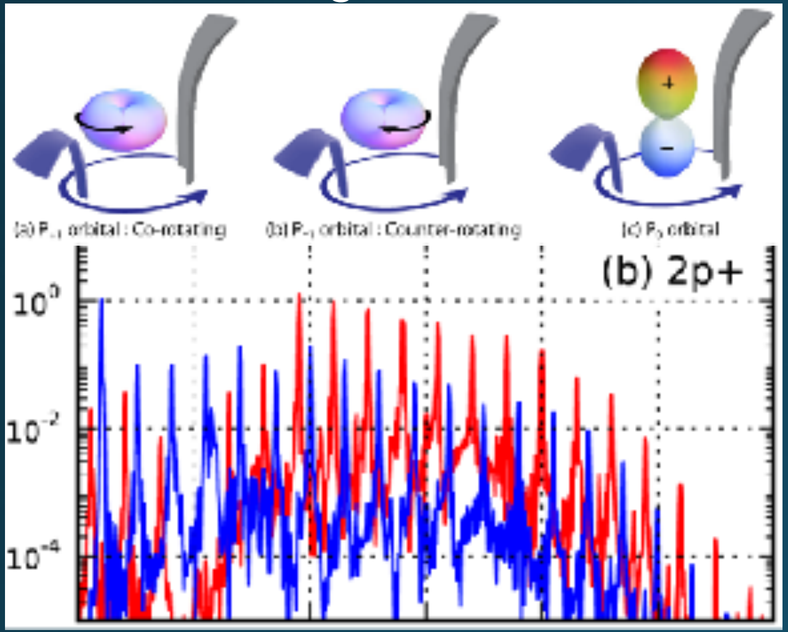

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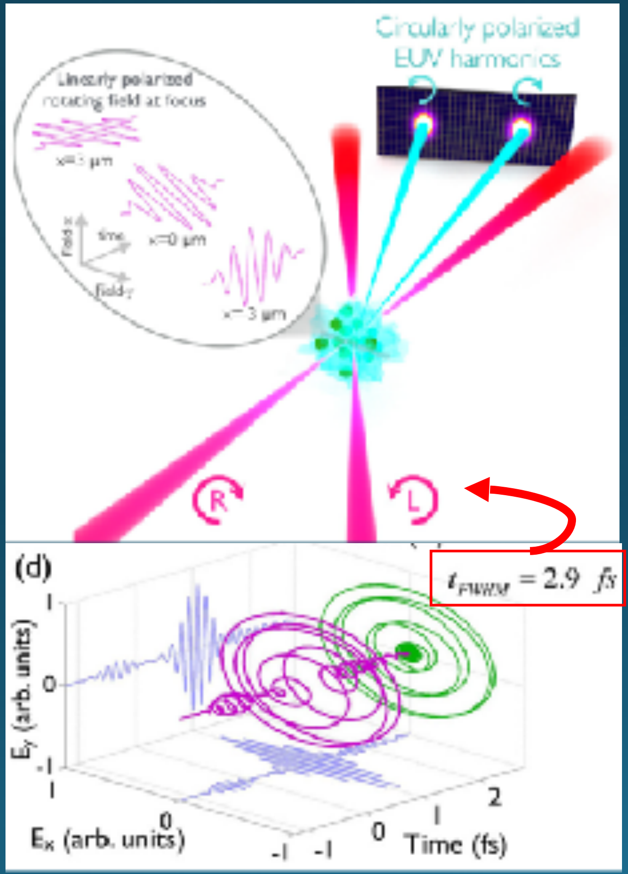

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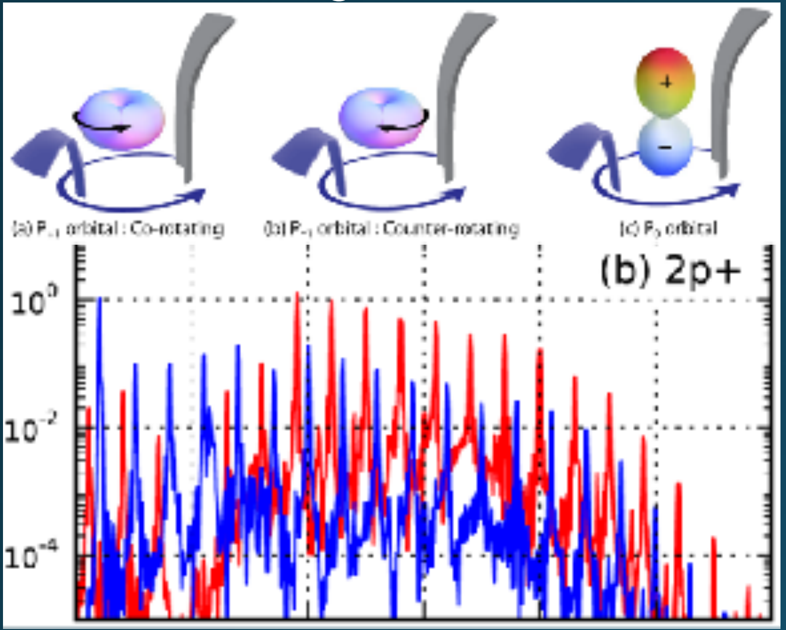

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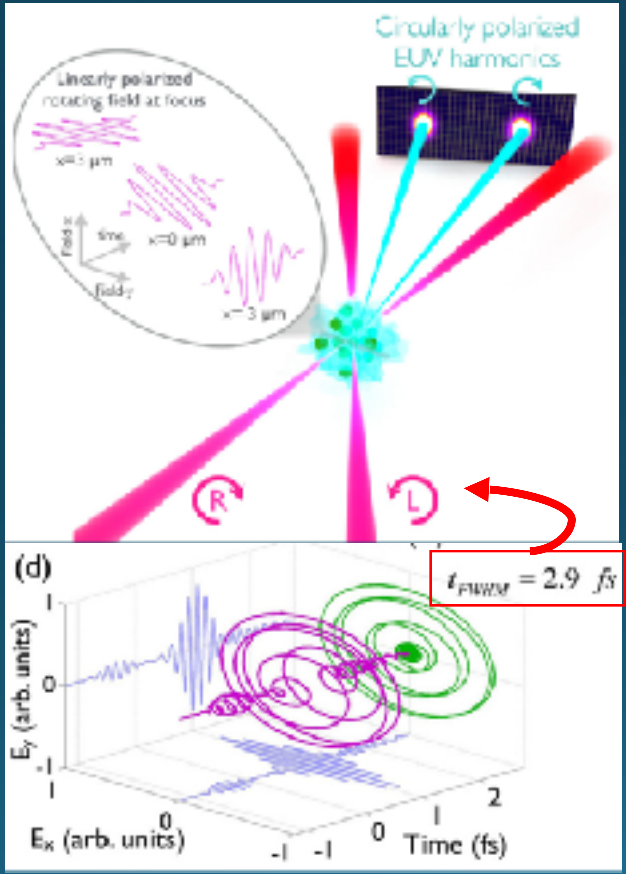
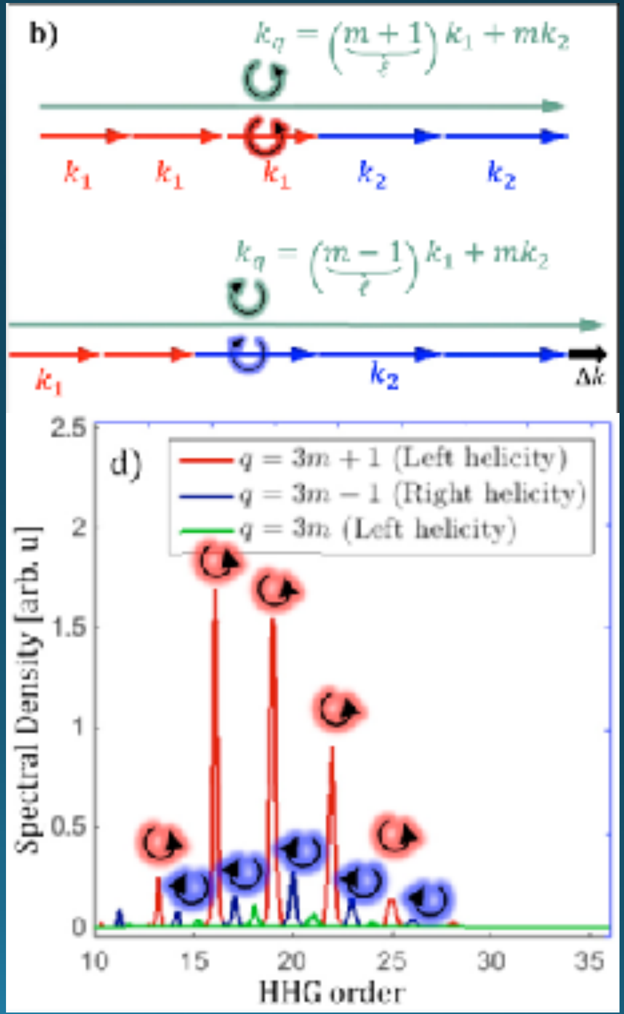
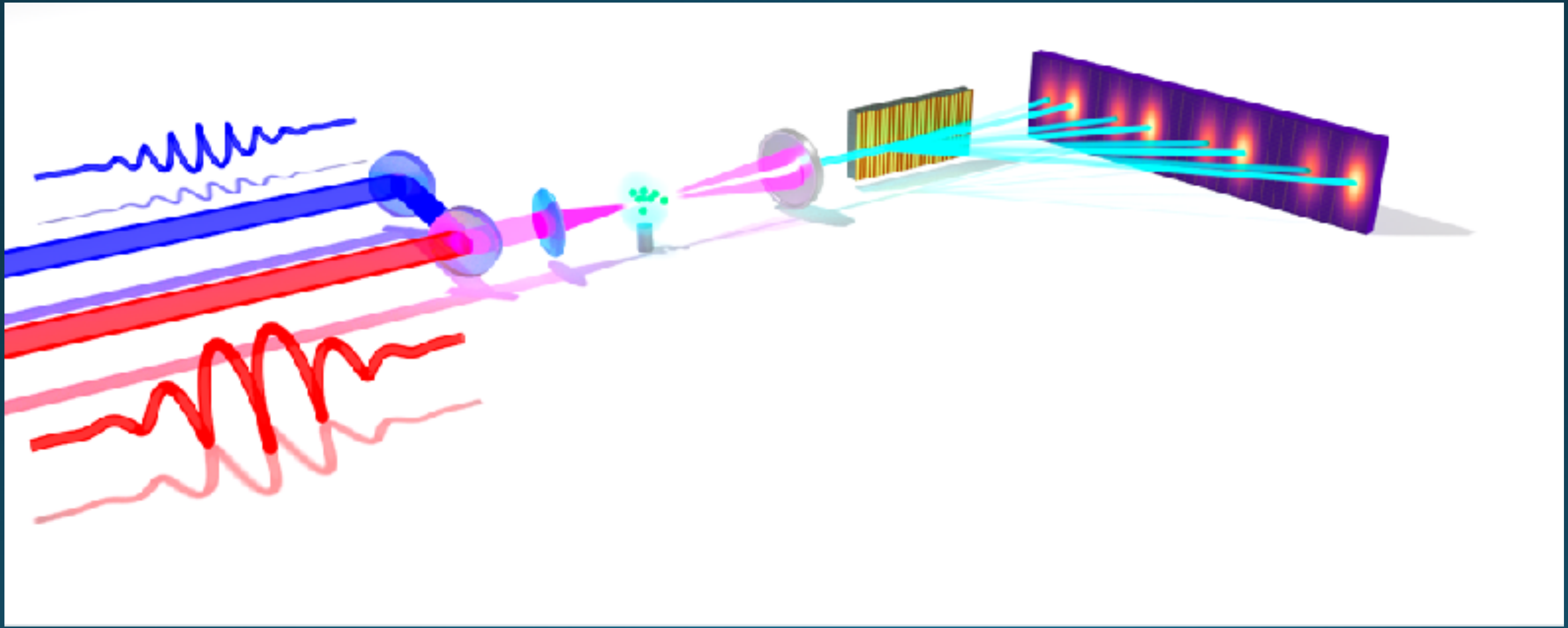


Macroscopic Control

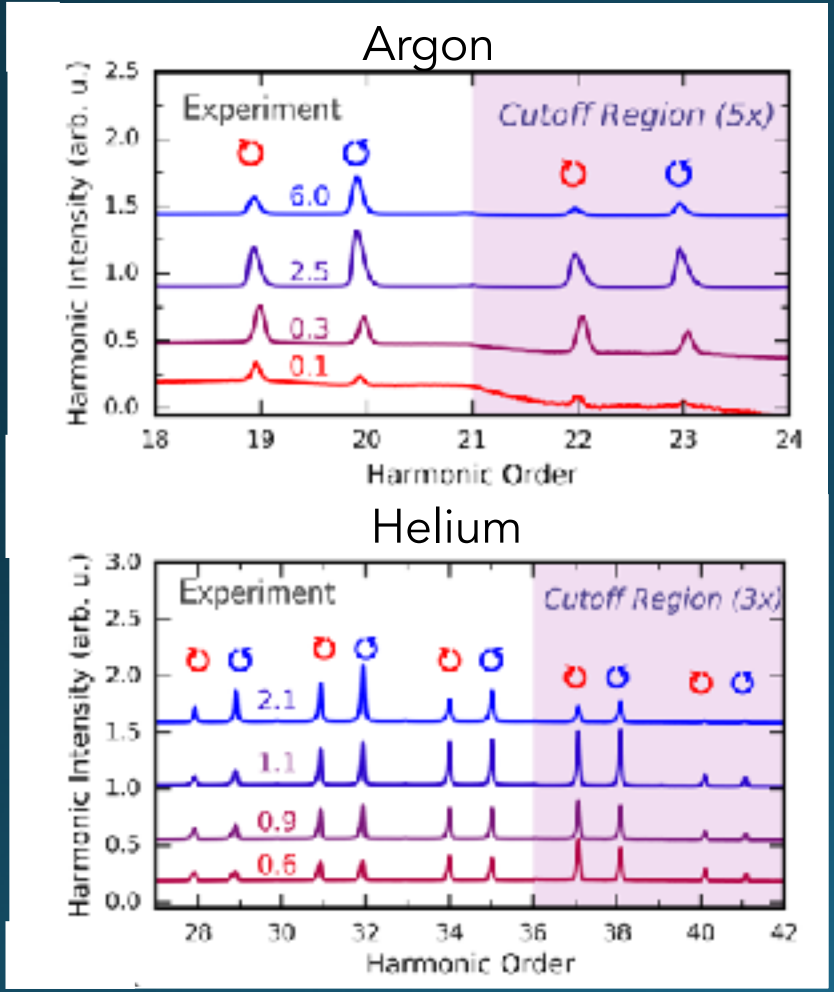


Active Control Over the Polarization of High-Harmonic Waveforms: Production of Elliptically Polarized Attosecond Pulse Trains!



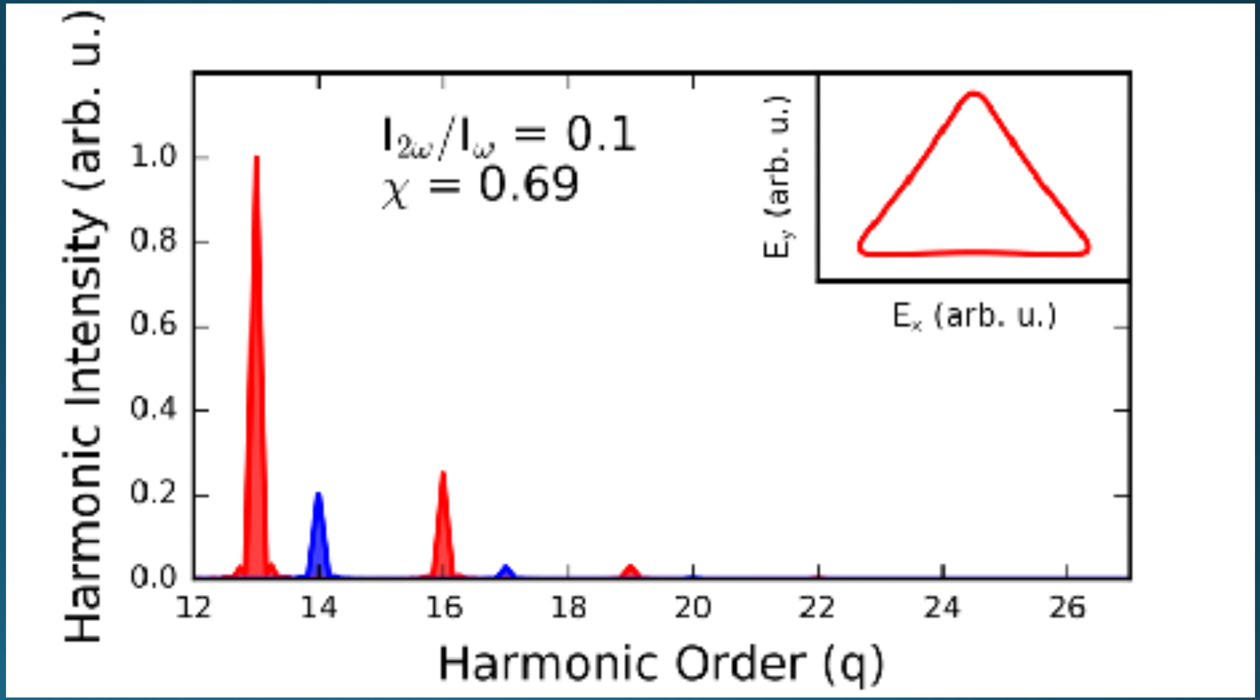
Dorney et. al., Phys. Rev. Lett., 2017, Accepted

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



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

SFA Simulation in Ar



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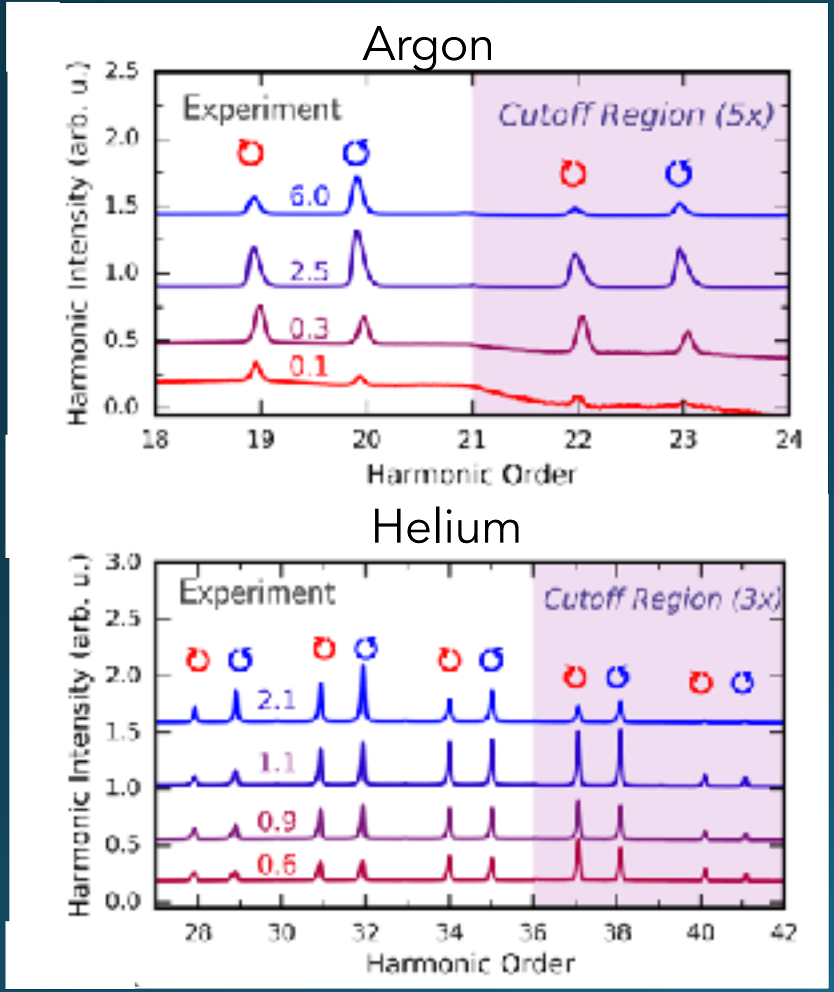

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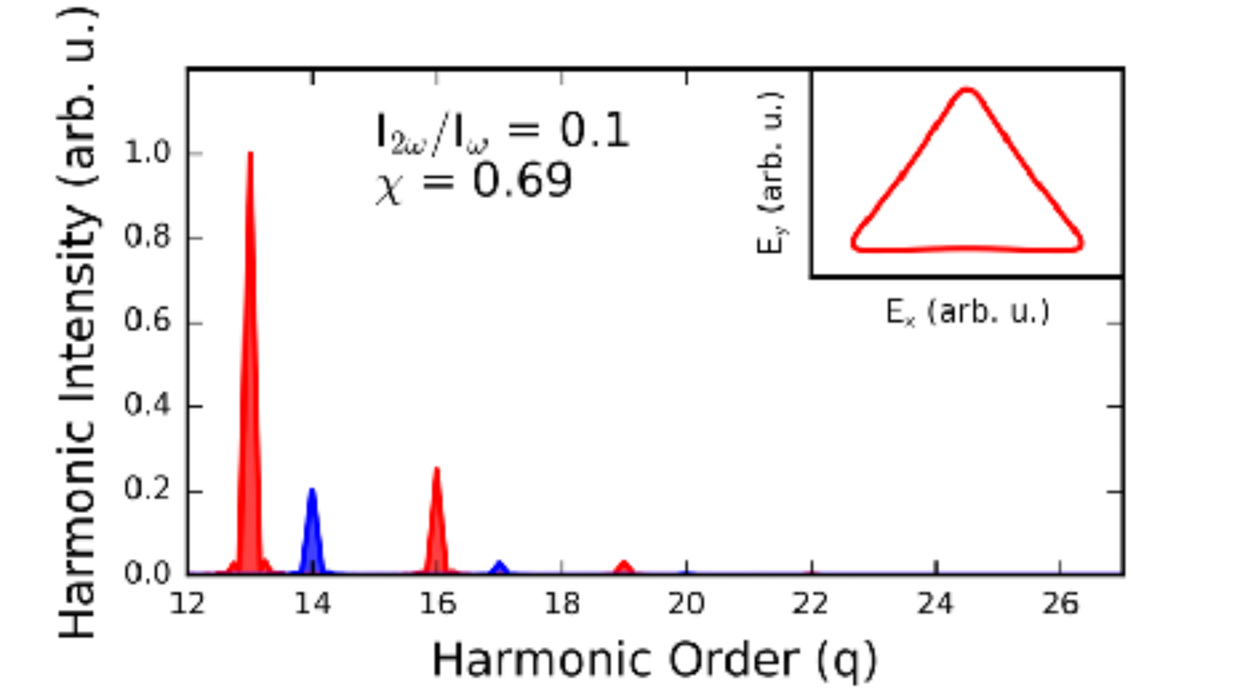

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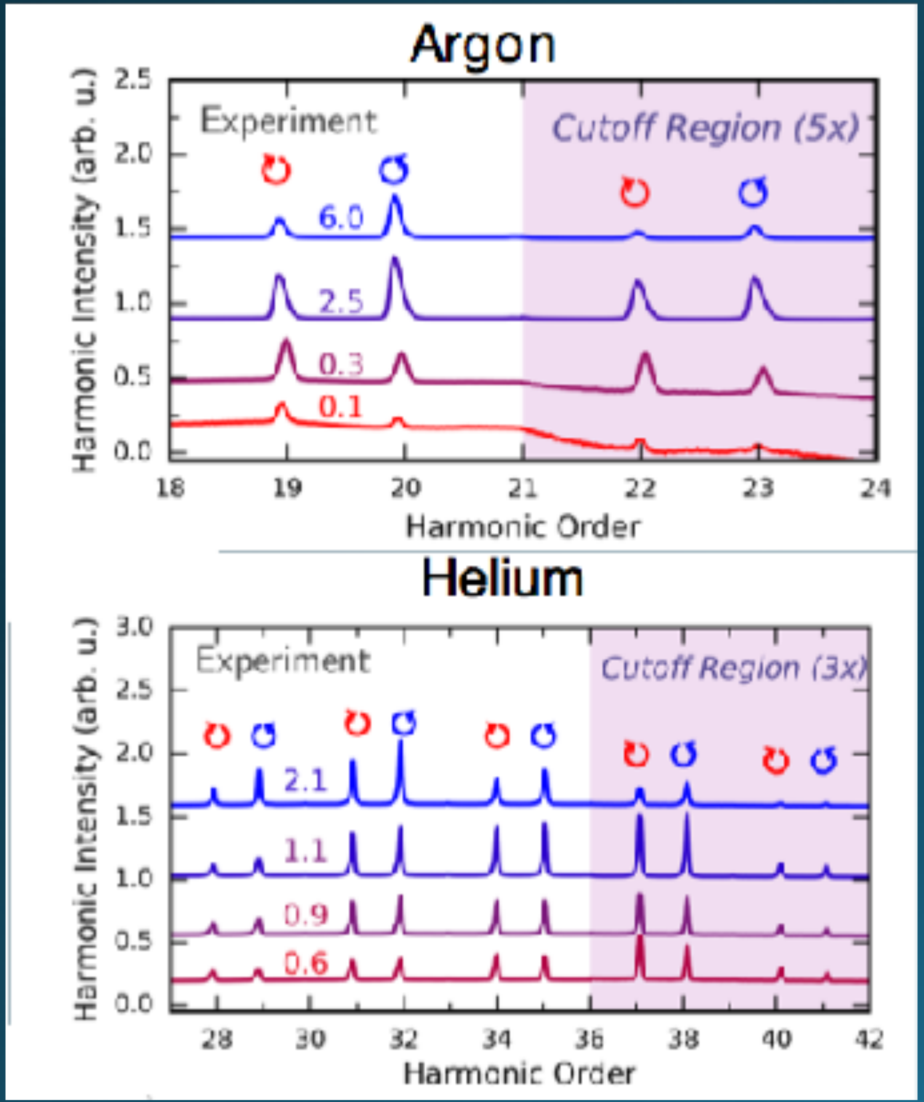
Chiral Control Independent of CPHHG Bandwidth



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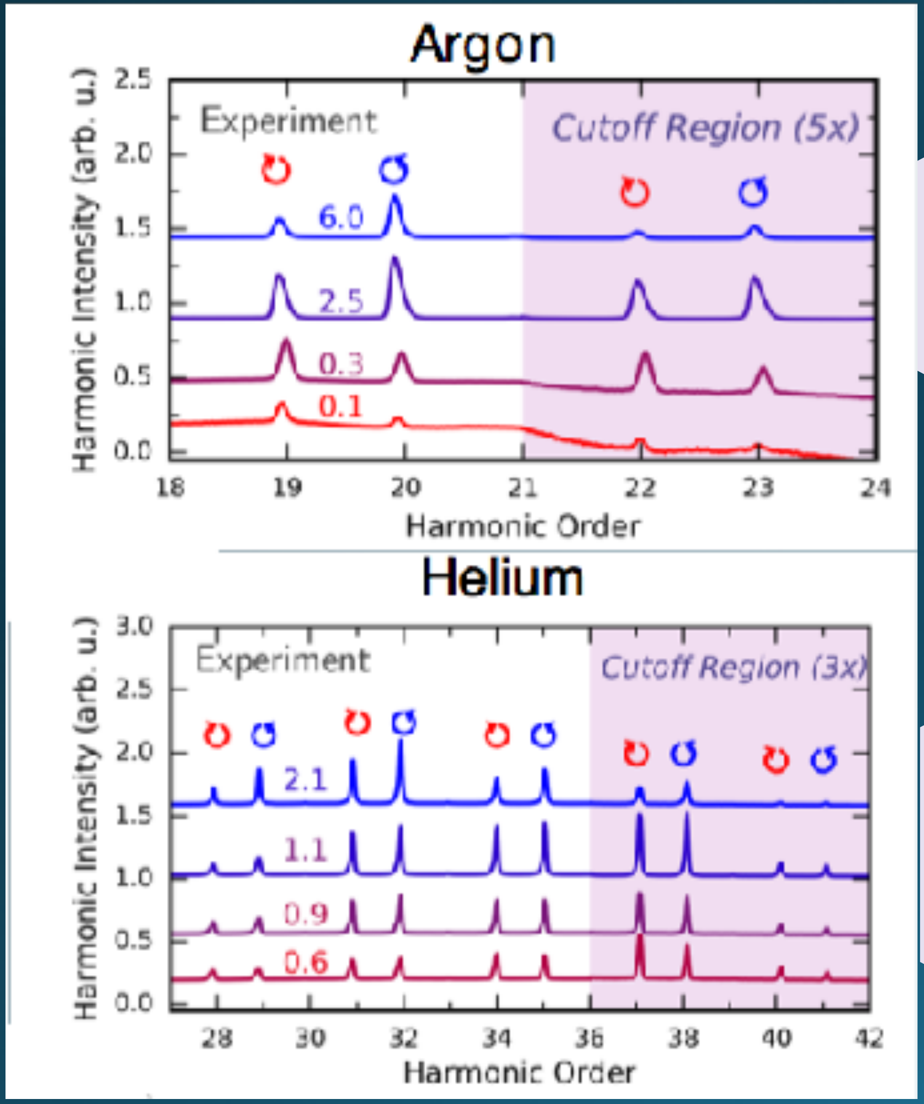

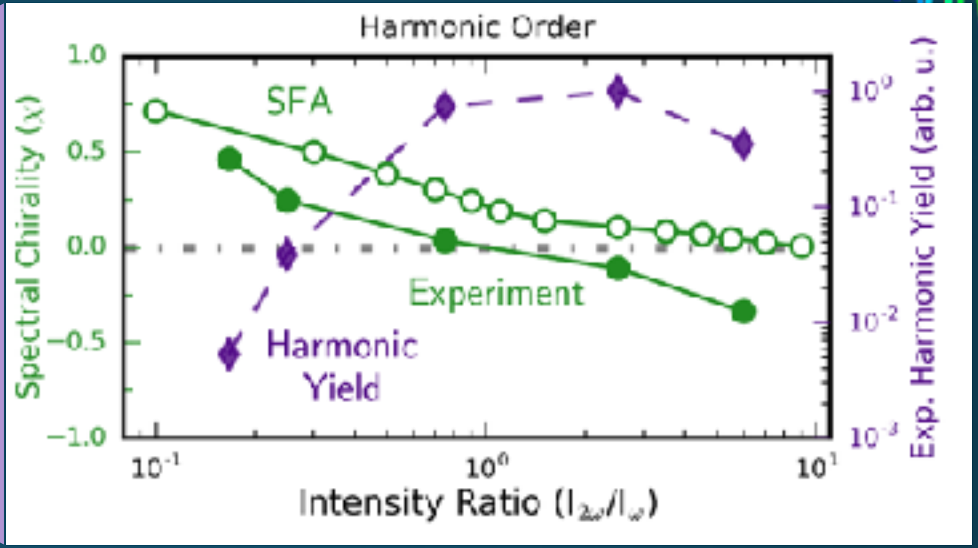

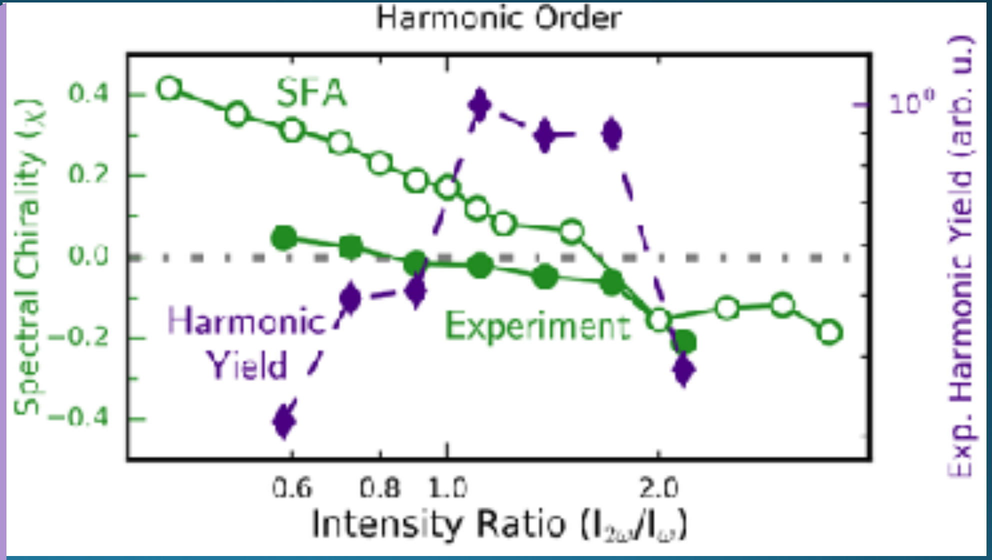

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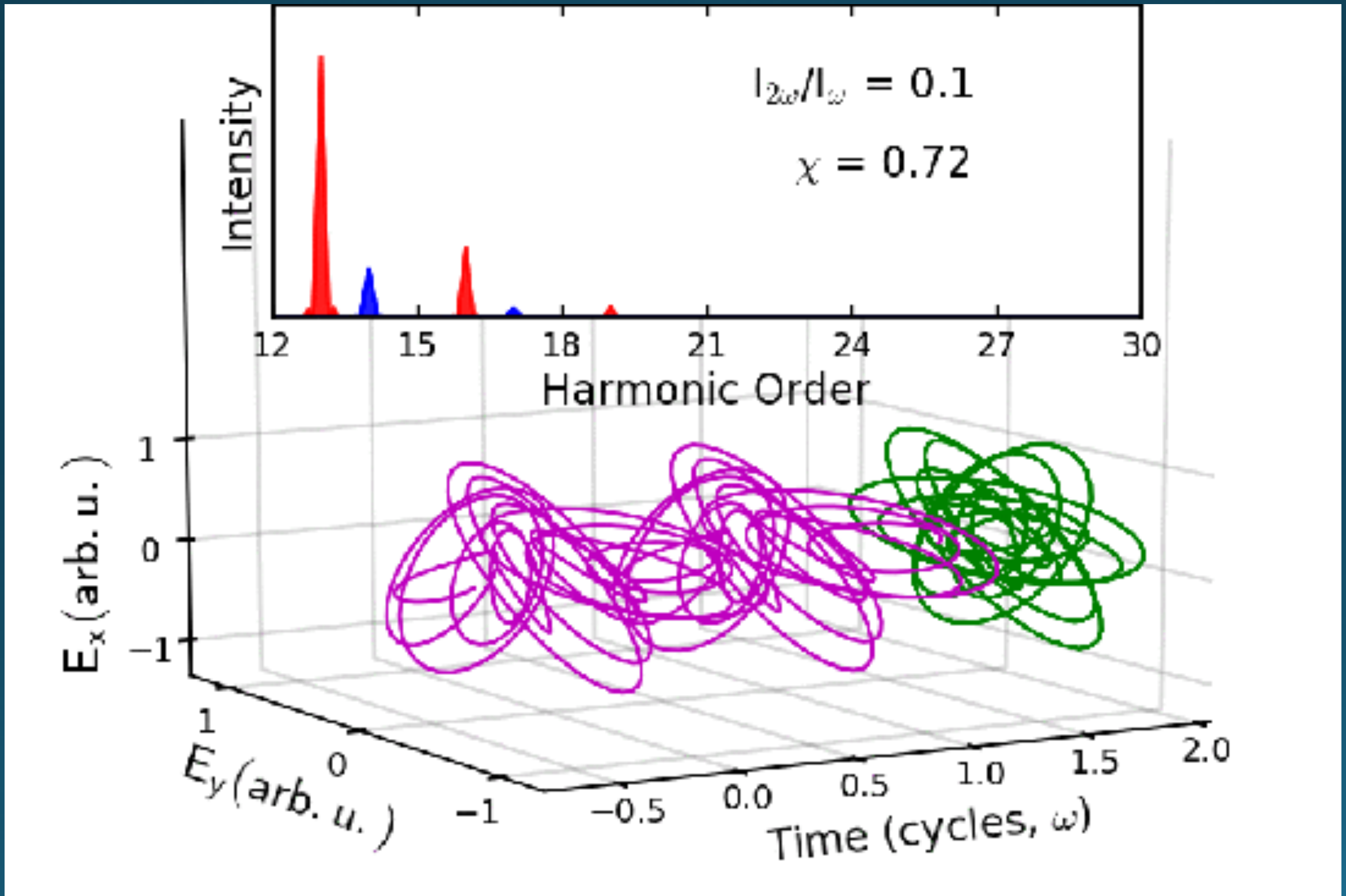
- **First demonstration of real-time polarization control of attosecond pulse trains in CPHHG!**



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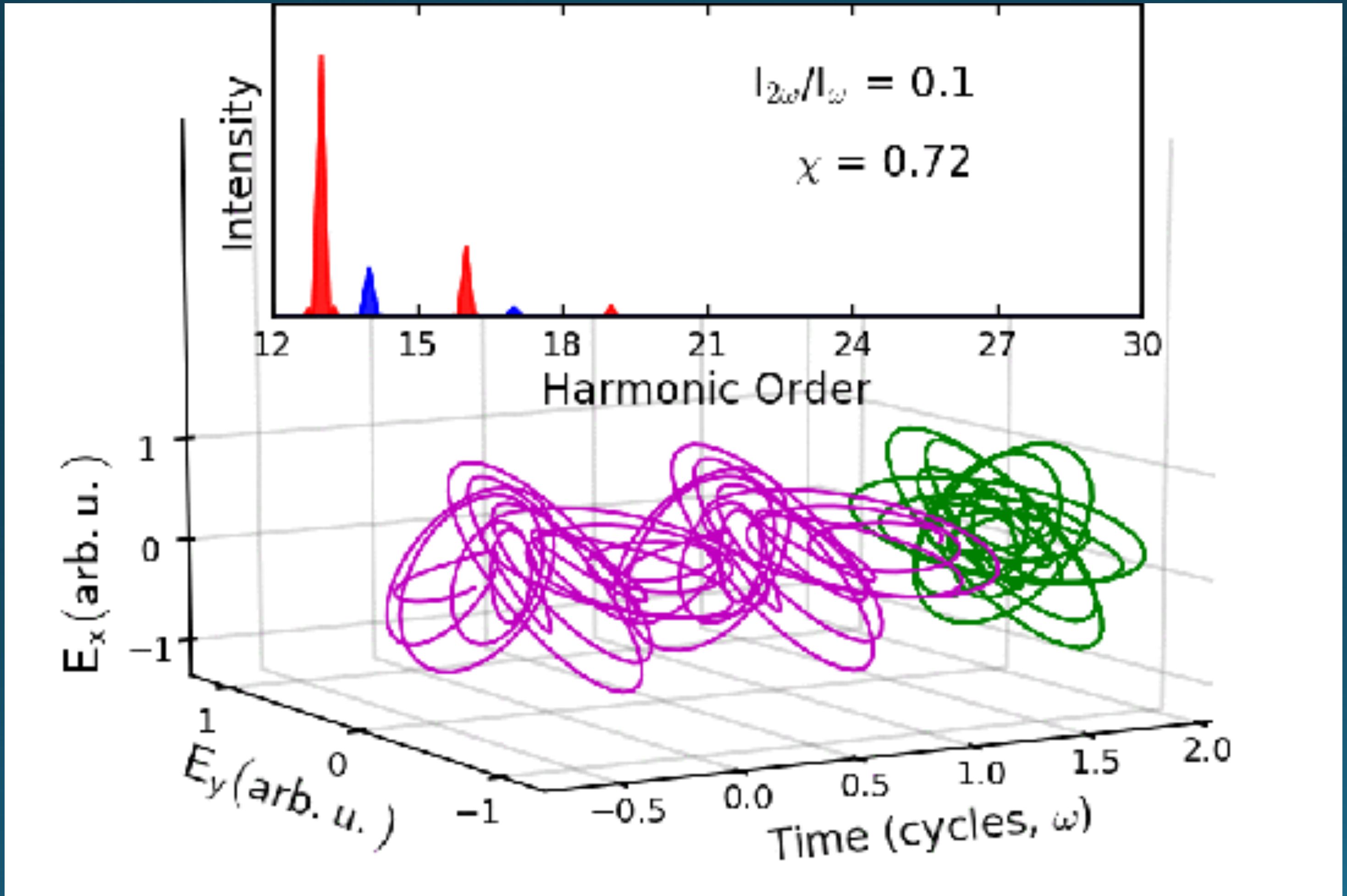
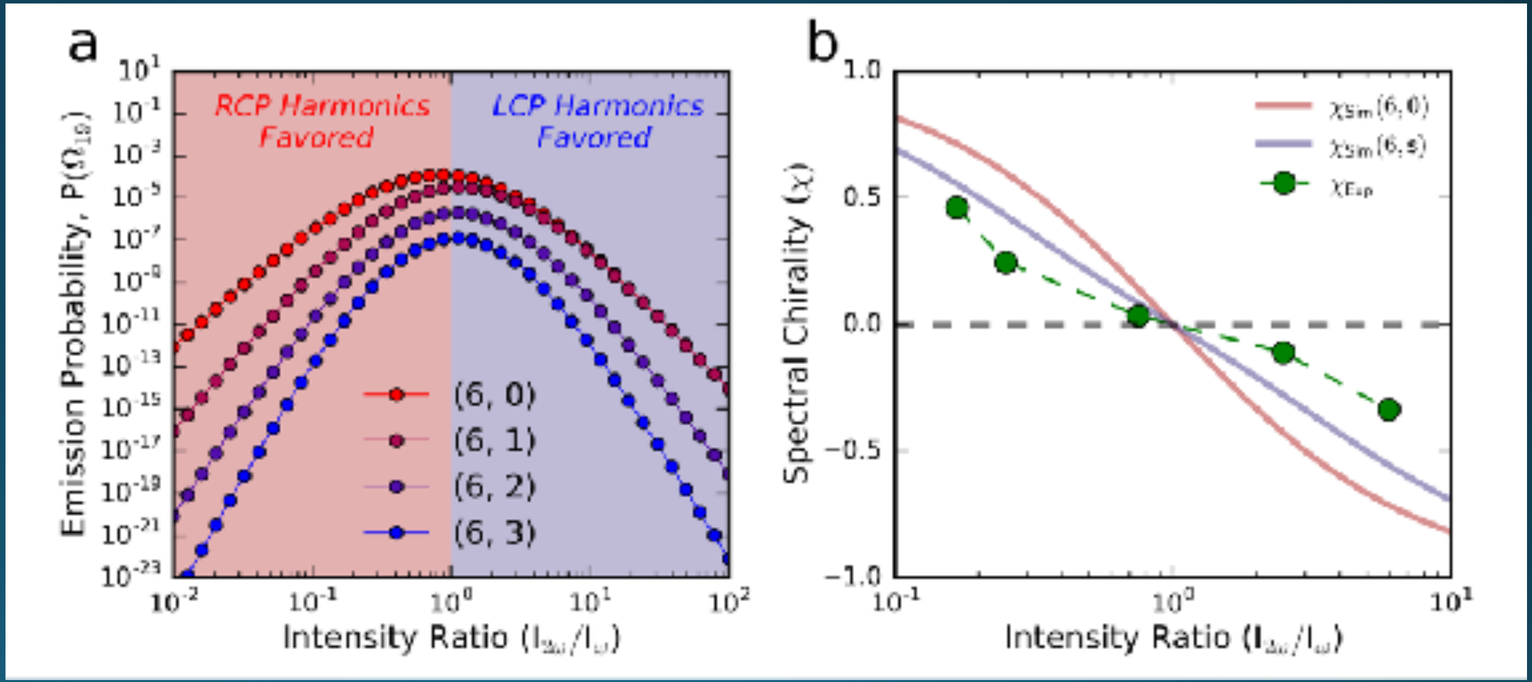


Simple Interpretation of Elliptical Control in CPHHG: Perturbative-esk Photon "Absorption"

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

Channel (n, s)	$H_{19,\ RCP}$ $(7\omega_1 + 6\omega_2)$	
	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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Attosecond Polarization Control at a Price…

Mancuso, Phys. Rev. A., 93, 2016
Dorney et. al., Phys. Rev. Lett., 2017, Accepted

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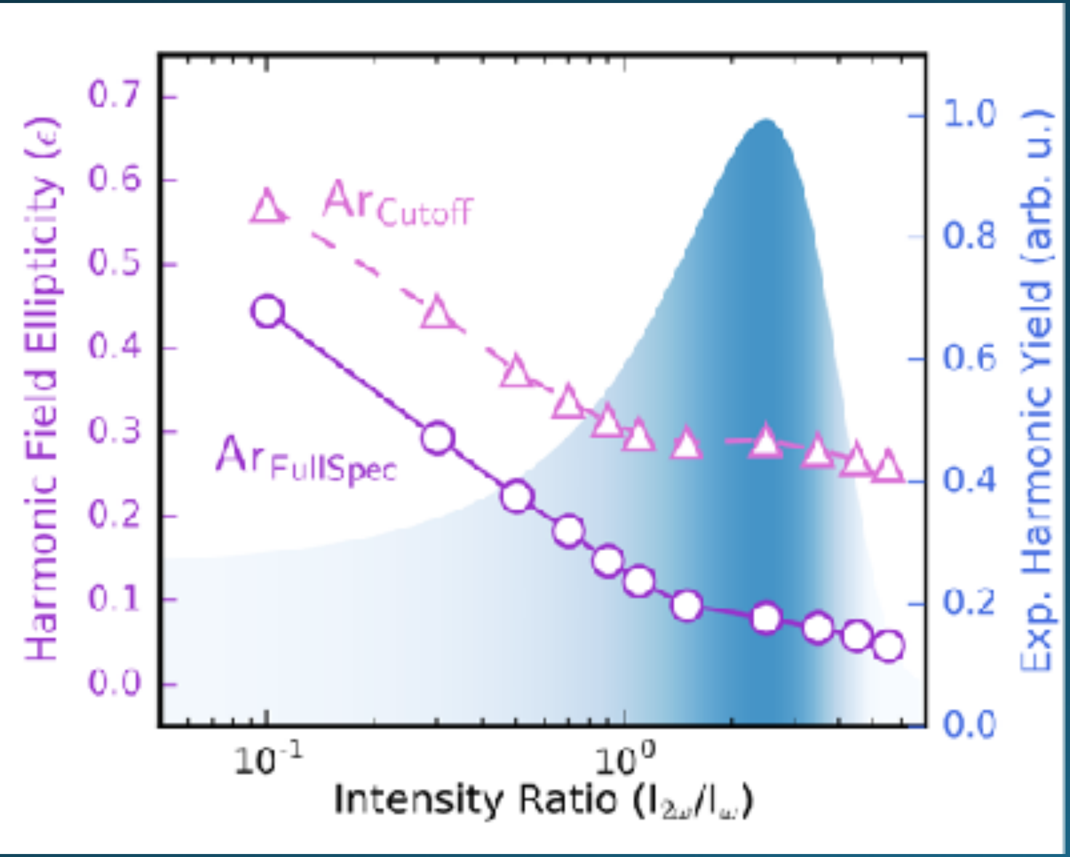

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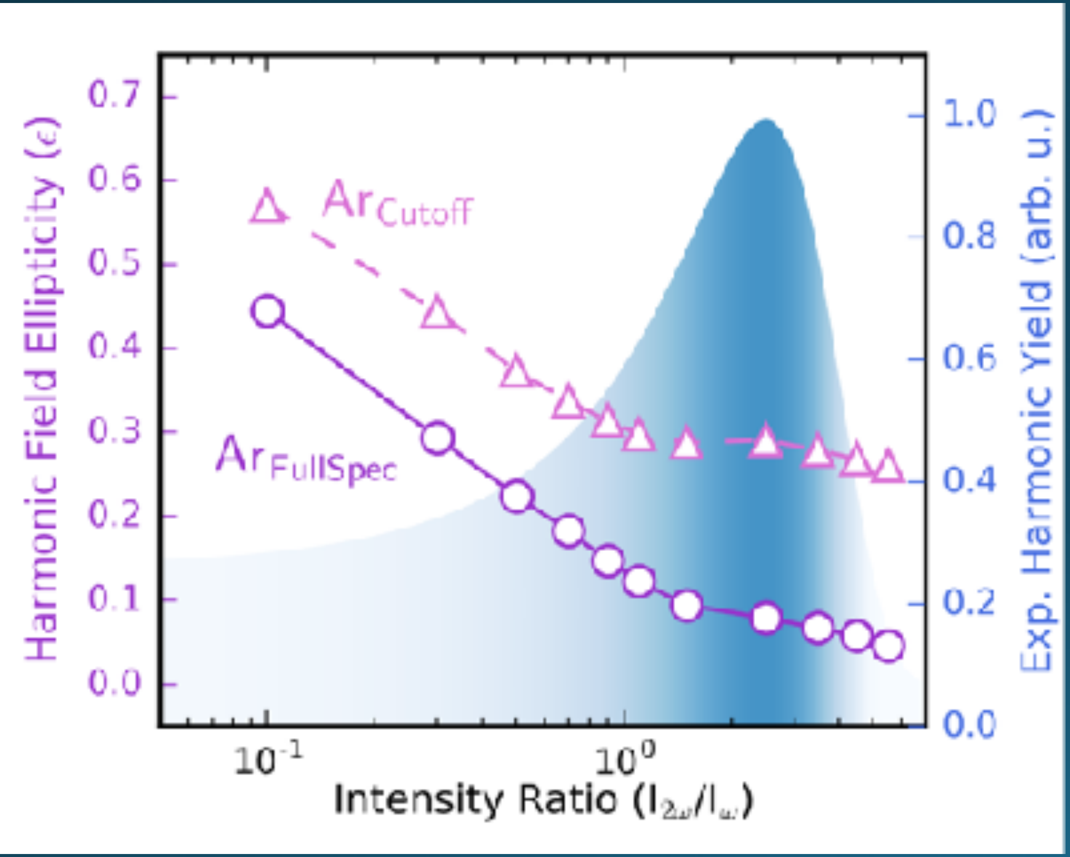

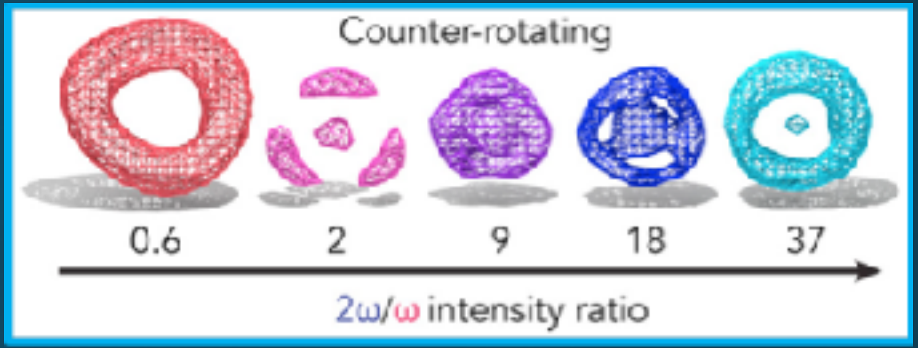

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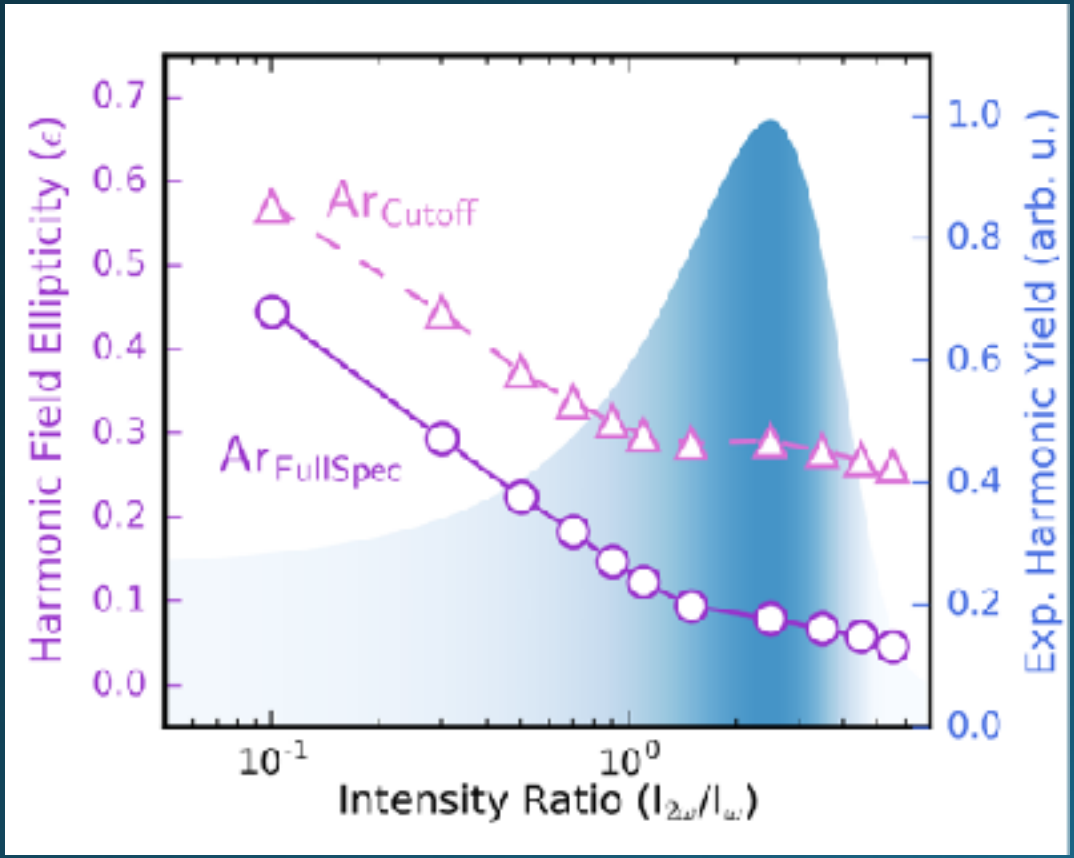

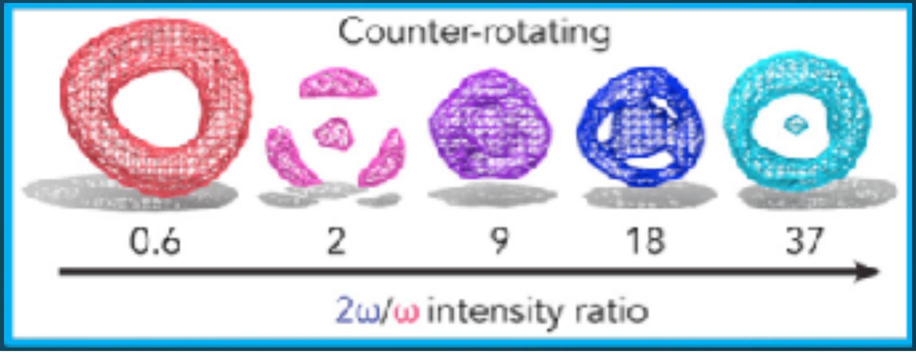

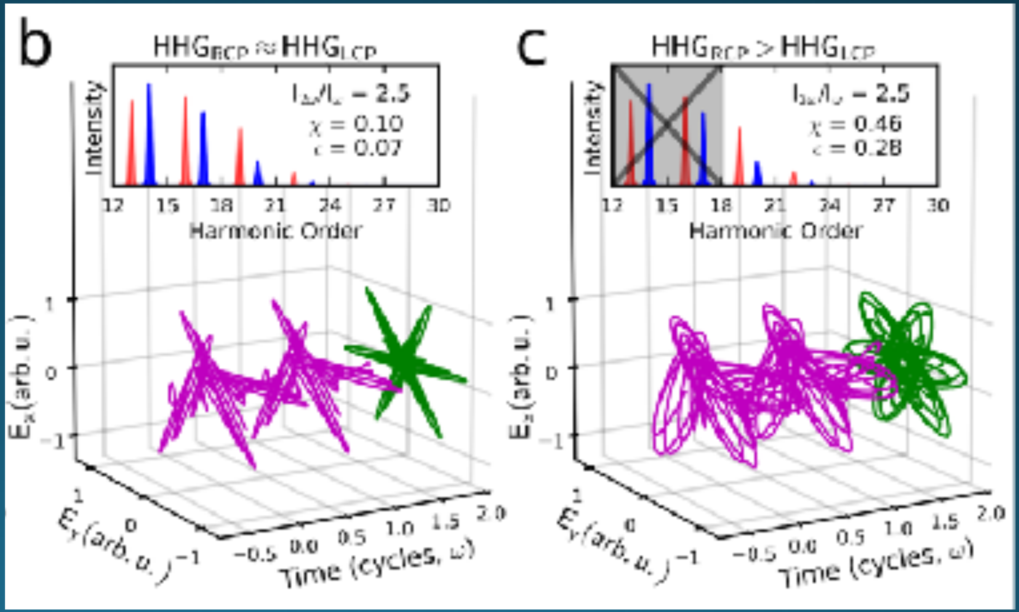

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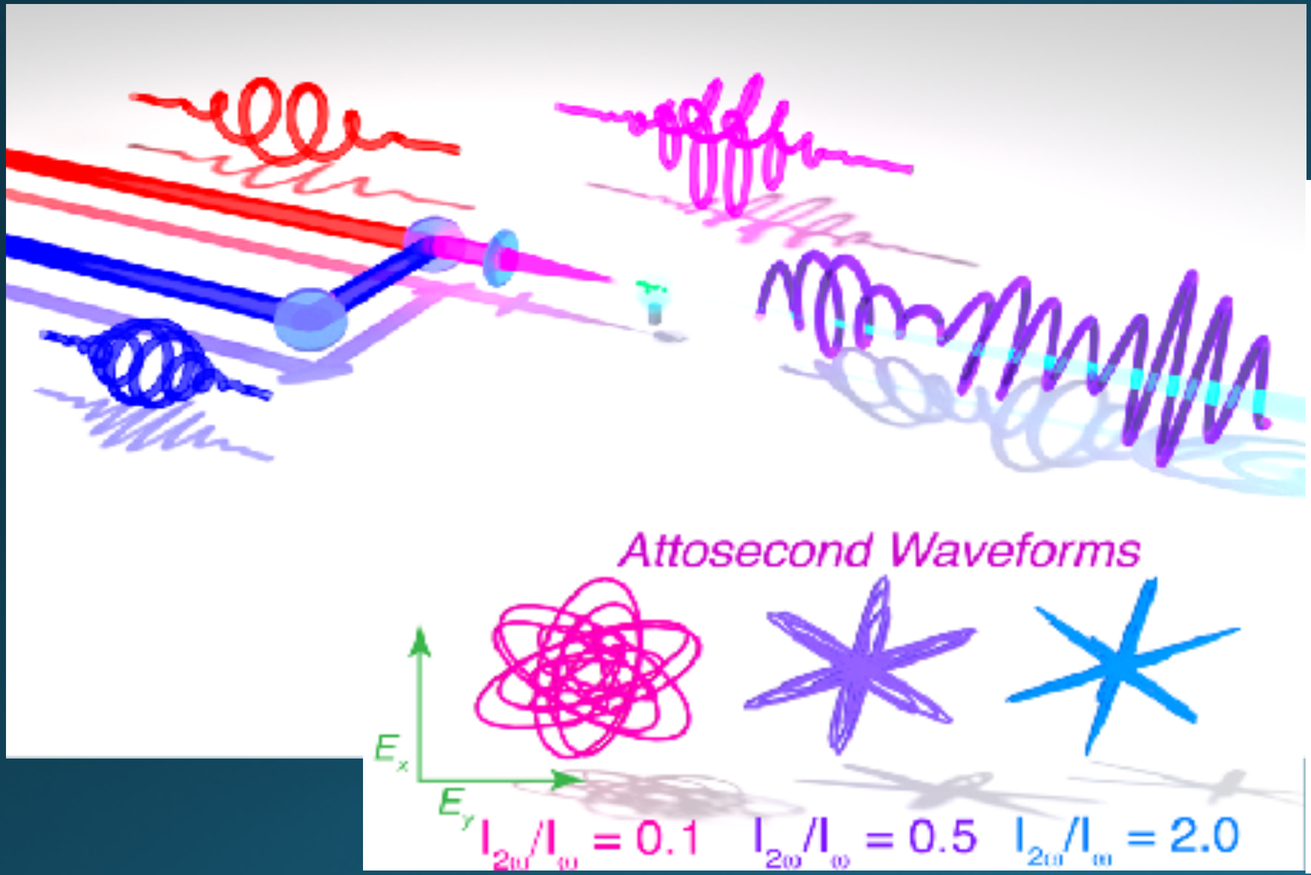
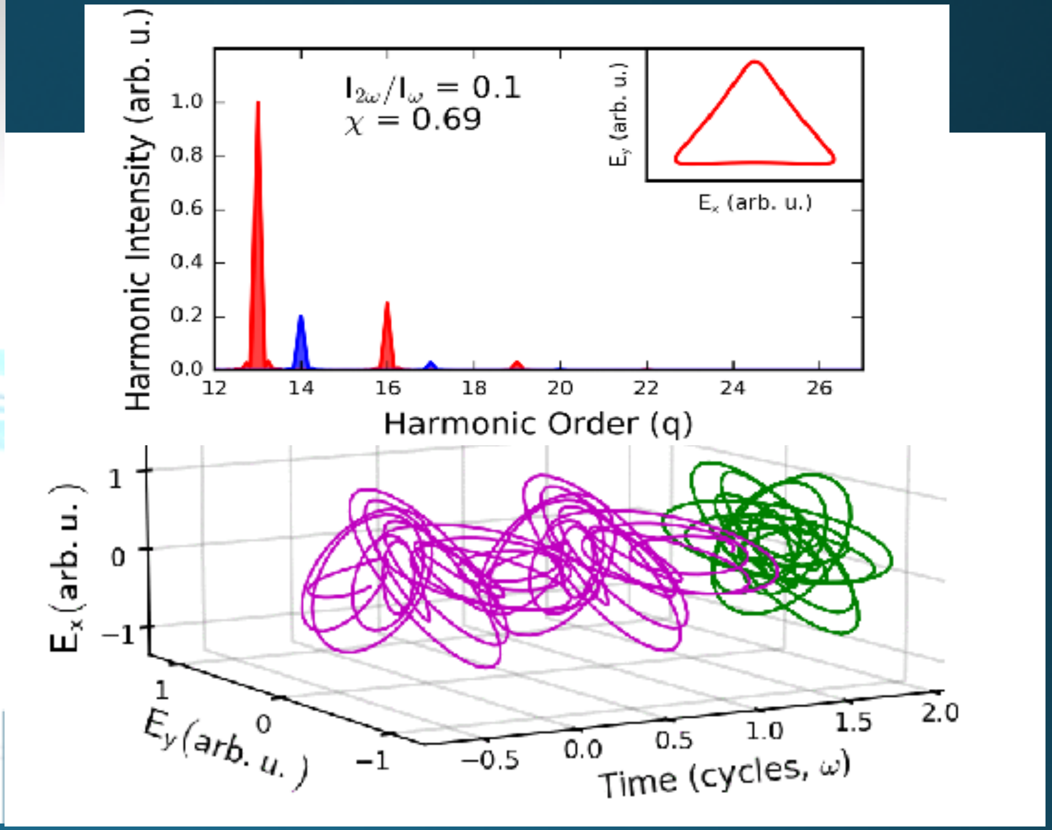
Production of Elliptically Polarized Attosecond Waveforms: Custom Attosecond Pulses for Chiral Spectroscopies





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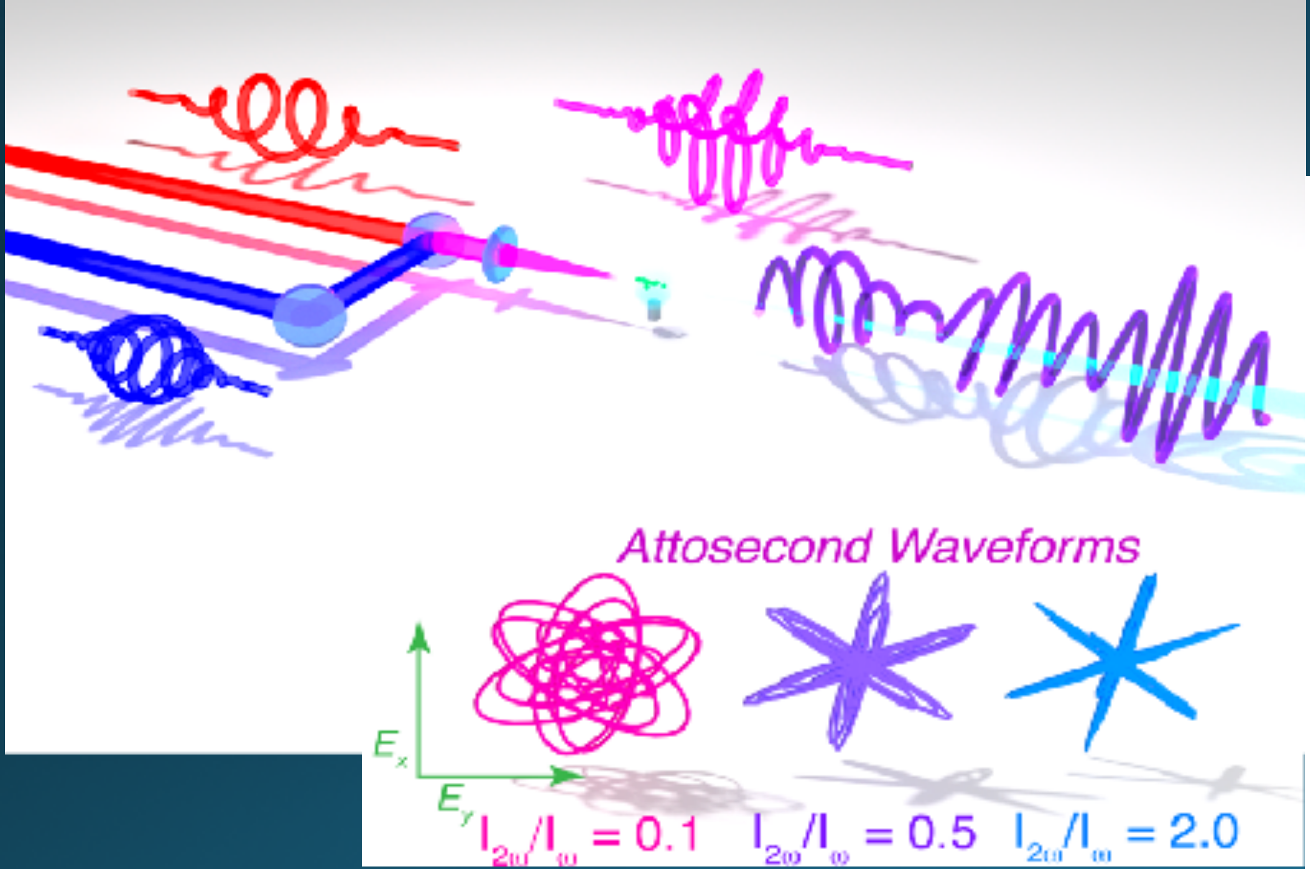

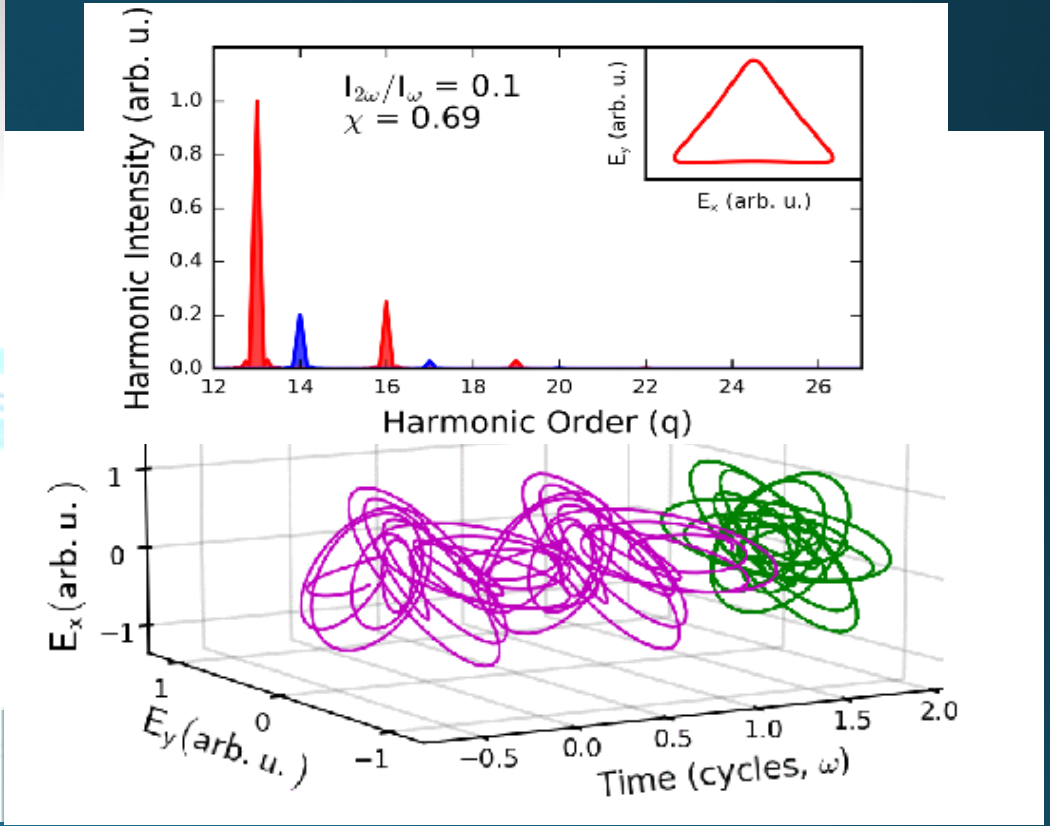

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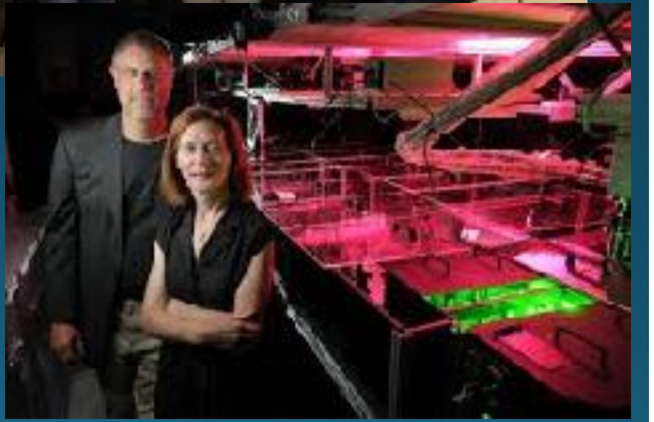
Excellent students, collaborators, and **advisors**

$$$$

<u>JILA, Univ. Colo. Boulder, NIST</u>

Jennifer L. Ellis, Daniel D. Hickstein,
Christopher A. Mancuso, Nathan Brooks,
Tingting Fan, Patrik Grychtol,
Dmitriy Zusin, Christian Gentry

Henry C. Kapteyn
Margaret M. Murnane



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