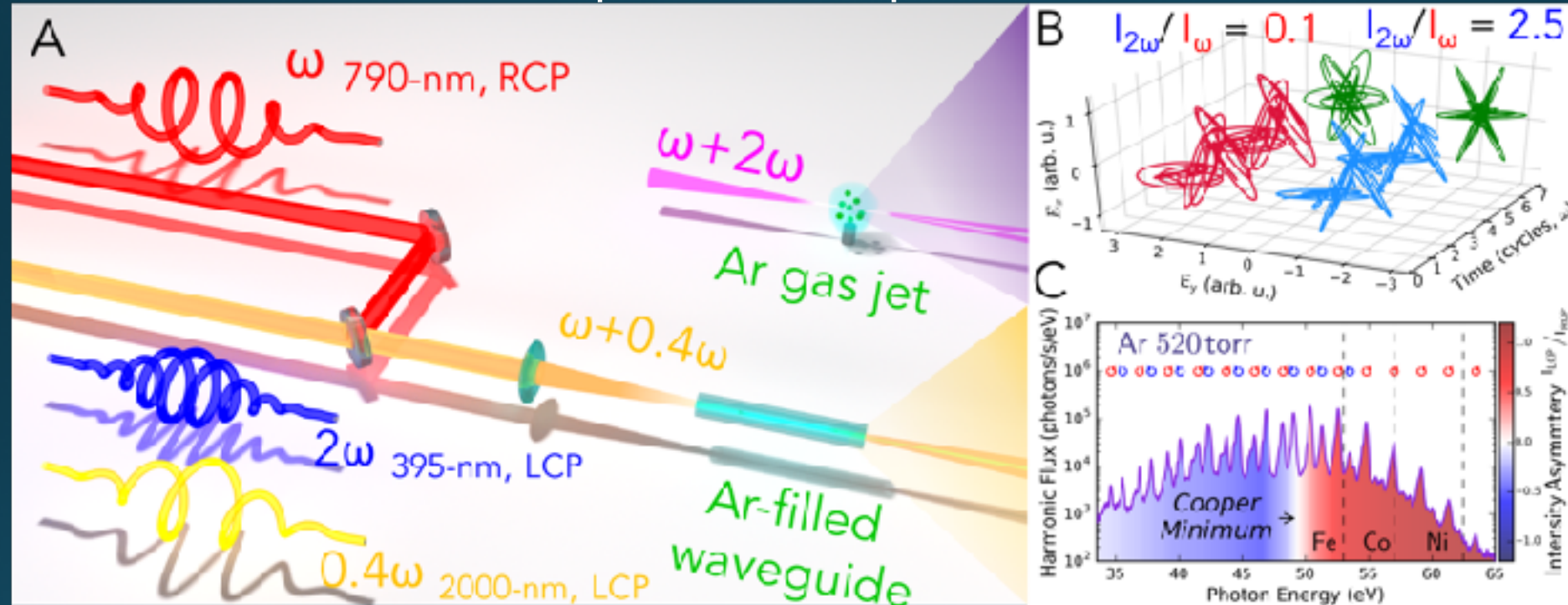


Helicity Gets a New Twist: Straightforward Production of Polarization Sculpted High-Harmonic Attosecond Waveforms for Chiral Spectroscopies and Imaging



Kevin M. Dorney

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



From Snowy Mountains to Warm... Beautiful...
Snow-covered Beaches?





From Snowy Mountains to Warm... Beautiful... Snow-covered Beaches?

Boulder, CO (10-02-2018)





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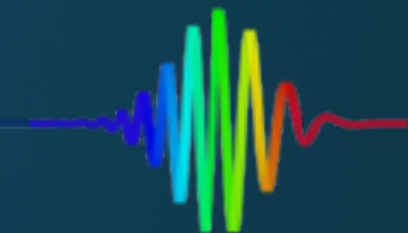


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Boulder, CO (10-02-2018)



Barcelona, ESP (10-02-2018)



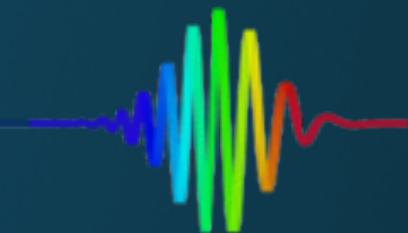


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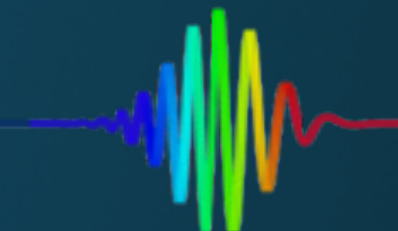


Barcelona, ESP (10-02-2018)





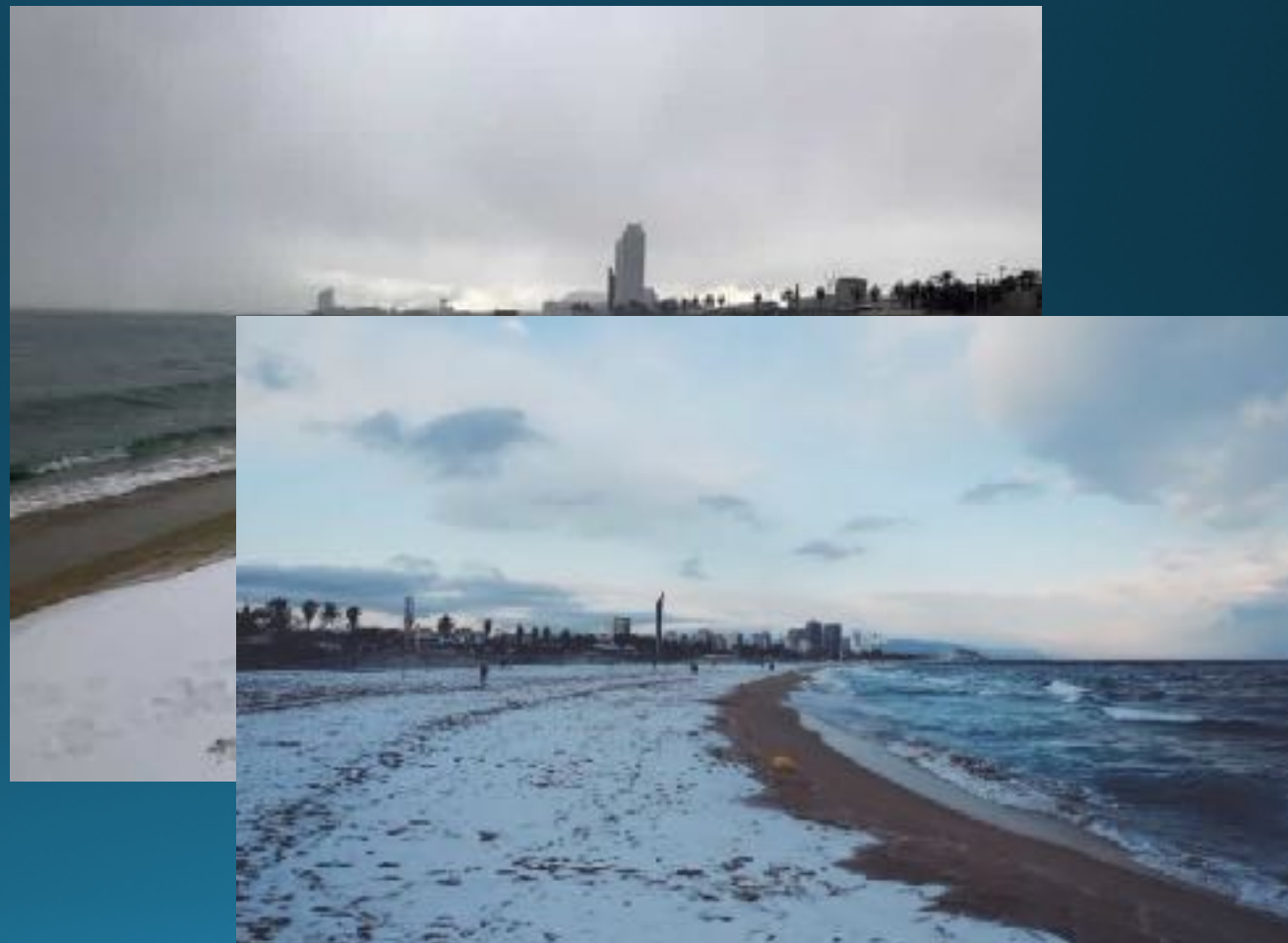
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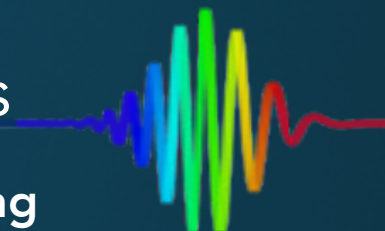
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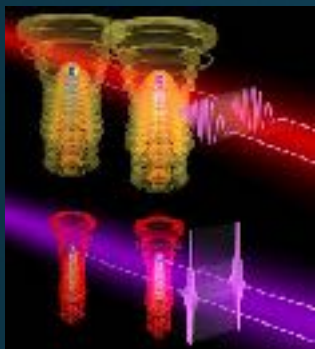
Barcelona, ESP (10-02-2018)



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

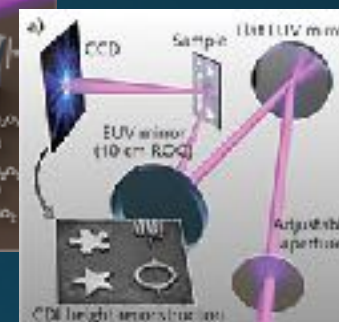
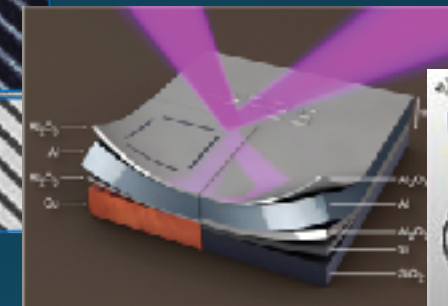


Attosecond Extreme Nonlinear Optics



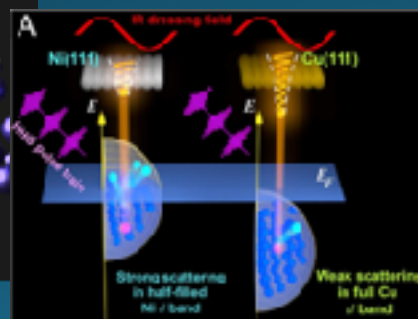
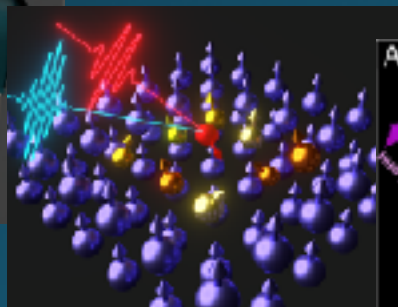
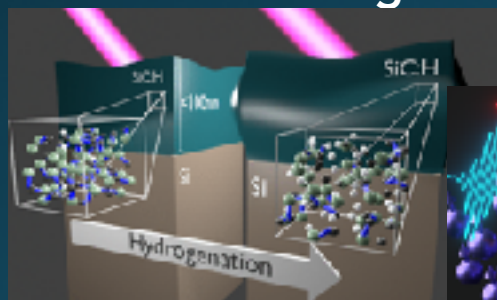
Popmintchev, Science 6265, 2015
Dorney, PRL 118, 2017

Coherent X-ray Imaging



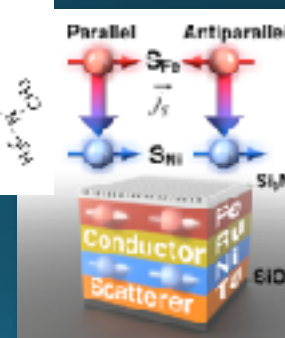
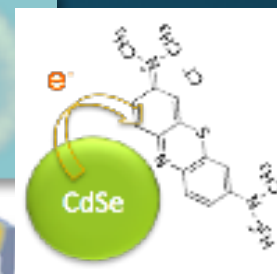
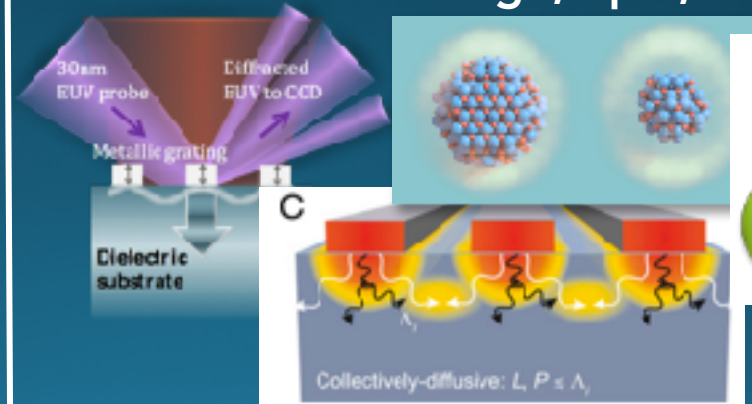
Gardener, Nat. Photon. 11, 2017
Shanblatt, Nano Lett. 16, 2016

Uncovering New Ultrafast Materials Science



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

Nanoscale Charge, Spin, Energy Transport



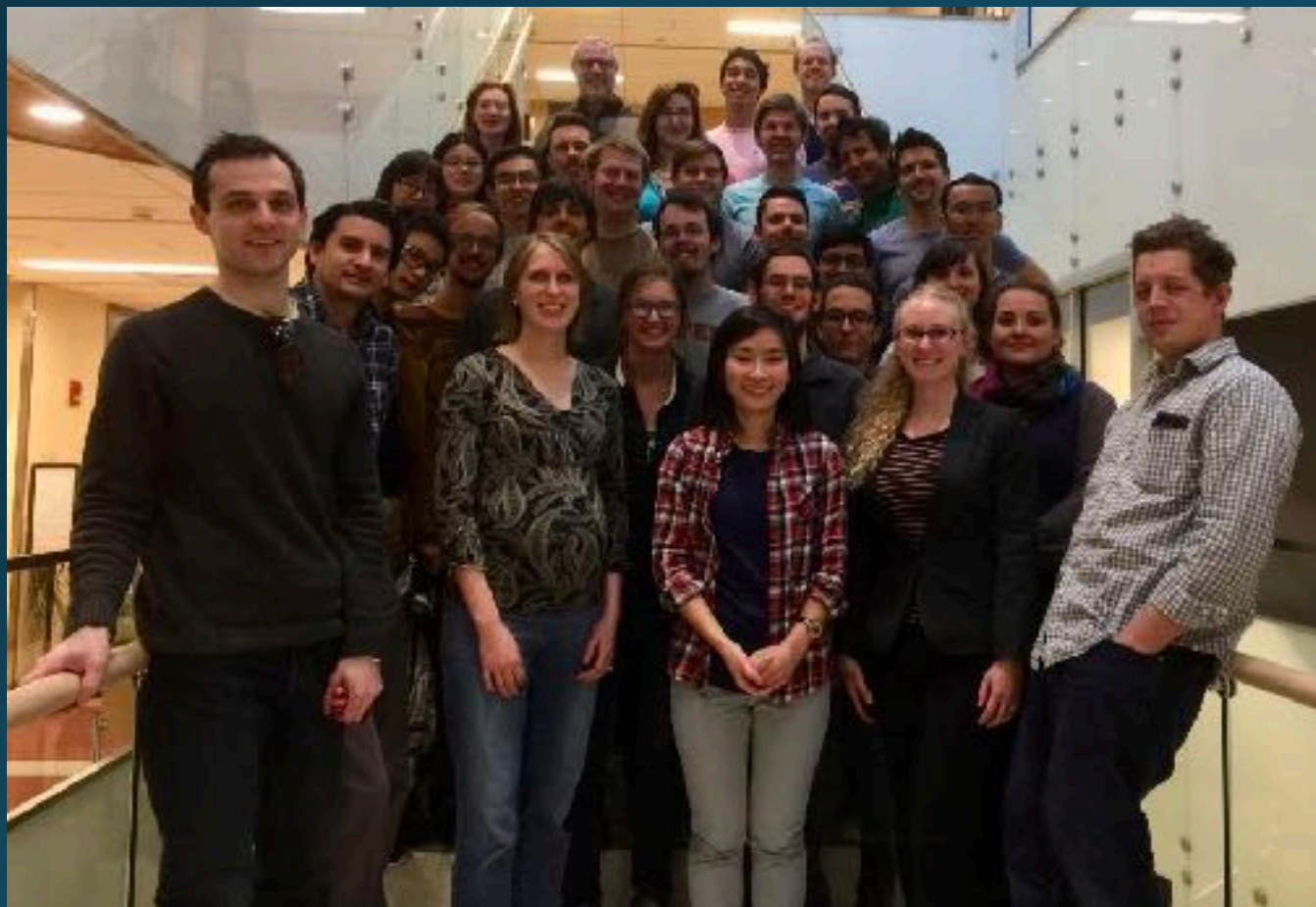
Hoogeboom-Pot, PNAS 112, 2015
Ellis, JACS, 11 2015



KM Group and JILA: Excellent students, collaborators, and **advisors**

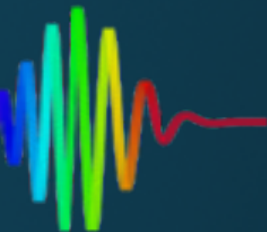


KM Group Spring 2017





KM Group and JILA: Excellent students, collaborators, and **advisors**



KM Group Spring 2017



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KM Group Spring 2017



Collaborators



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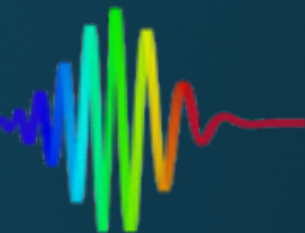


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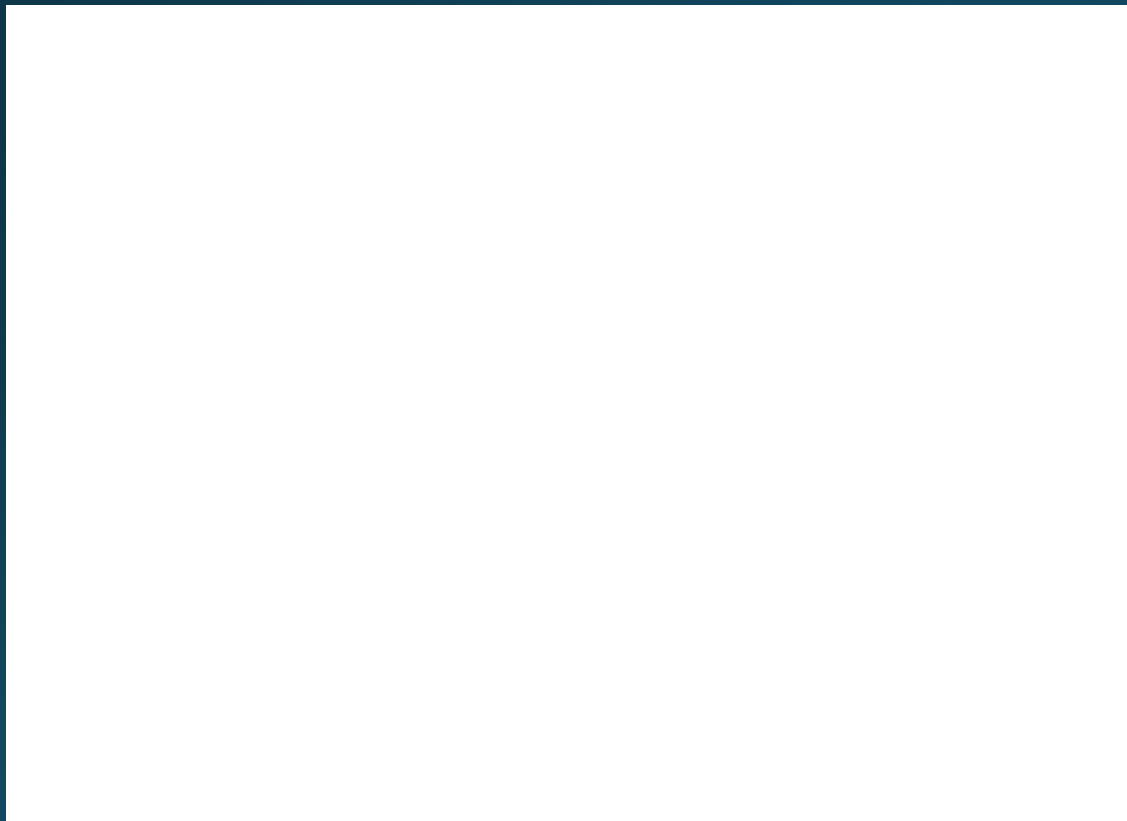


High Harmonic Generation (HHG): Light Science at the Atomic Frontier and Beyond



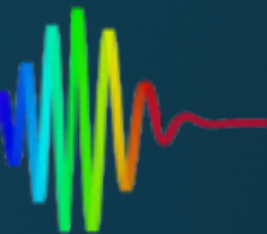
The White Whale of the Physical Sciences

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



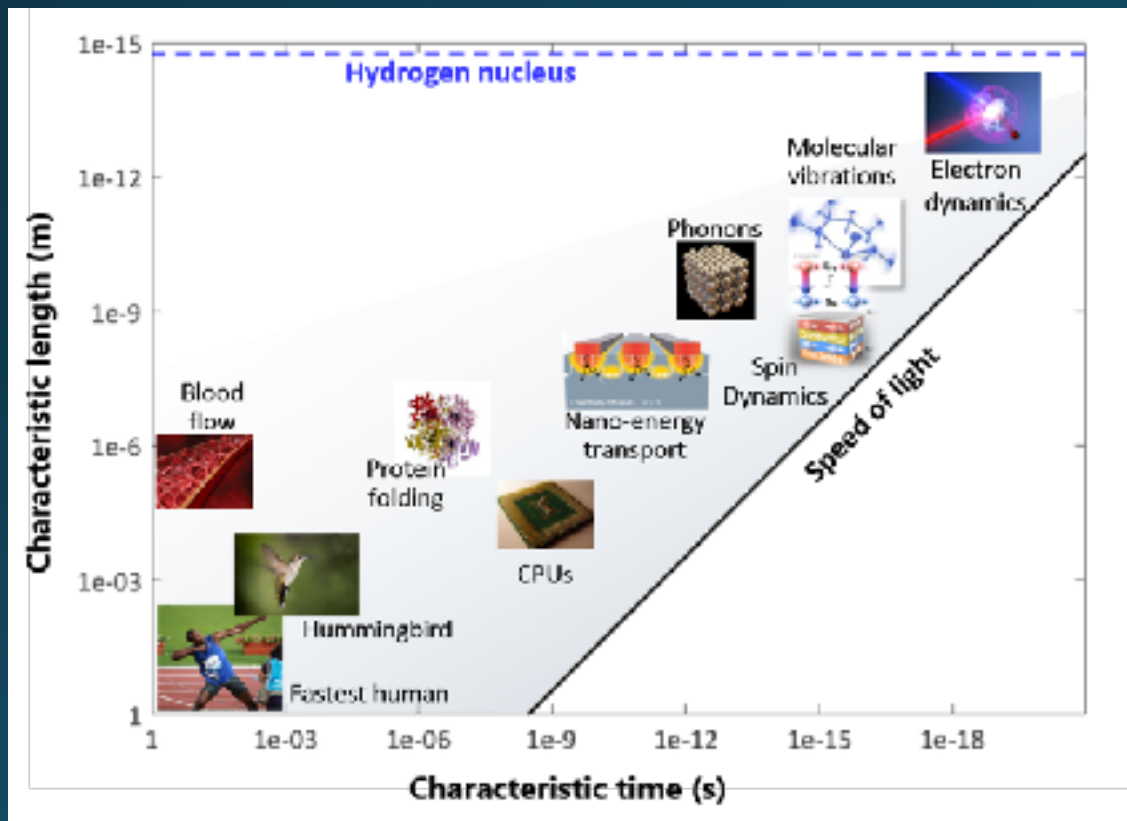
Courtesy: Nico Hernandez Chupak, KM Group

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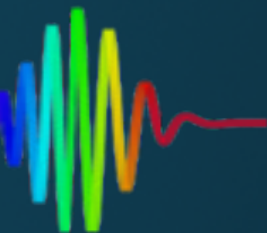
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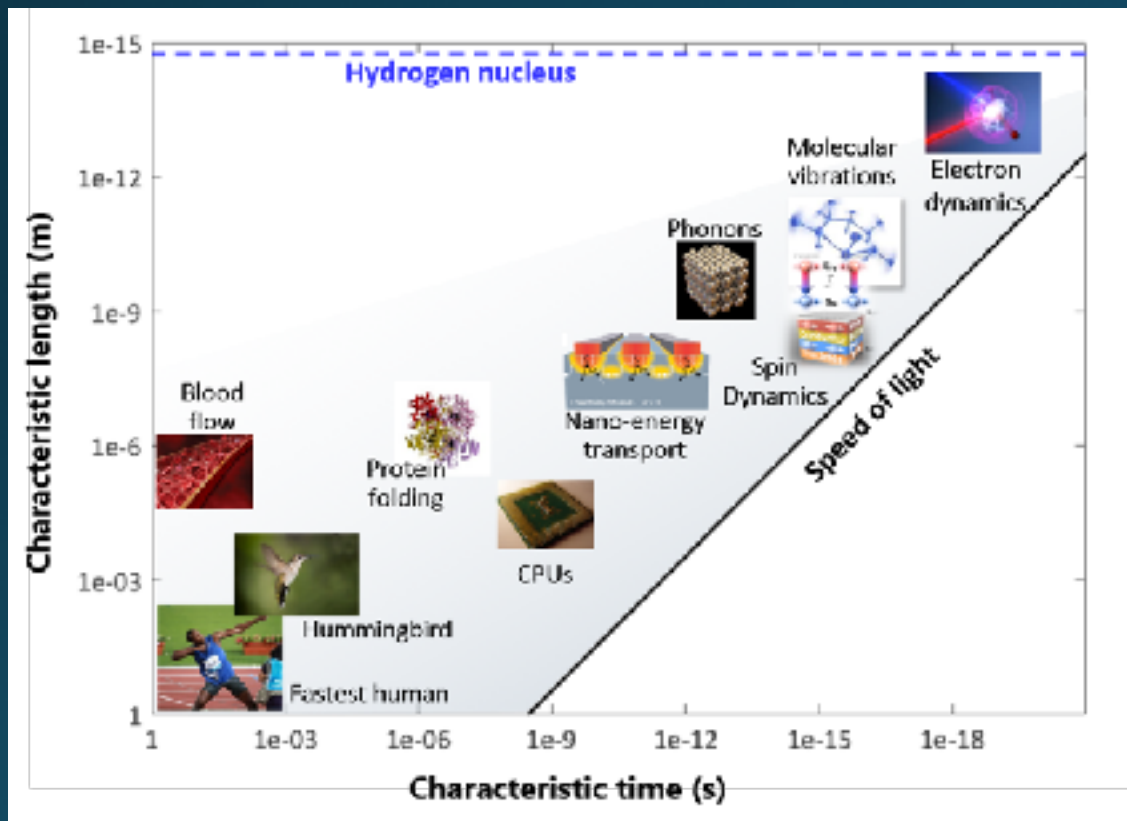
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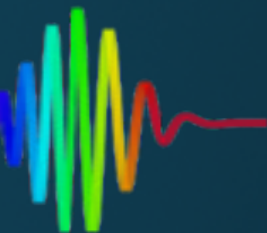


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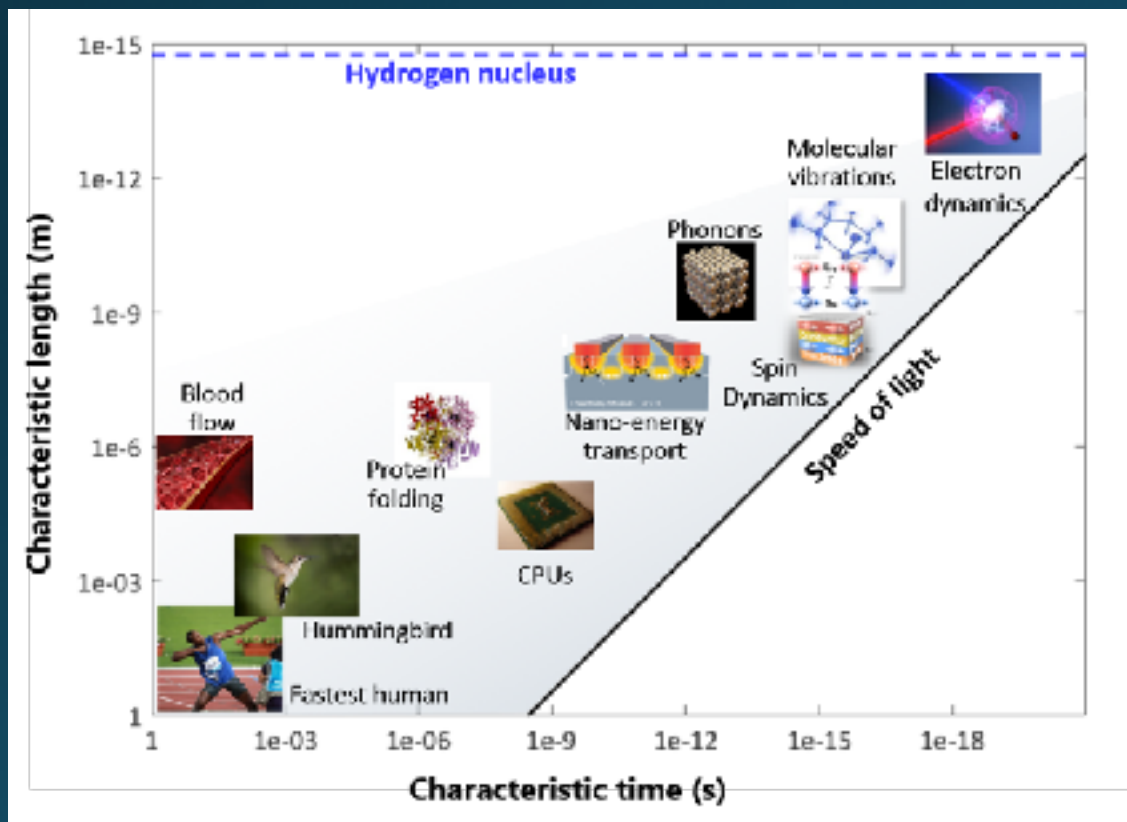


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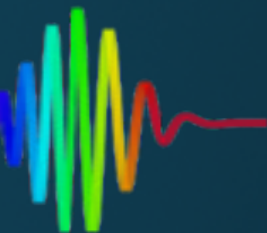
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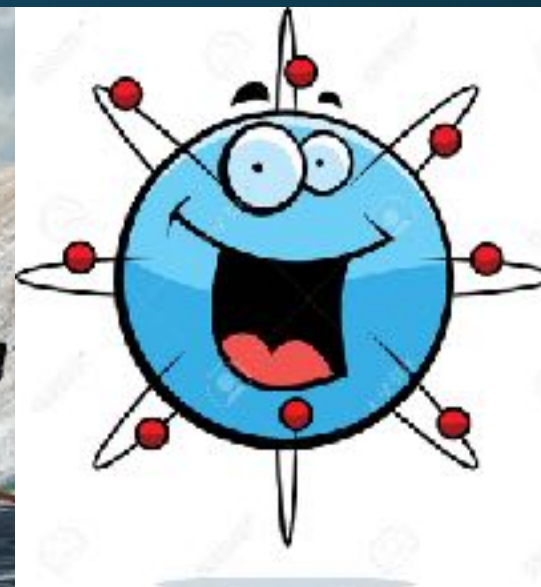
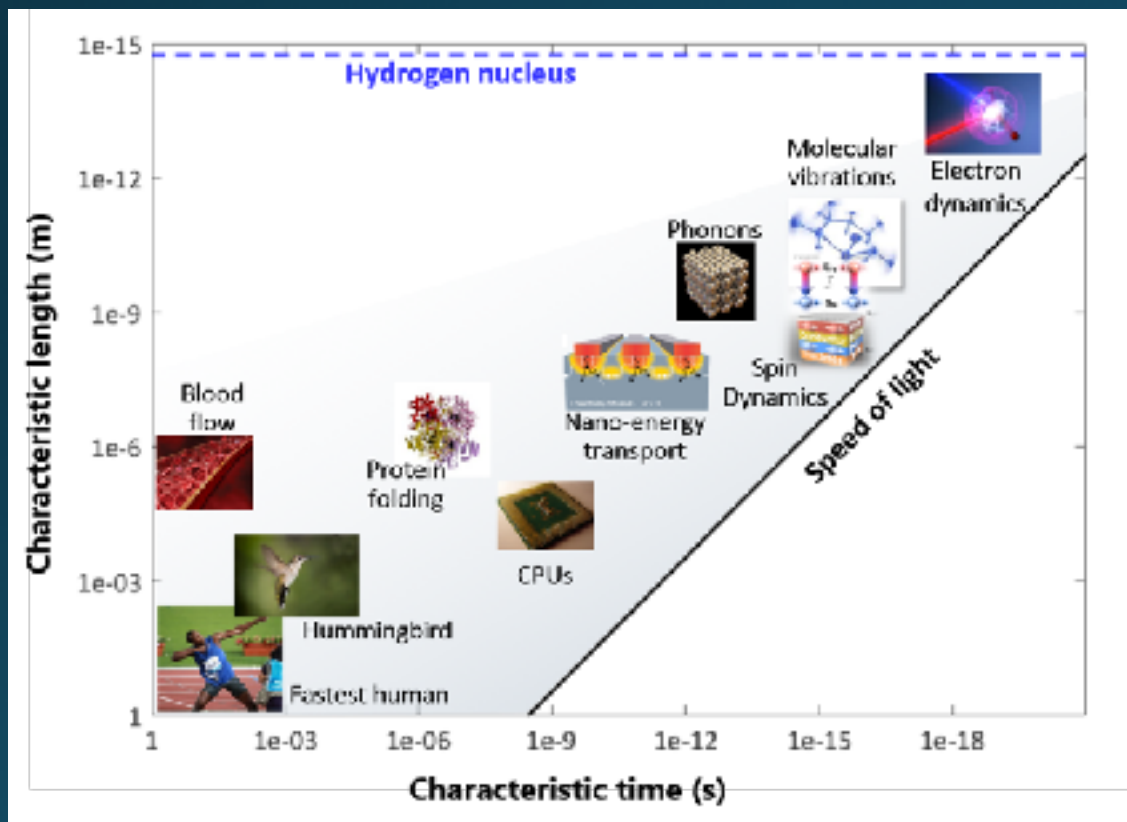
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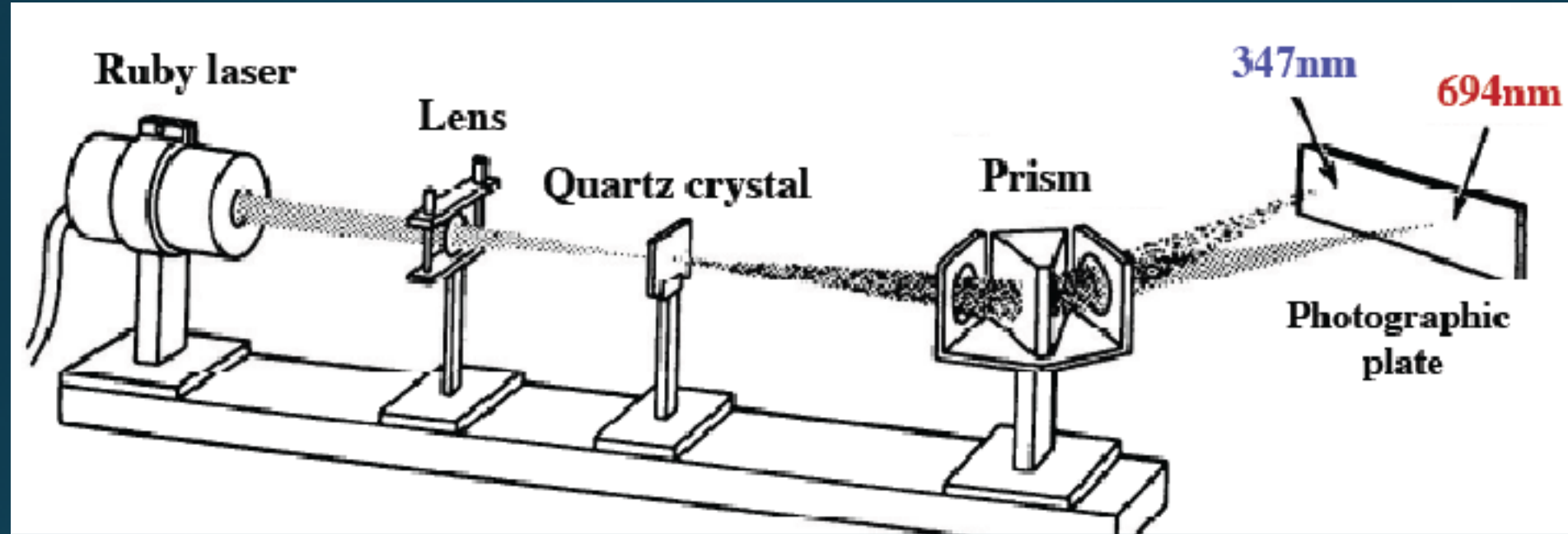
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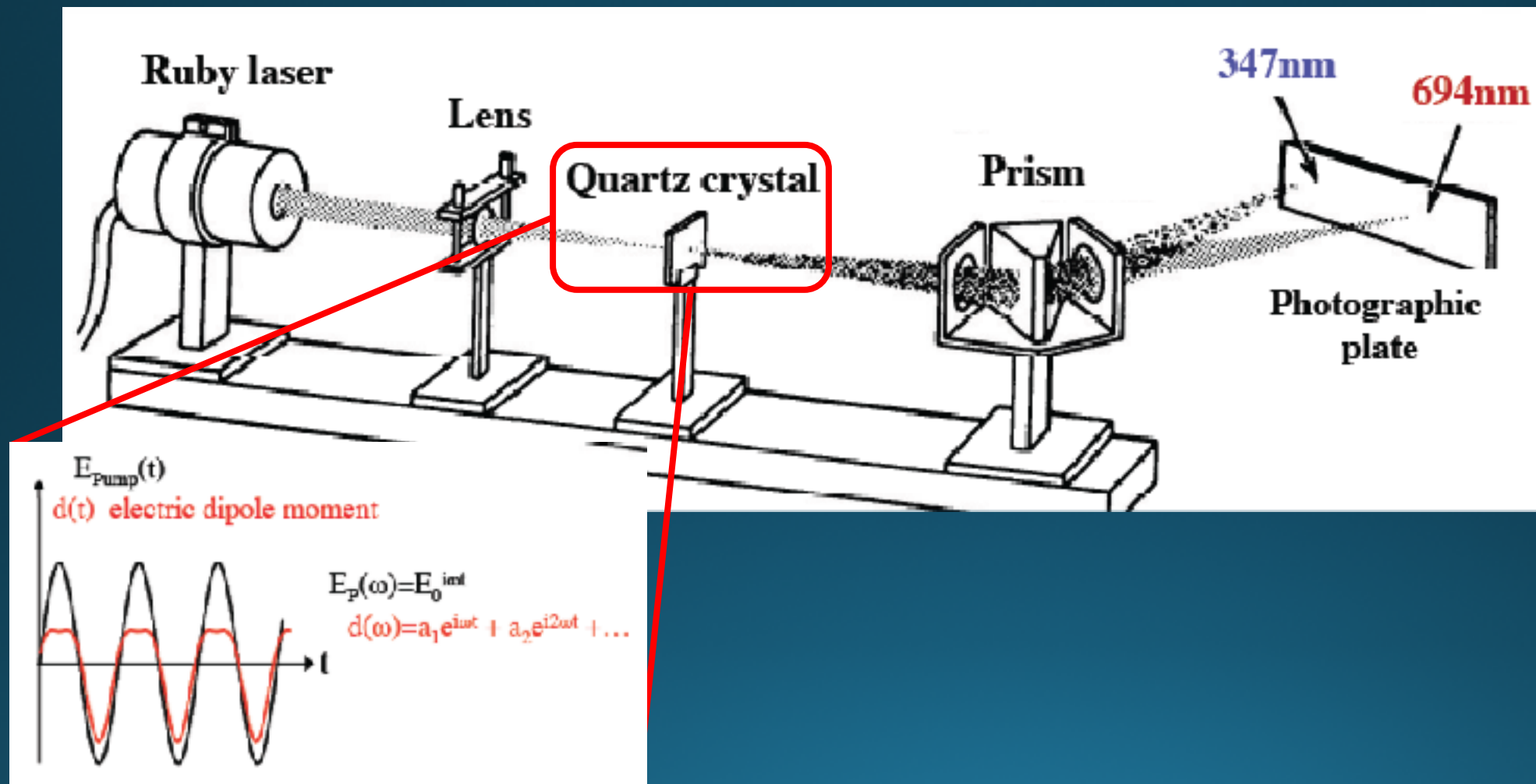


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HHG in the Beginning: A Barely Visible "Smudge" Revolutionizes Optical Science

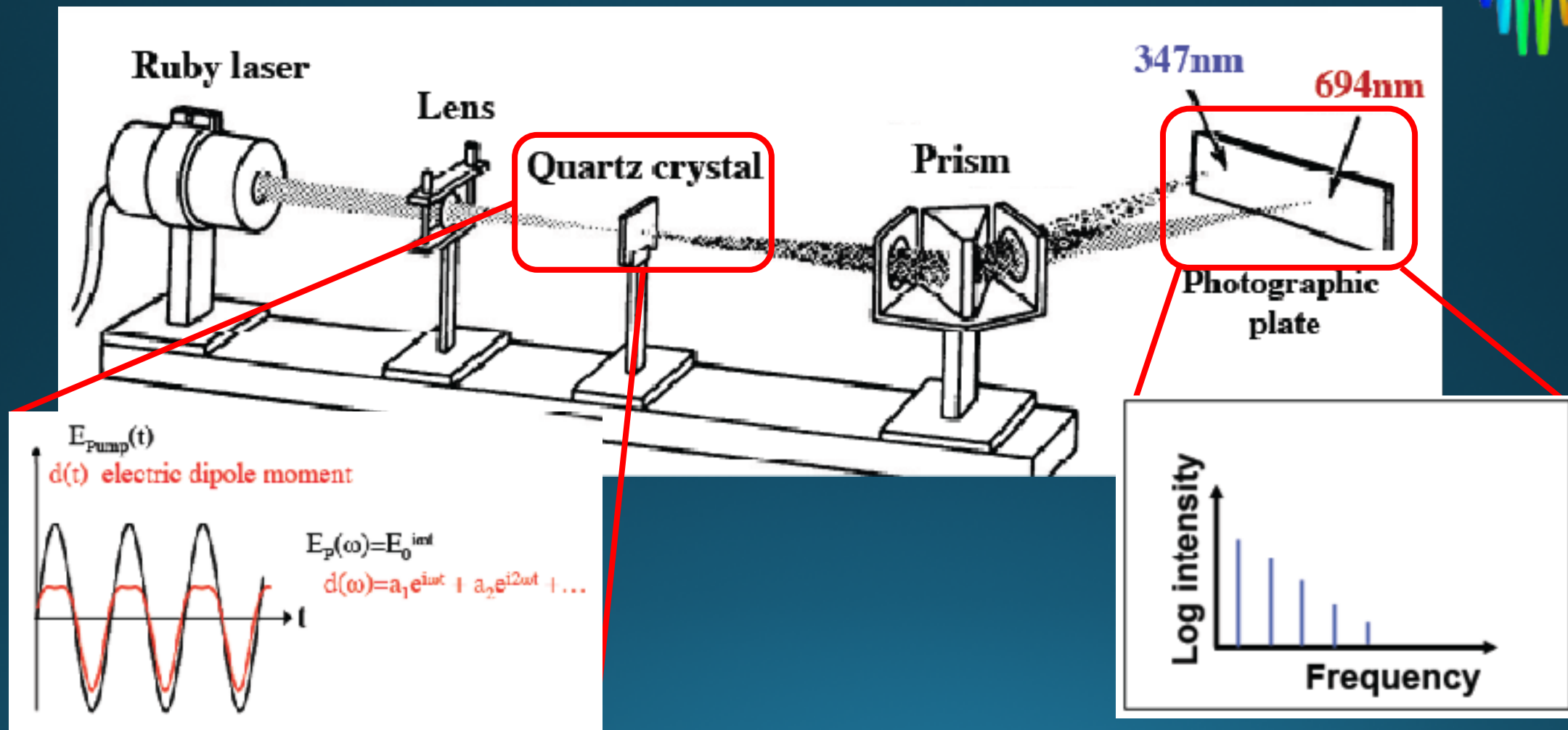


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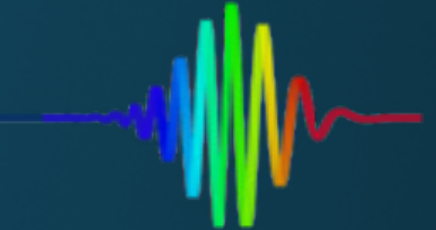
Franken et al. PRL, 7, 1961

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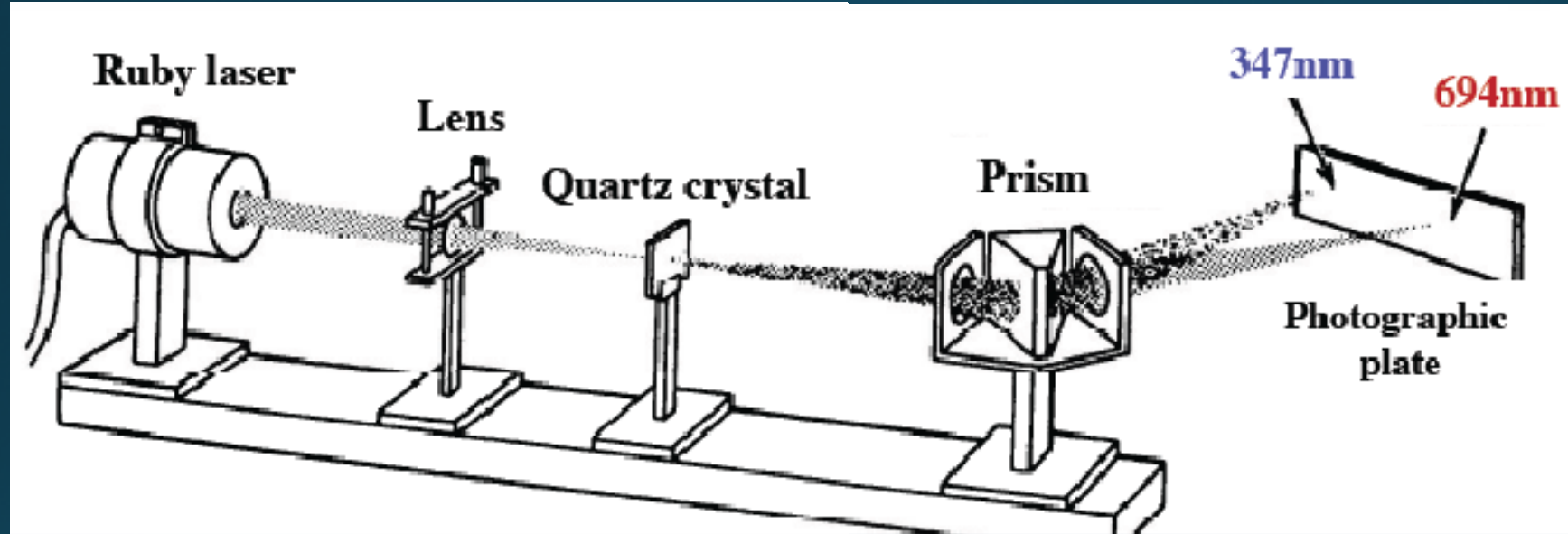


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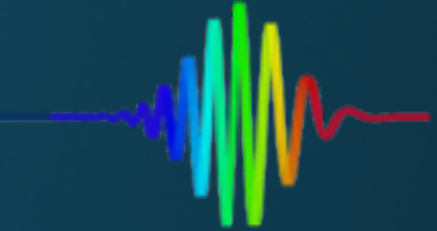
Evolution of HHG: Perturbative Optics to Extreme Nonlinear Optical Science



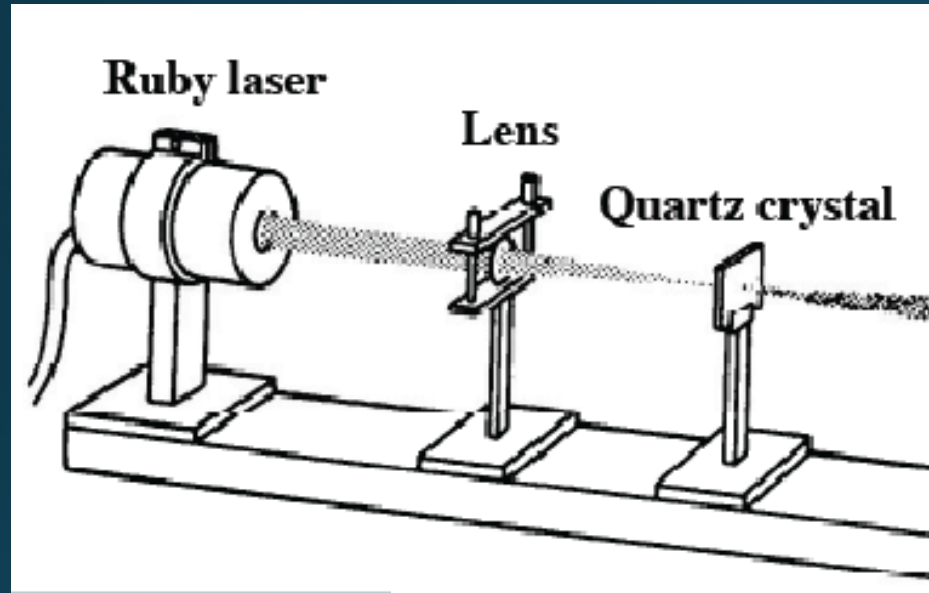
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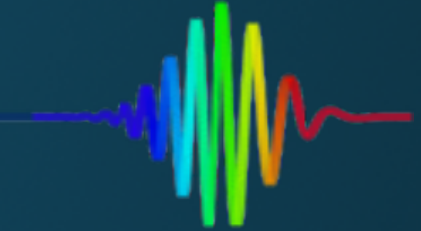
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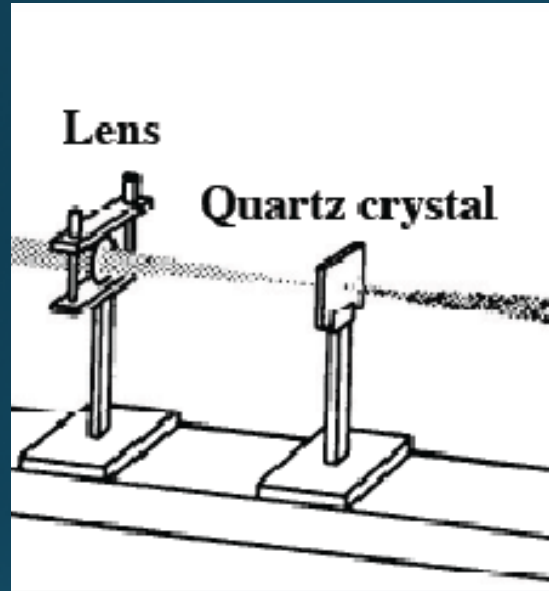
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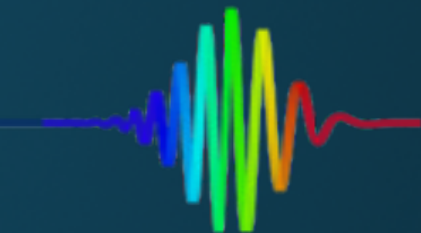
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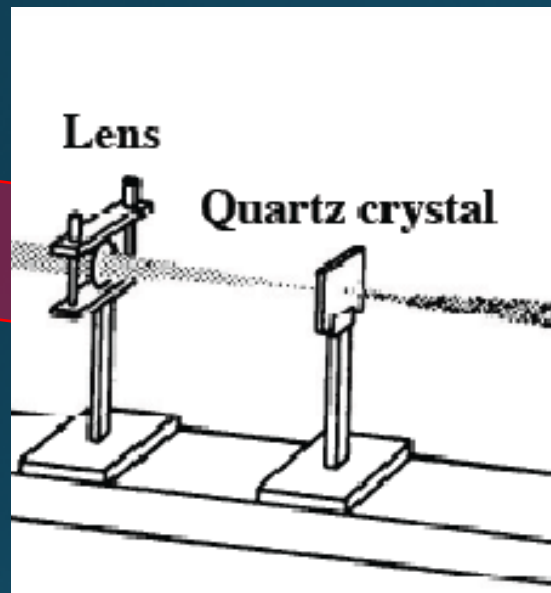
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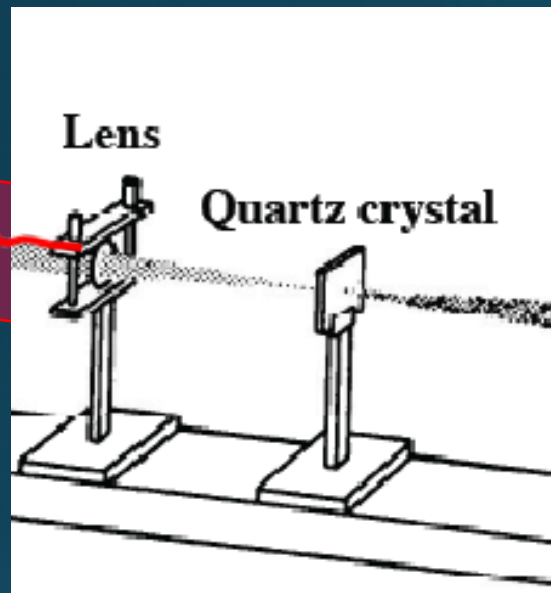


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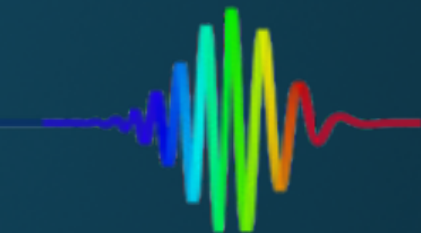


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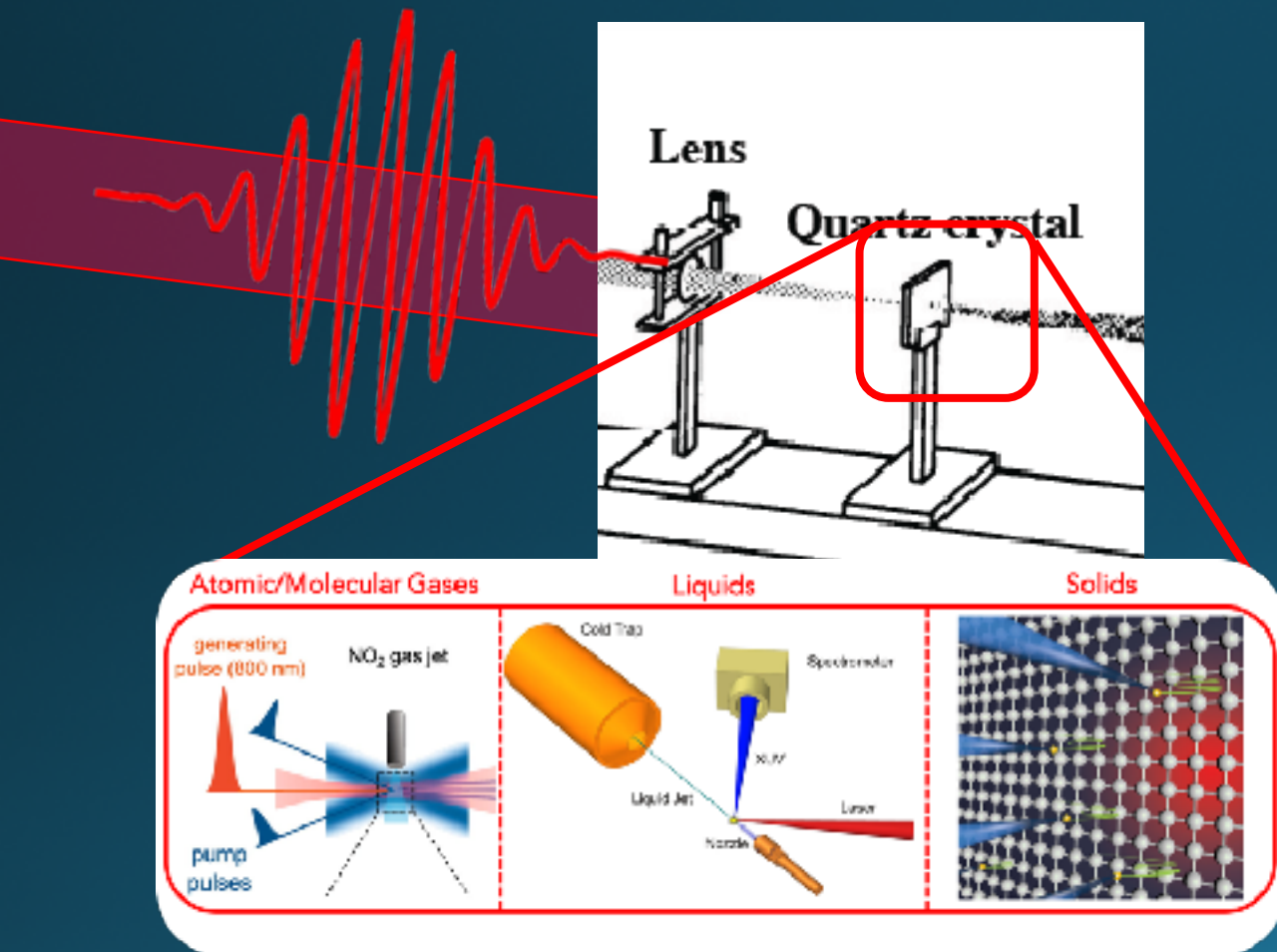
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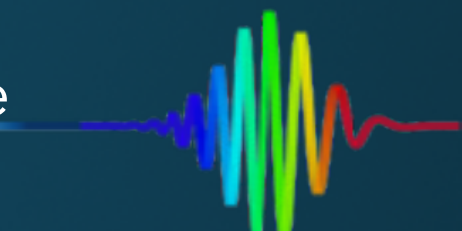
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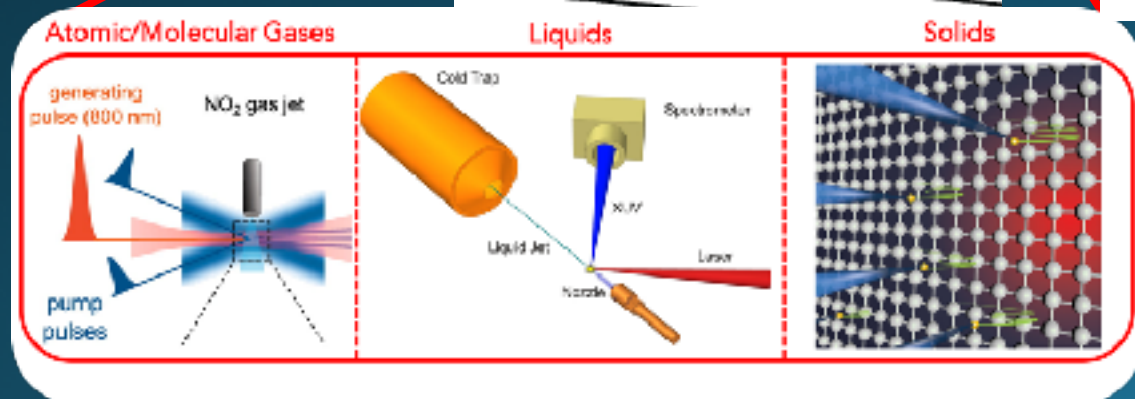
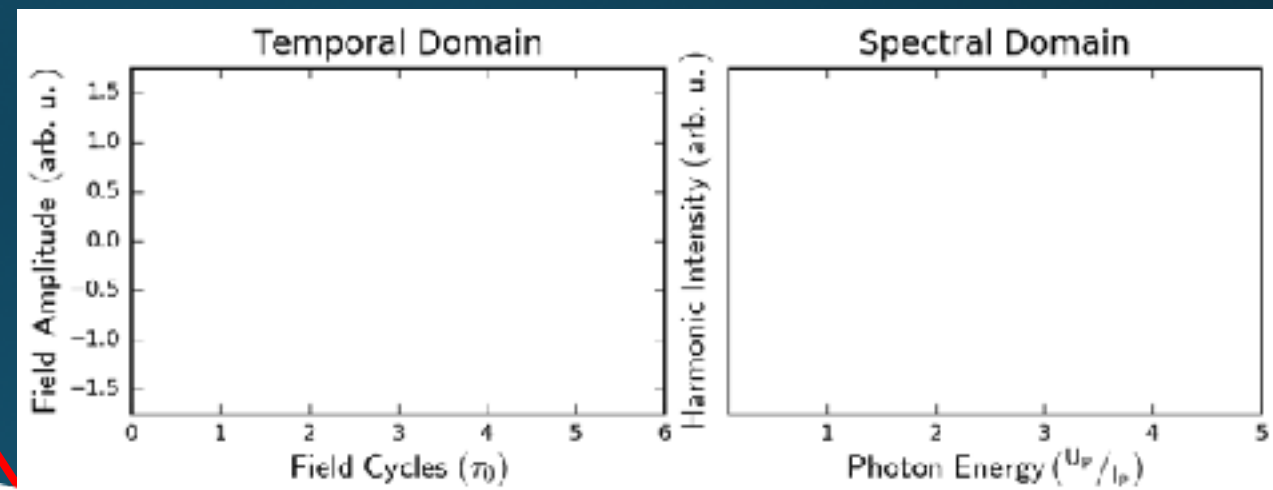
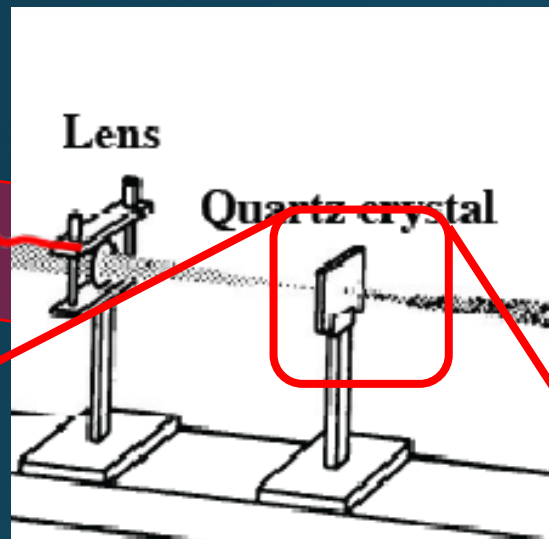
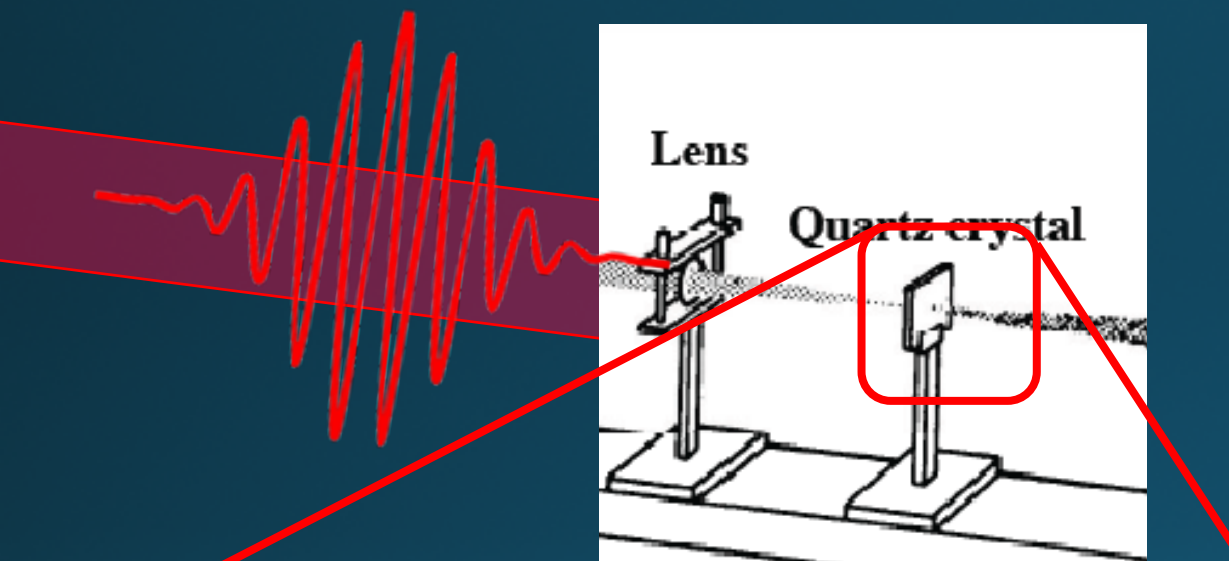
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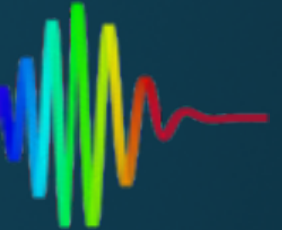
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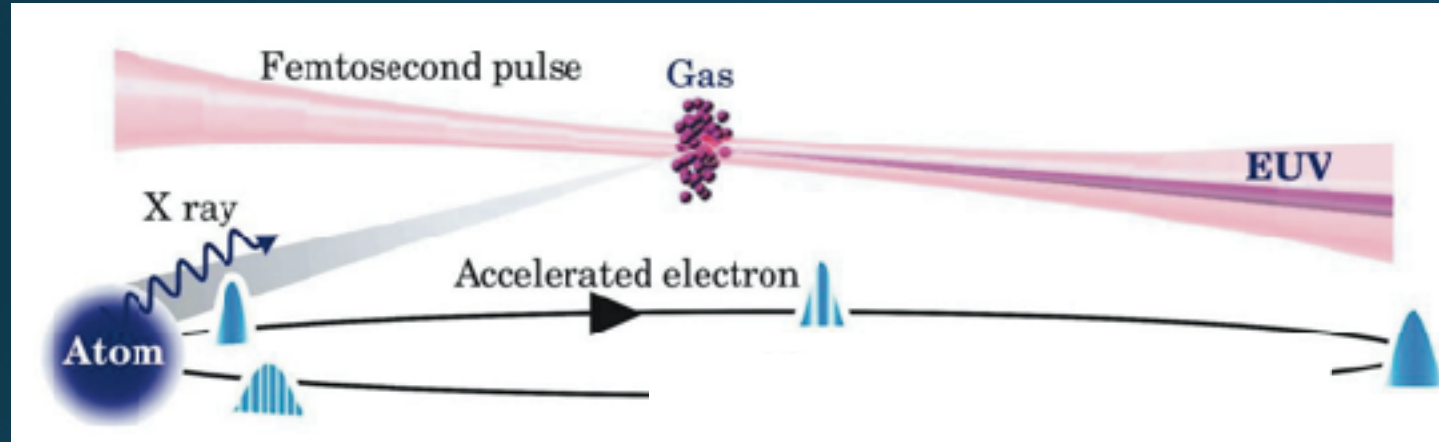
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Microscopic Mechanism of HHG: Epitome of Classical Correspondence Principle



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



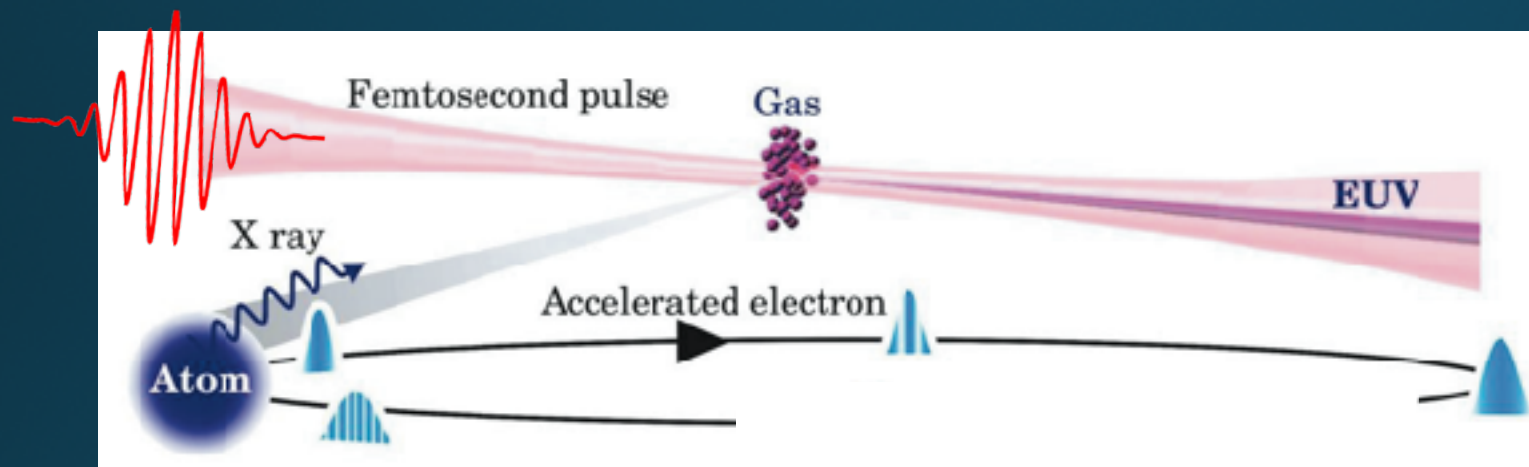
Kuchiev, JETP, **45**. 404 (1987)

Classical: Corkum. PRL 1993

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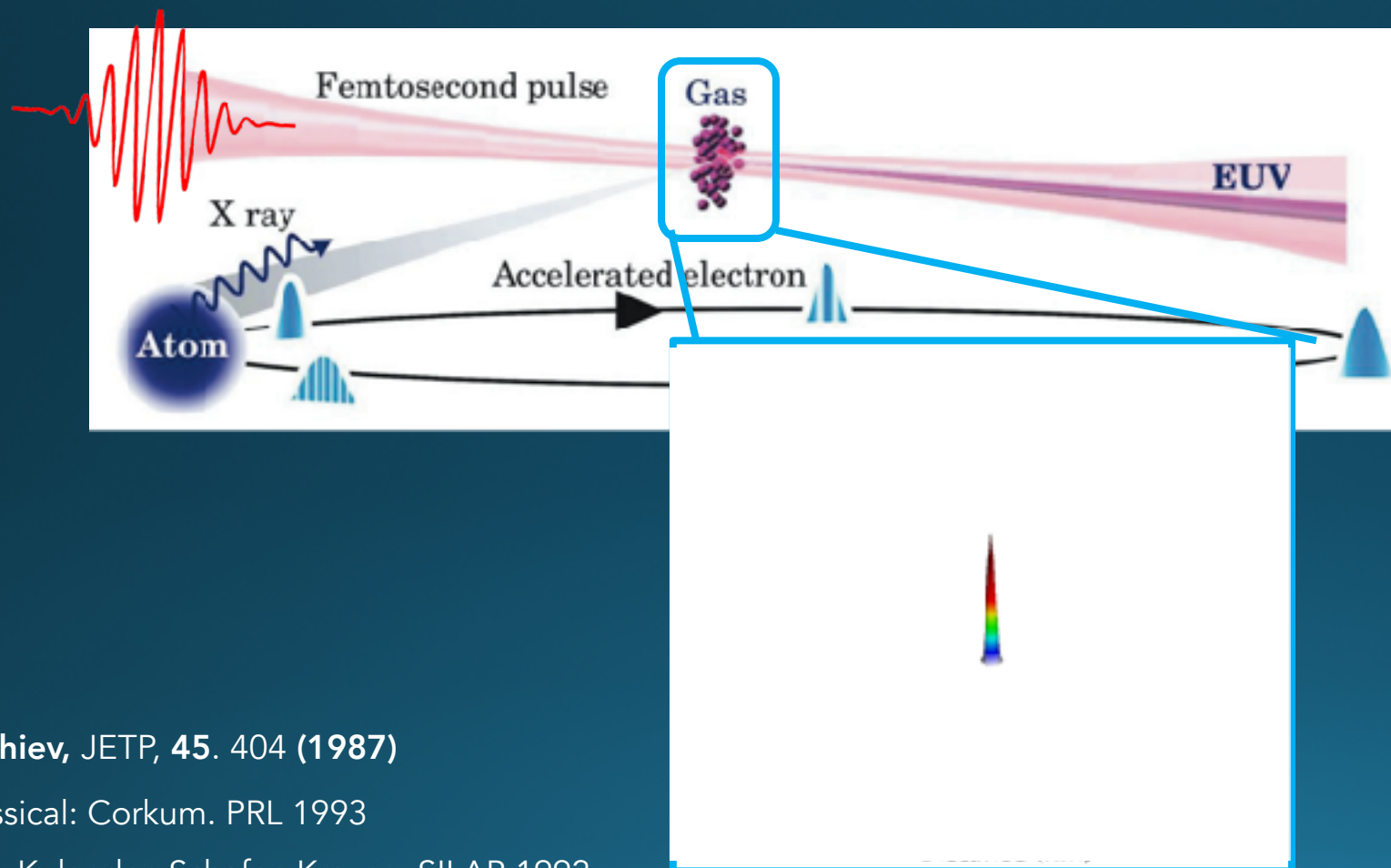
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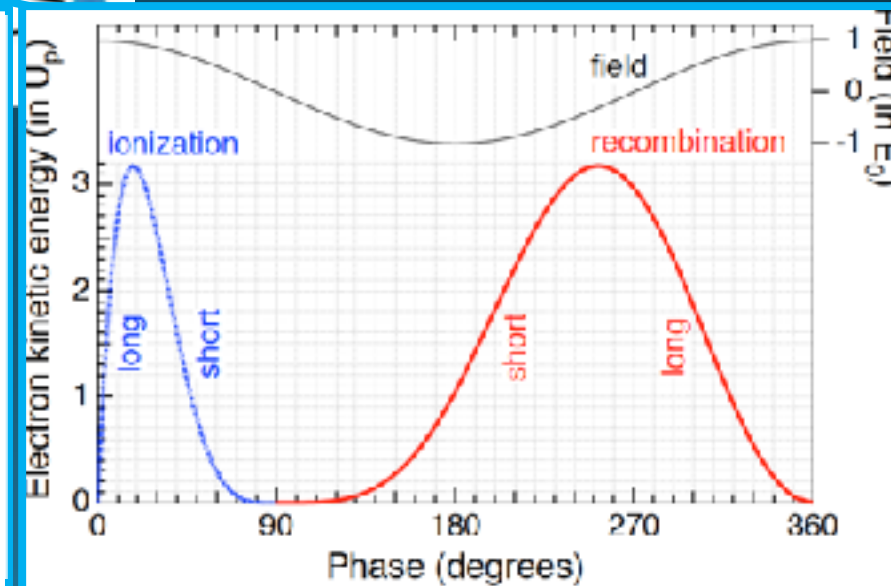
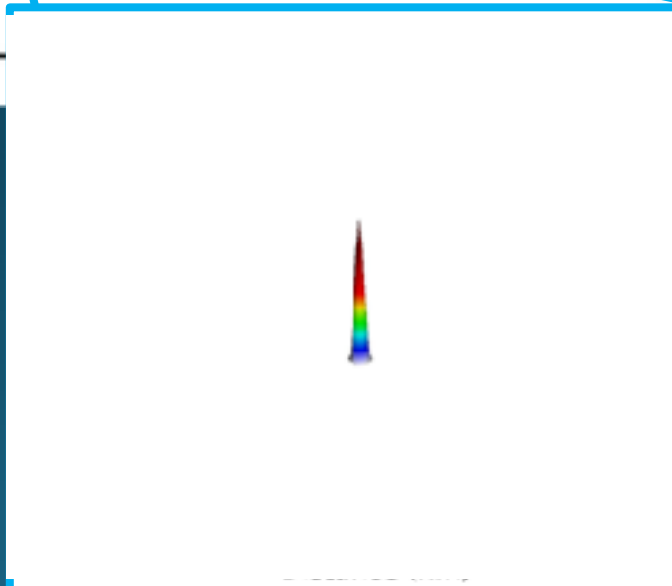
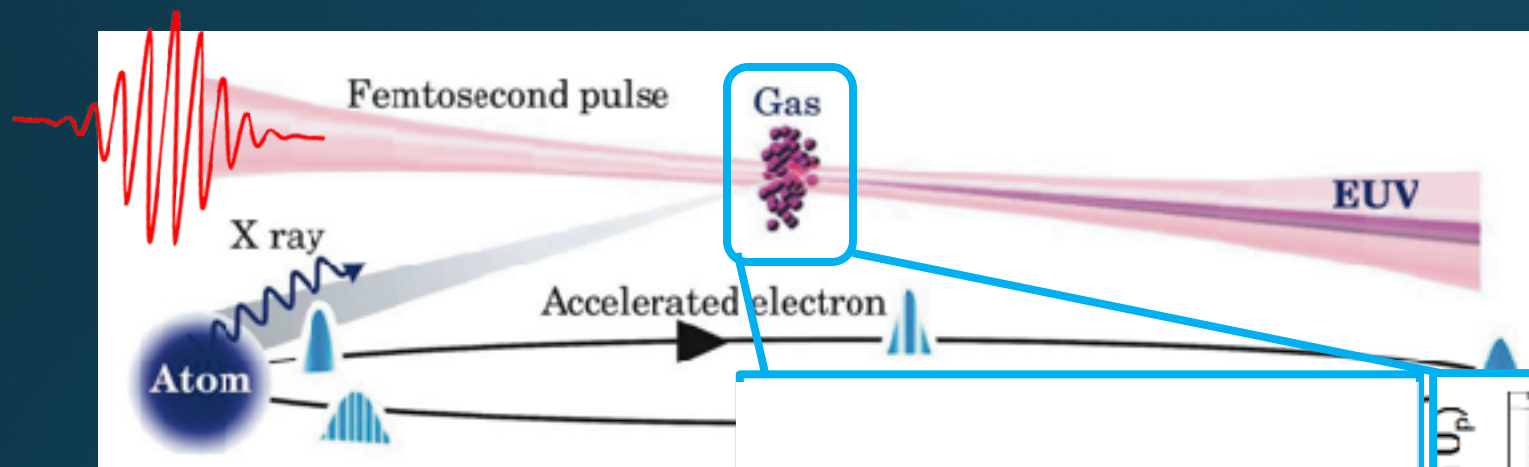
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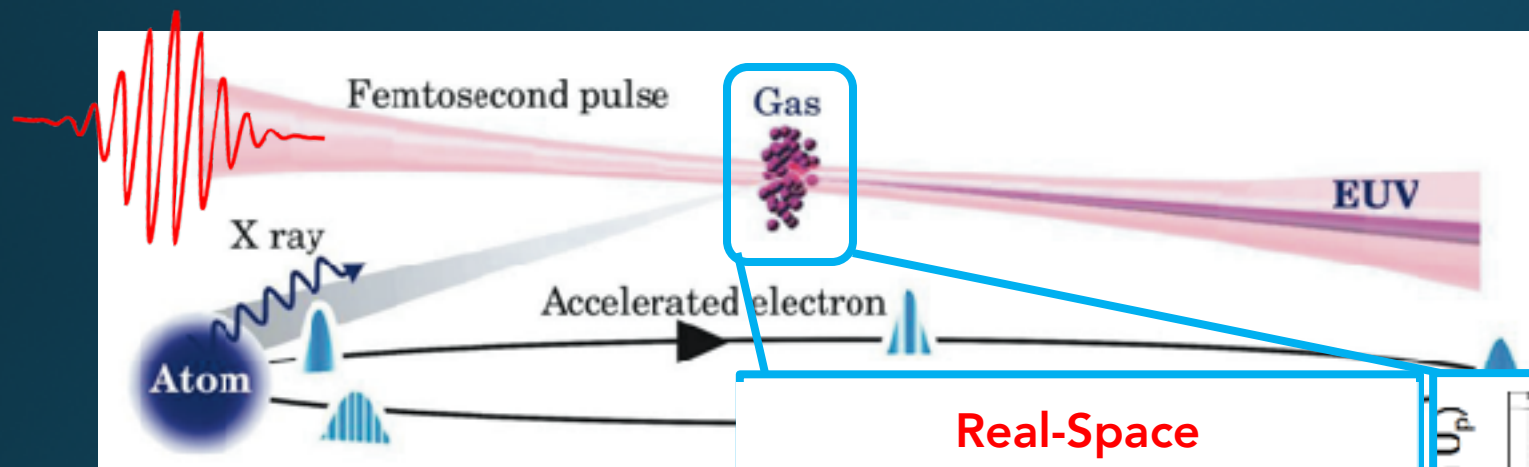
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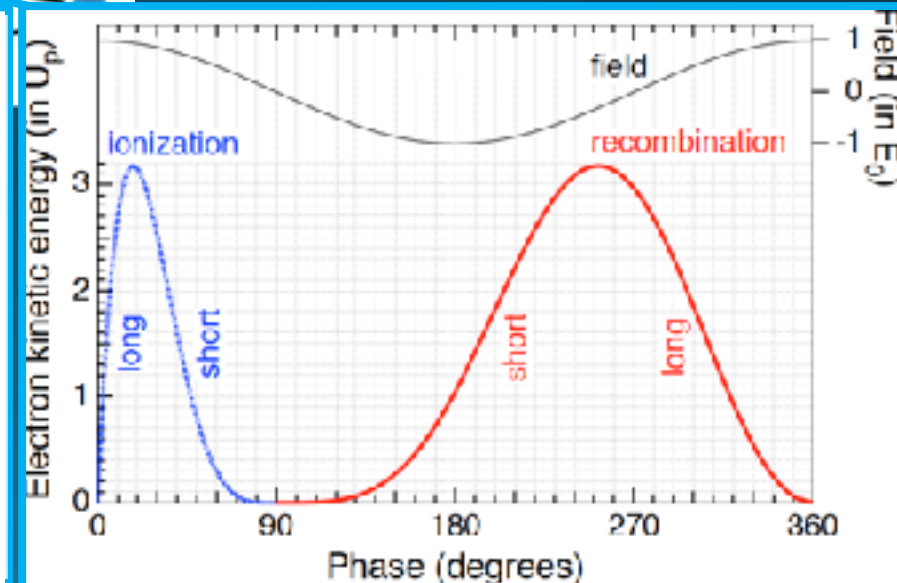
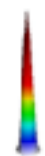
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Real-Space Wavepacket Motion



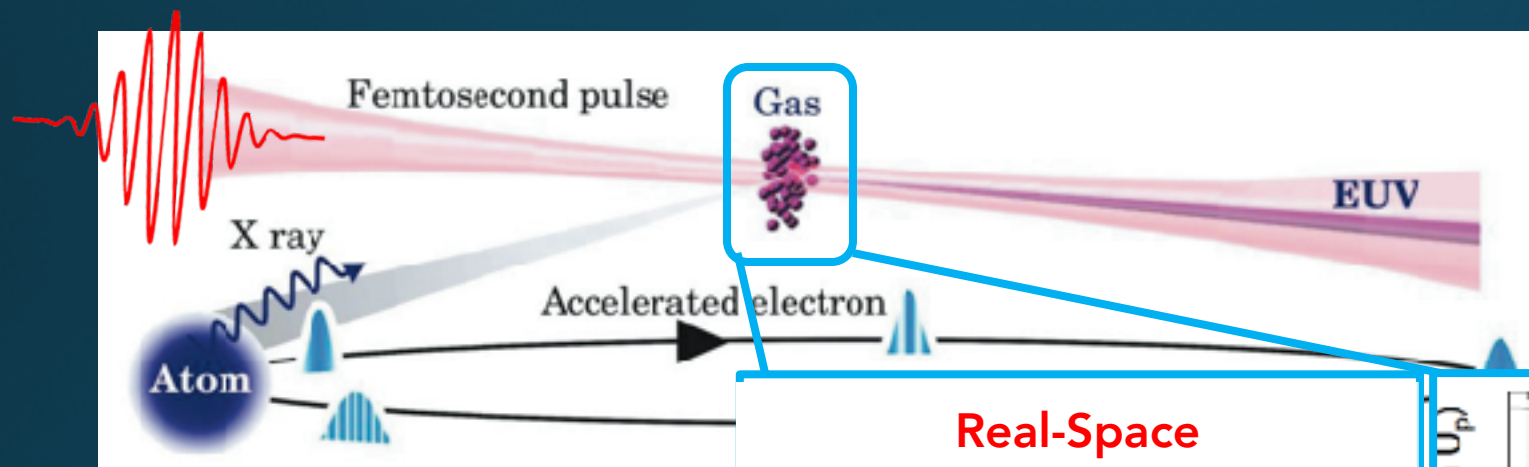
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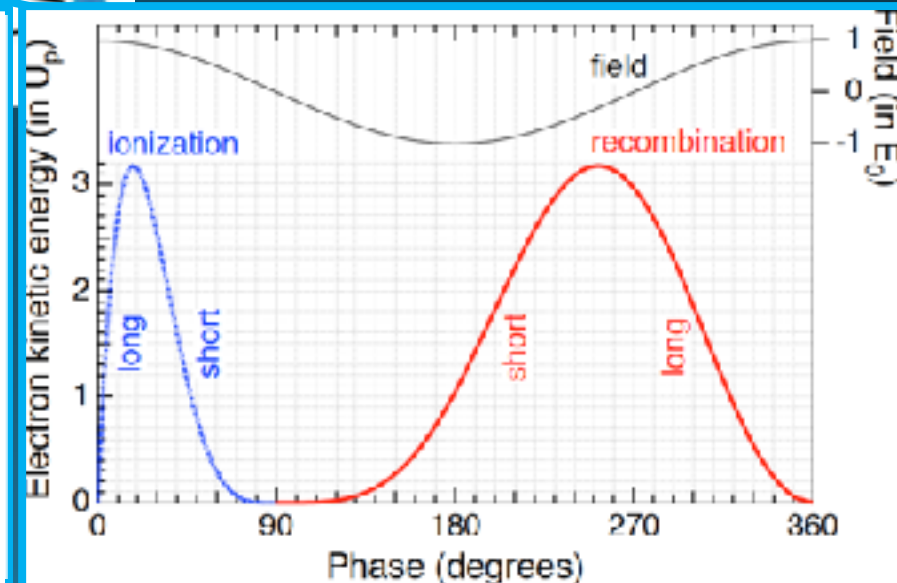
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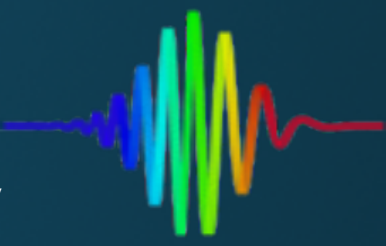
Macroscopic HHG: Attosecond Nonlinear Optics in a K (Nut) Shell



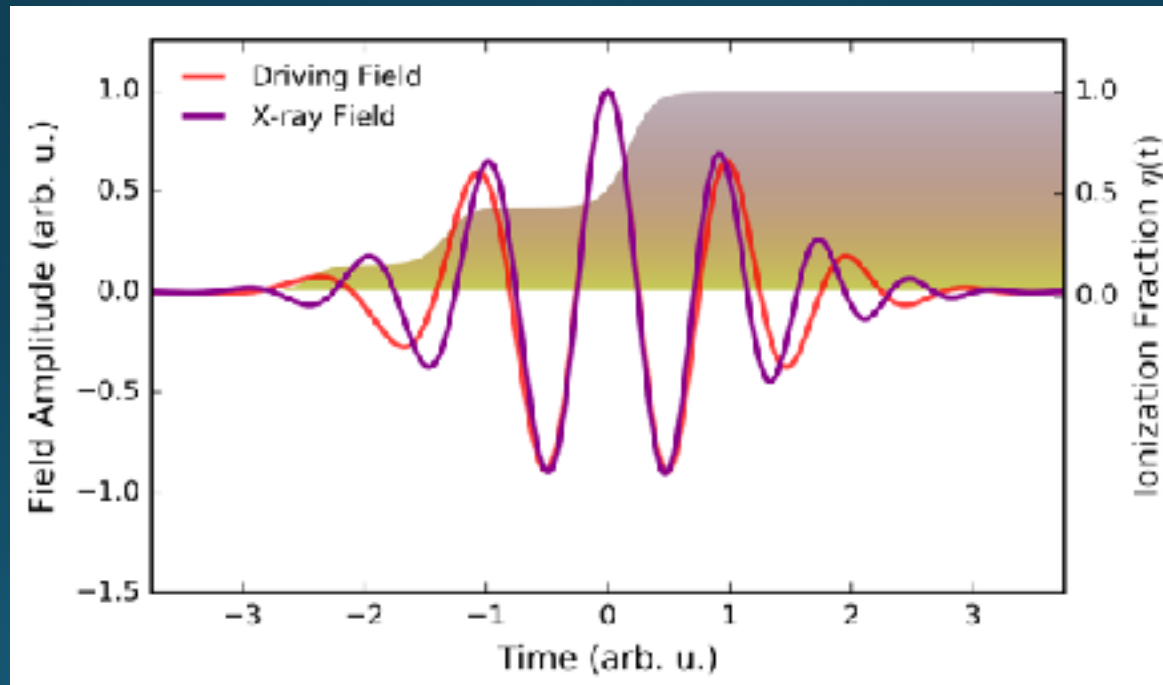
- The generation of bright, coherent beams of X-ray light demands that we solve the currency exchange problem inherent to nonlinear optics.
- Single atom yield $\sim \lambda^{-6.5}$

Rundquist, Science, 5368, 1998
Popmintchev, PNAS, 106, 2009
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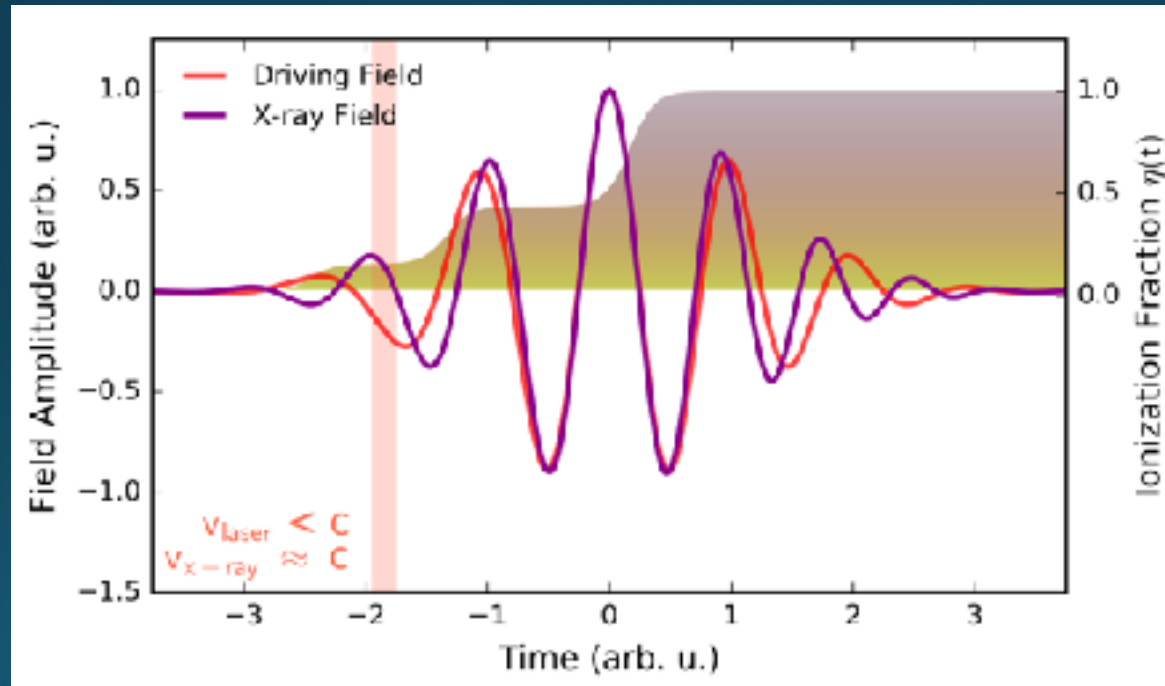
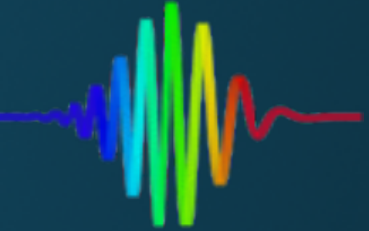
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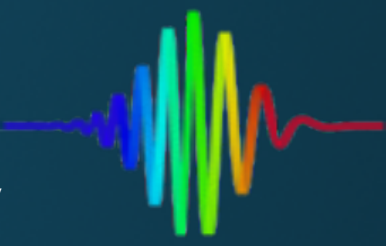
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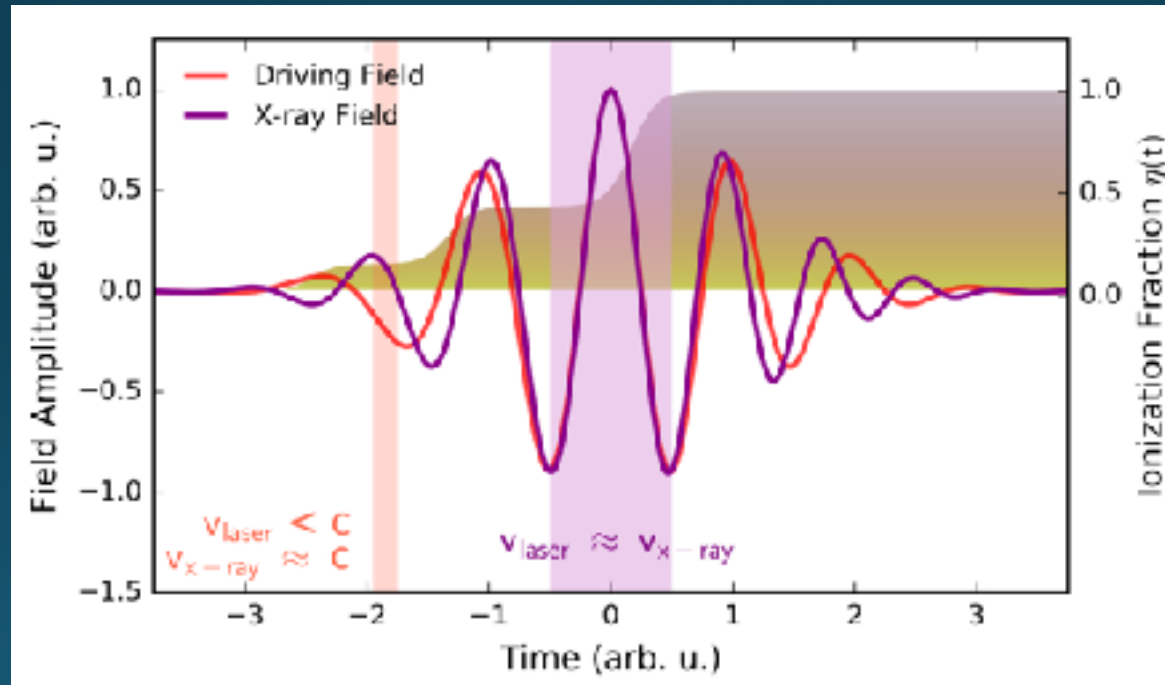


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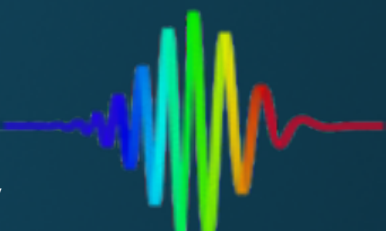


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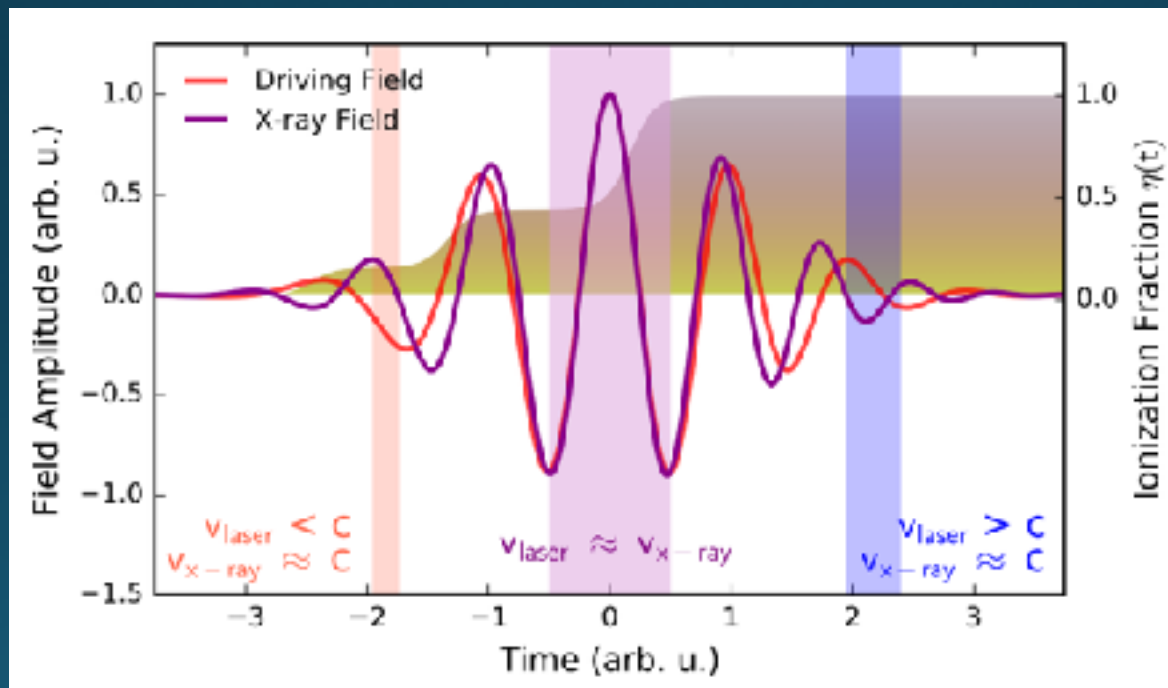


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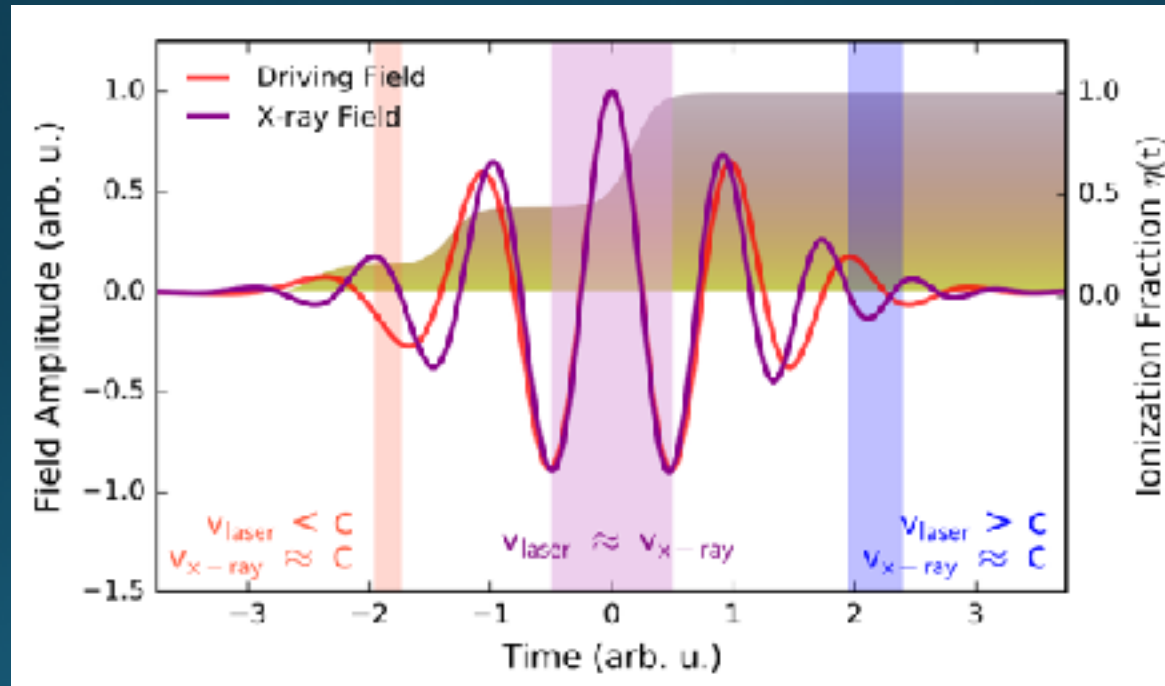
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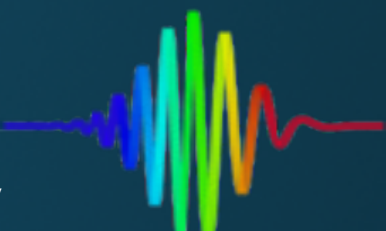
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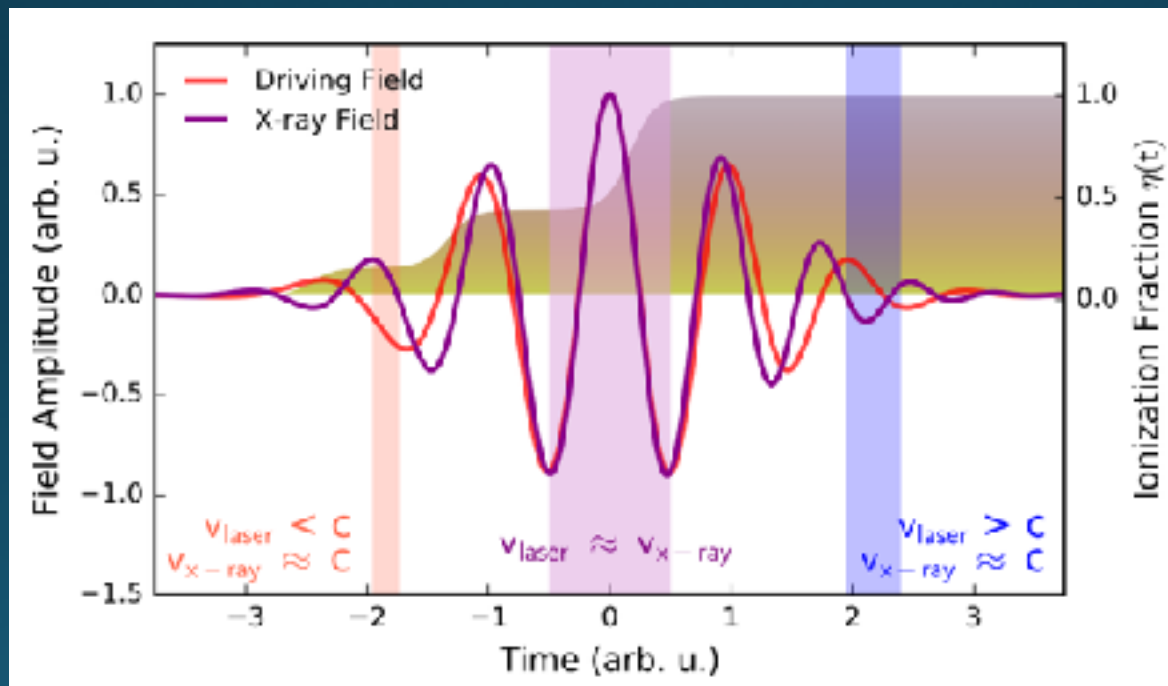


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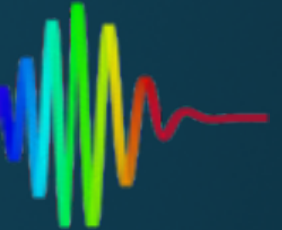


- The generation of bright, coherent beams of X-ray light demands that we solve the currency exchange problem inherent to nonlinear optics.
- Single atom yield $\sim \lambda^{-6.5}$



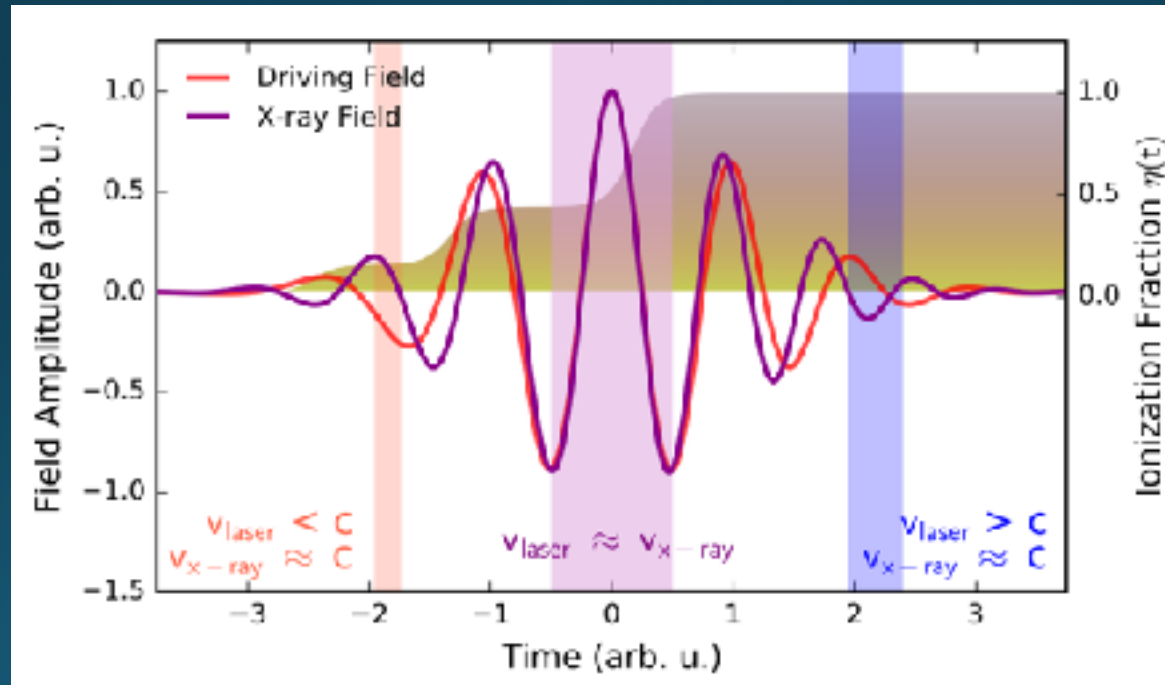
Rundquist, Science, 5368, 1998
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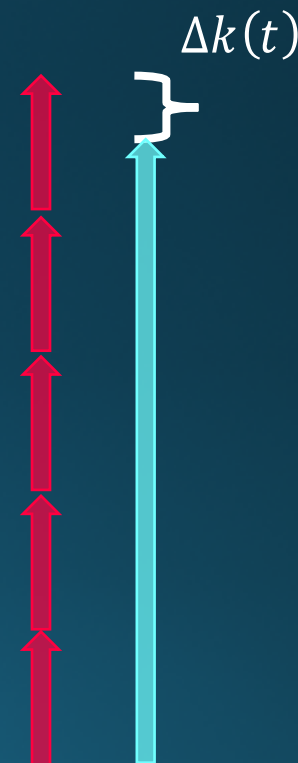
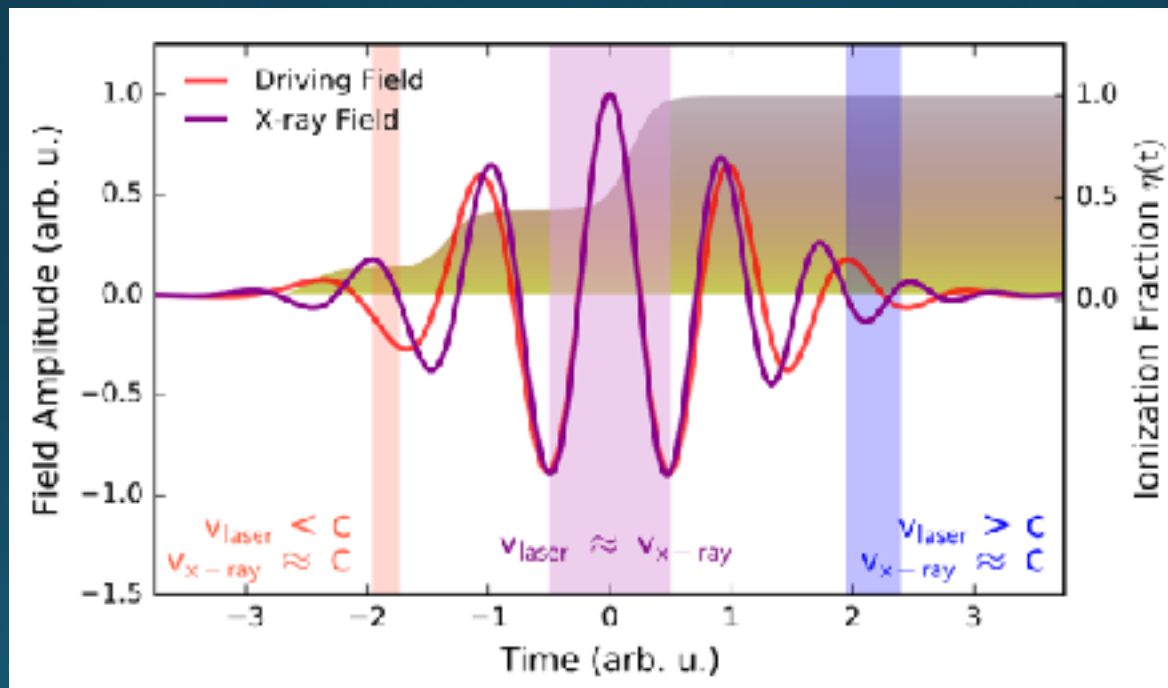
- Single atom yield $\sim \lambda^{-6.5}$
$$\Delta k(t) = -qP \left\{ [1 - \eta(t)] \frac{2\pi}{\lambda_L} \delta n - \eta(t) N_{atm} r_e \lambda_L \right\} + \Delta k_{geom} + \Delta k_{quantum}(t)$$



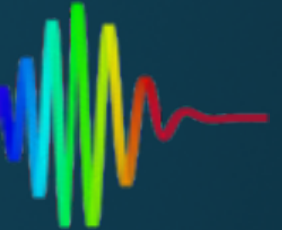
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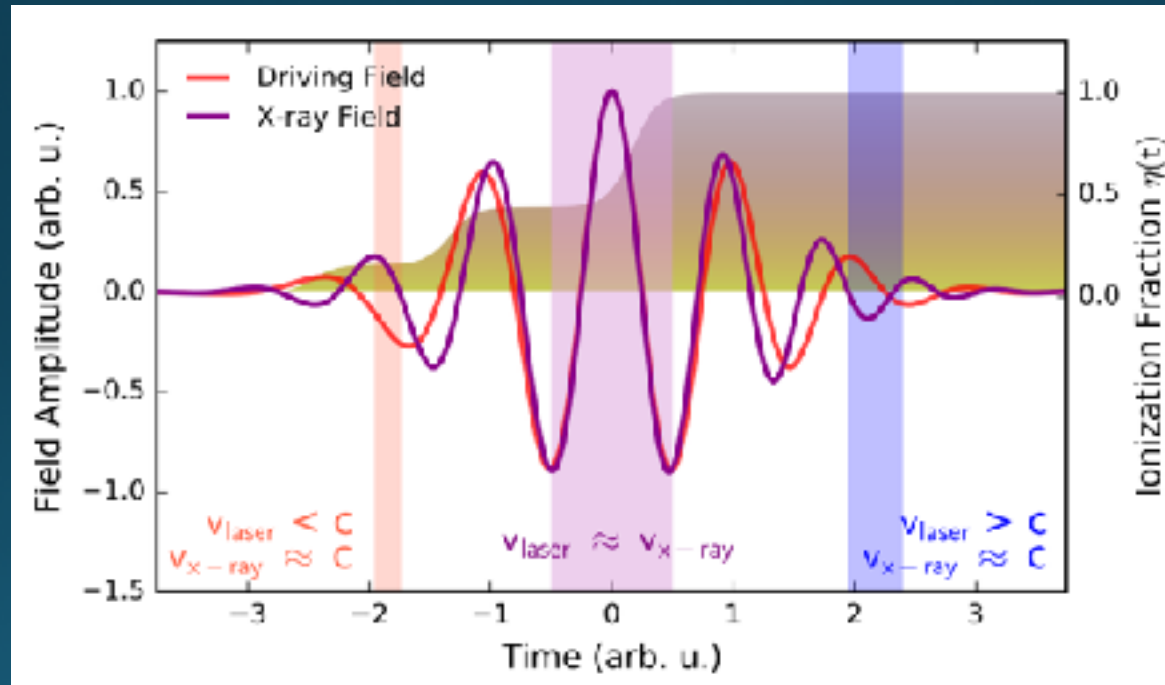


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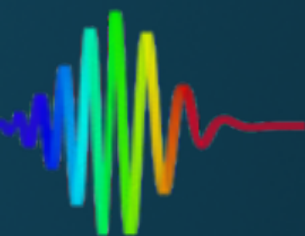


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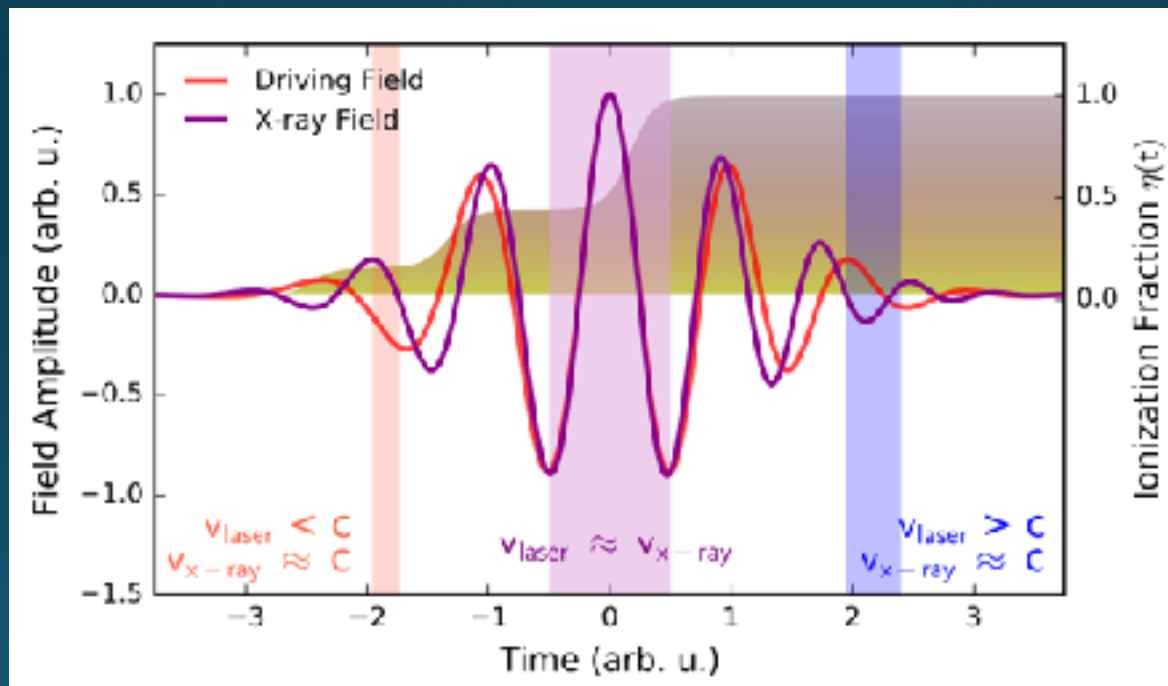


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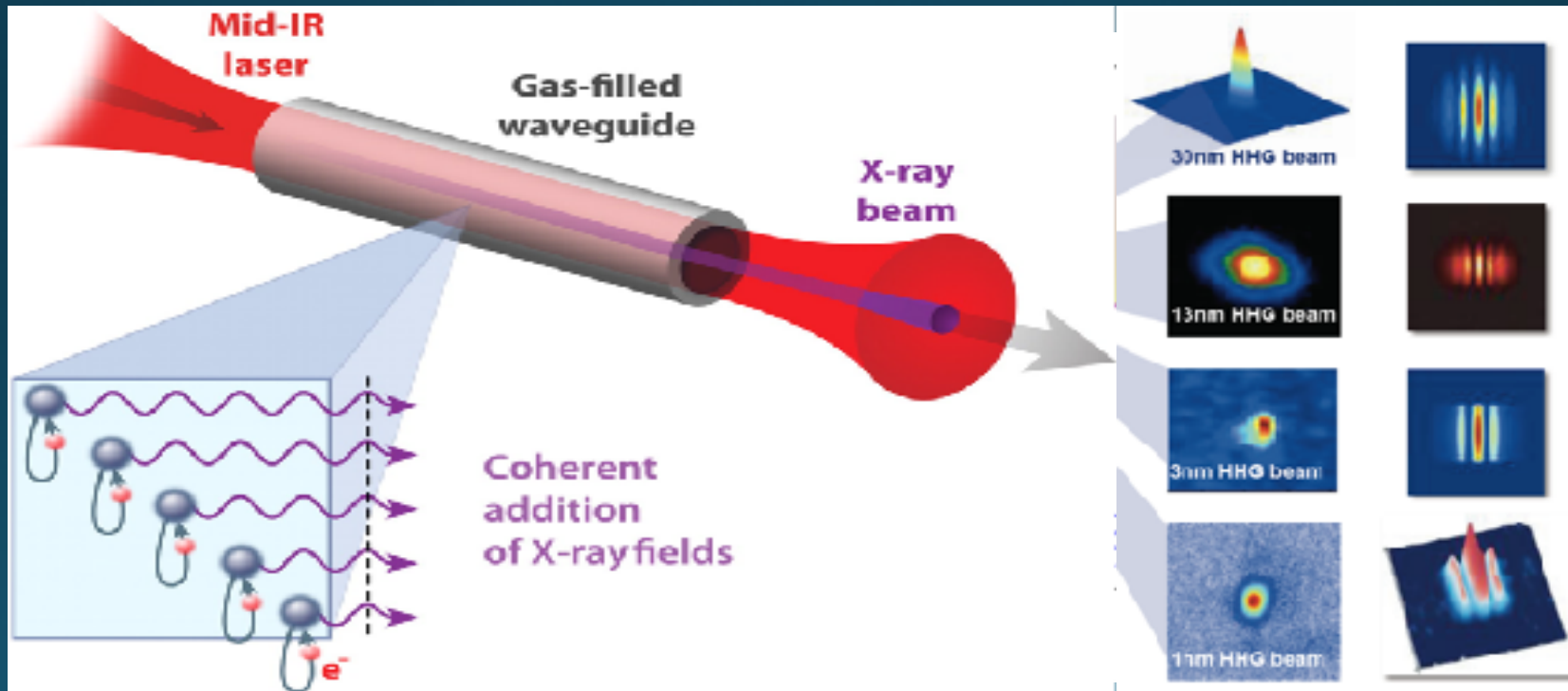
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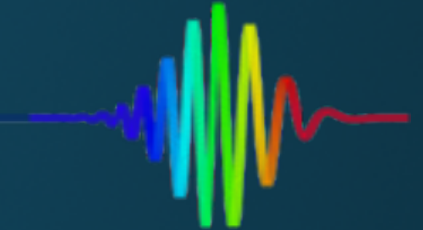
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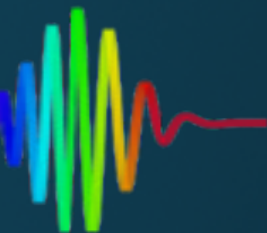


Custom Tailored EUV and X-ray Light From a Table-Top: Exquisite Control Over the Entire Up-Conversion Process

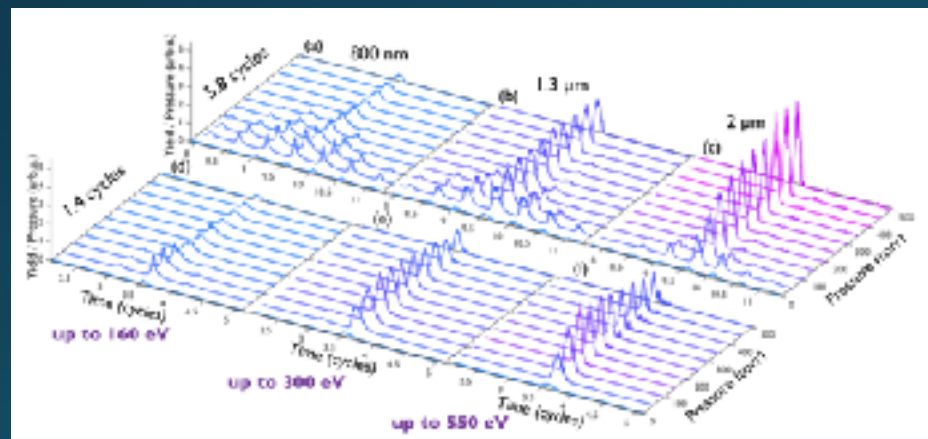
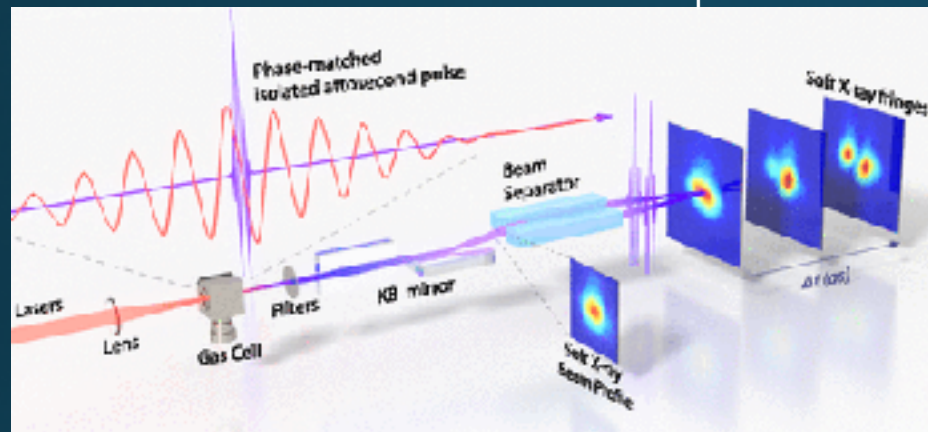


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Phase-matched isolated as pulses



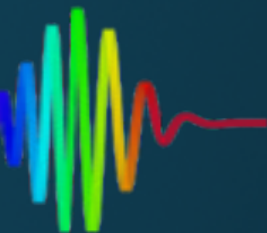
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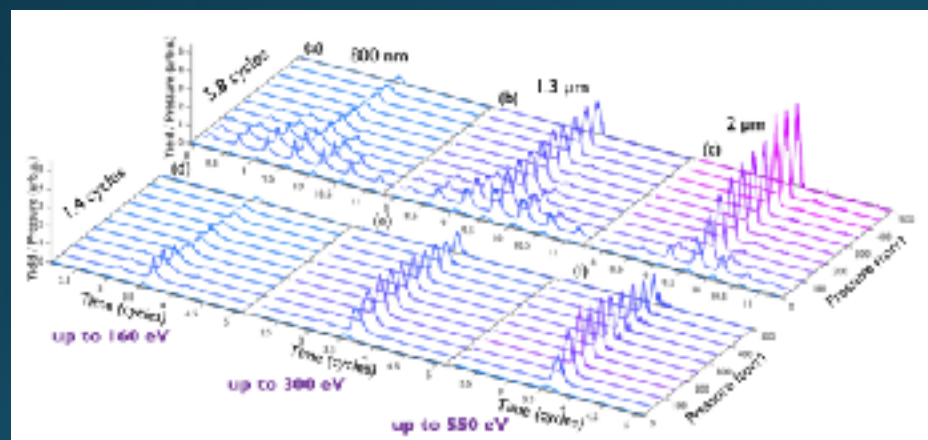
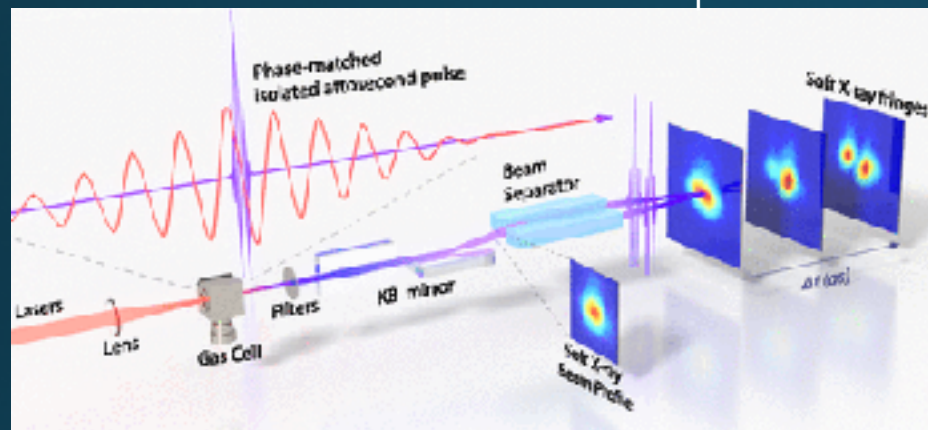
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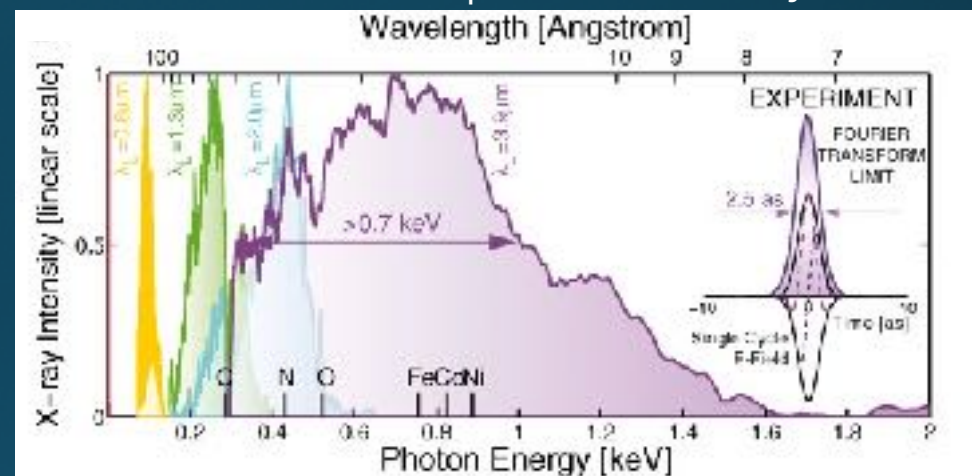
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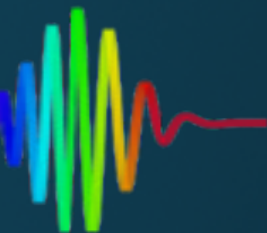


Coherent, zeptosecond x-rays

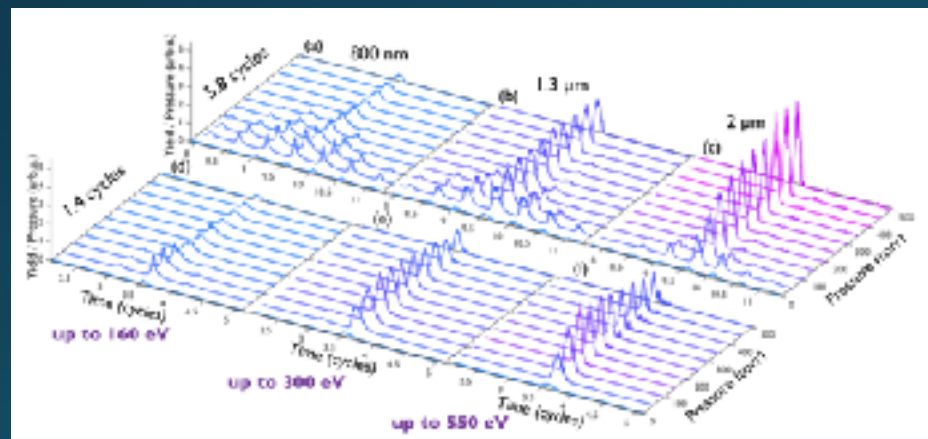
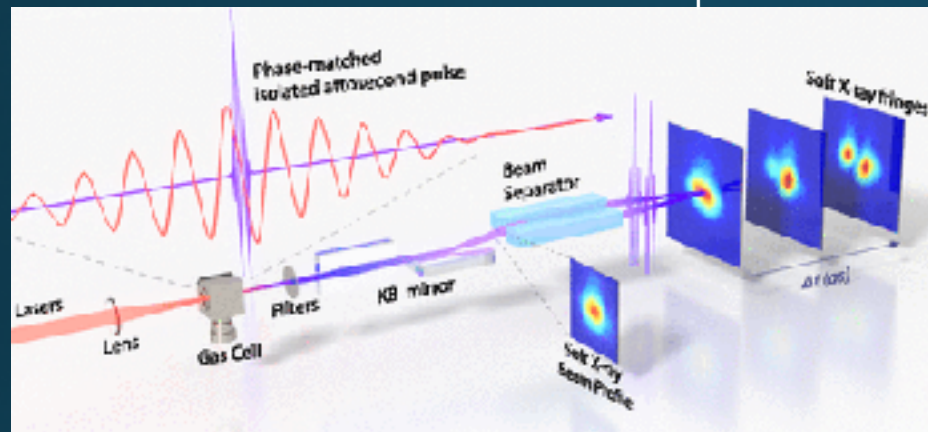


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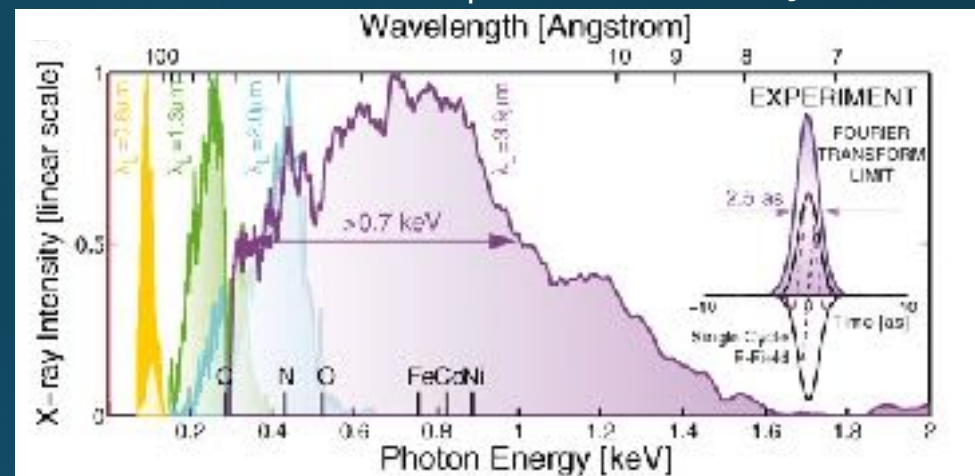
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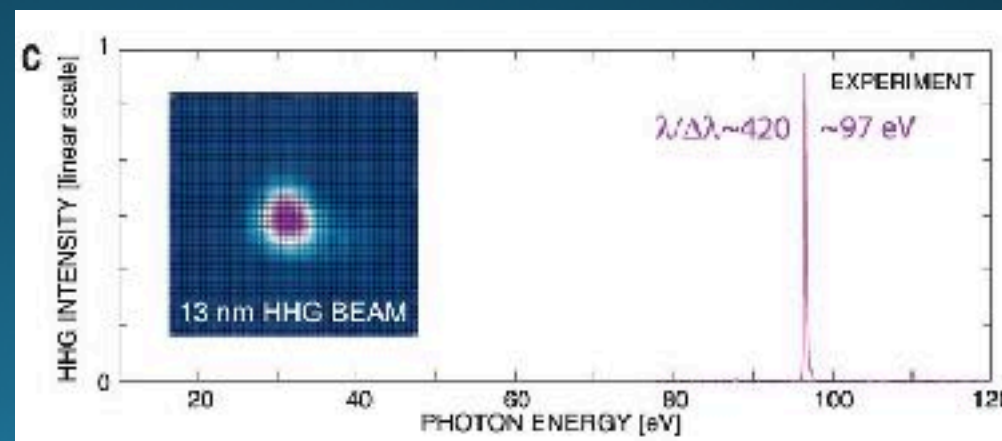
Phase-matched isolated as pulses



Coherent, zeptosecond x-rays



Bright, isolated harmonics



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Hernandez-Garcia, Opt Exp, 106, 2017

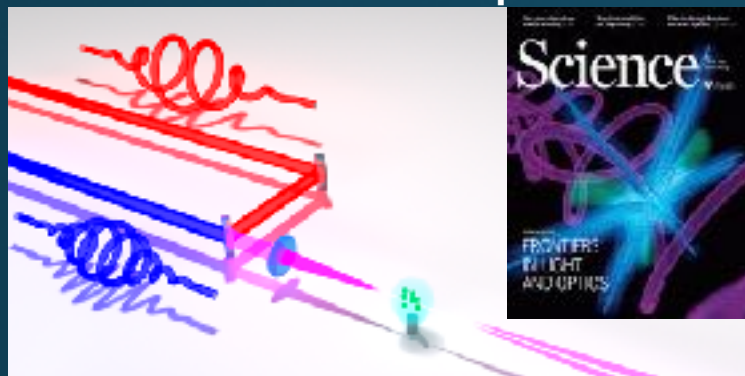
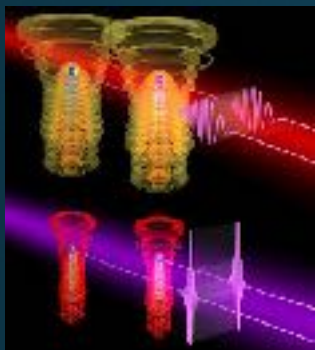
Popmintchev, Science, 6086, 2012

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

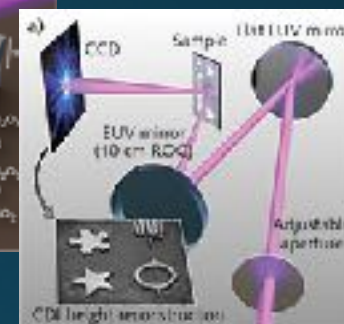
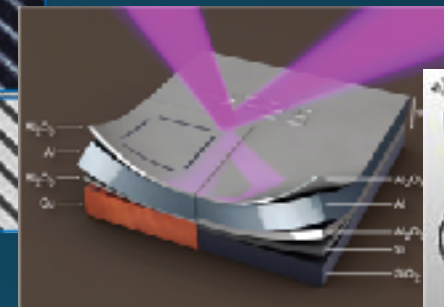


Attosecond Extreme Nonlinear Optics



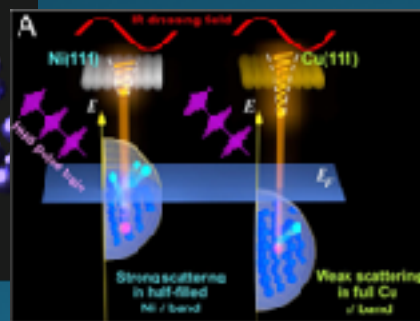
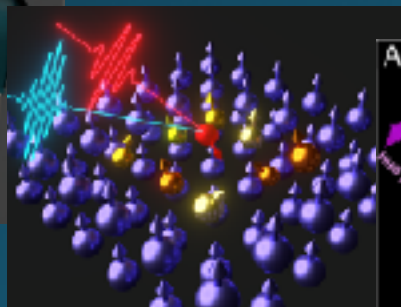
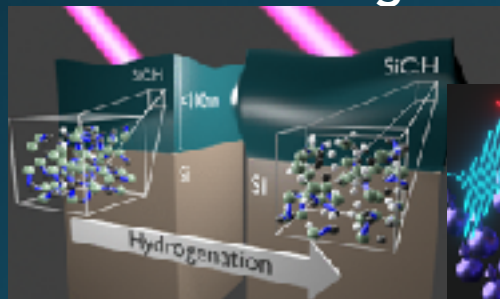
Popmintchev, Science 6265, 2015
Dorney, PRL 118, 2017

Coherent X-ray Imaging



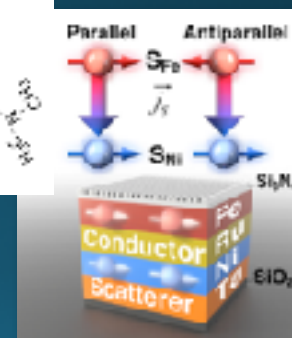
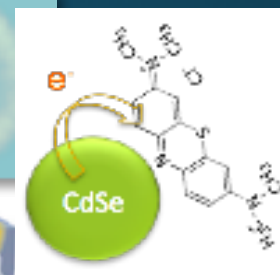
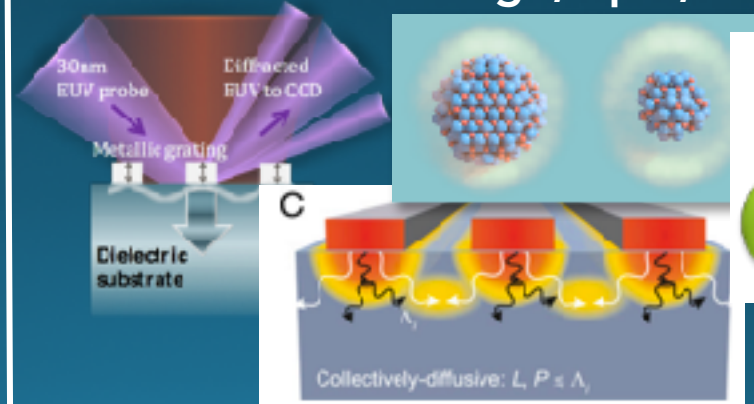
Gardener, Nat. Photon. 11, 2017
Shanblatt, Nano Lett. 16, 2016

Uncovering New Ultrafast Materials Science



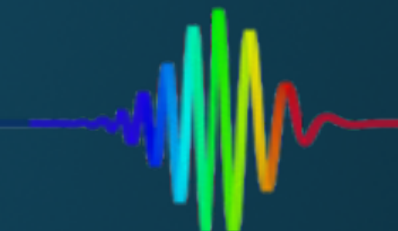
Hernandez-Charapak, Nano Lett. 2017
Tao, Science 353, 2016
Chen, PNAS 114, 2017

Nanoscale Charge, Spin, Energy Transport



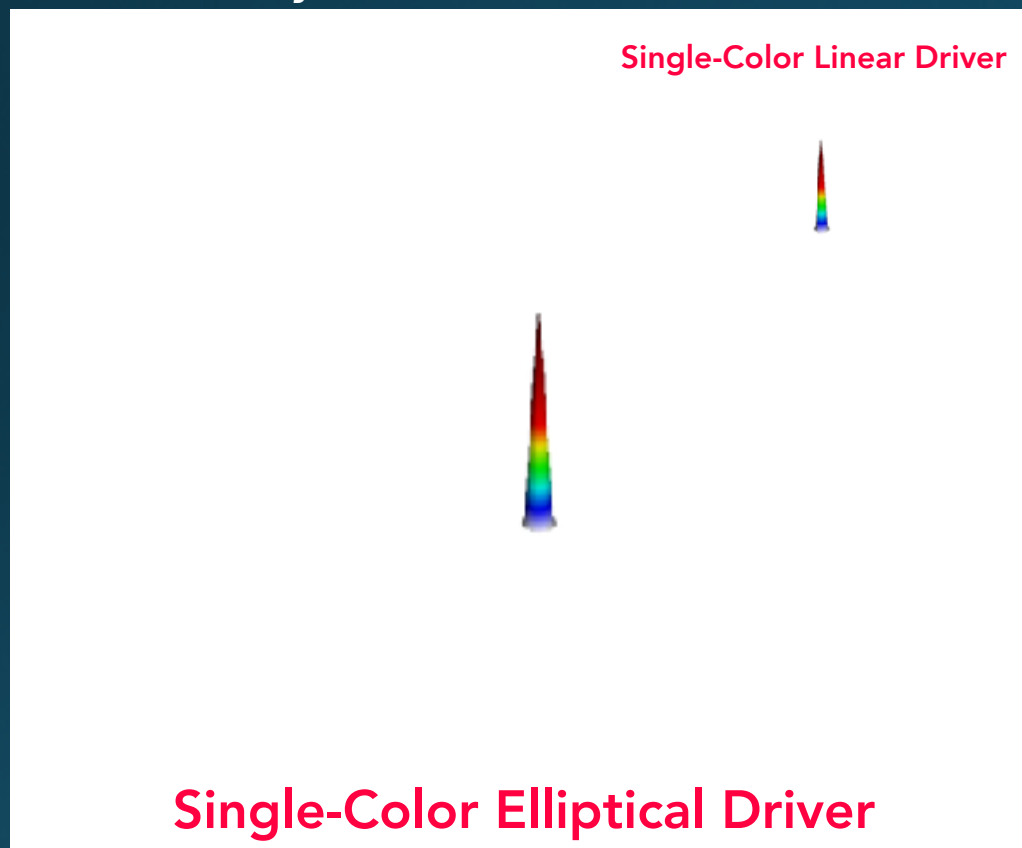
Hoogeboom-Pot, PNAS 112, 2015
Ellis, JACS, 11 2015

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



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

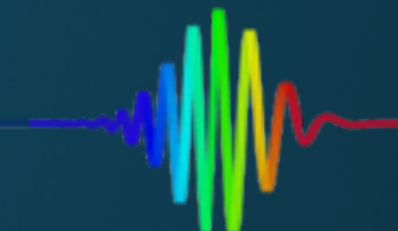
Video: Courtesy of Carlos Hernandez-Garcia



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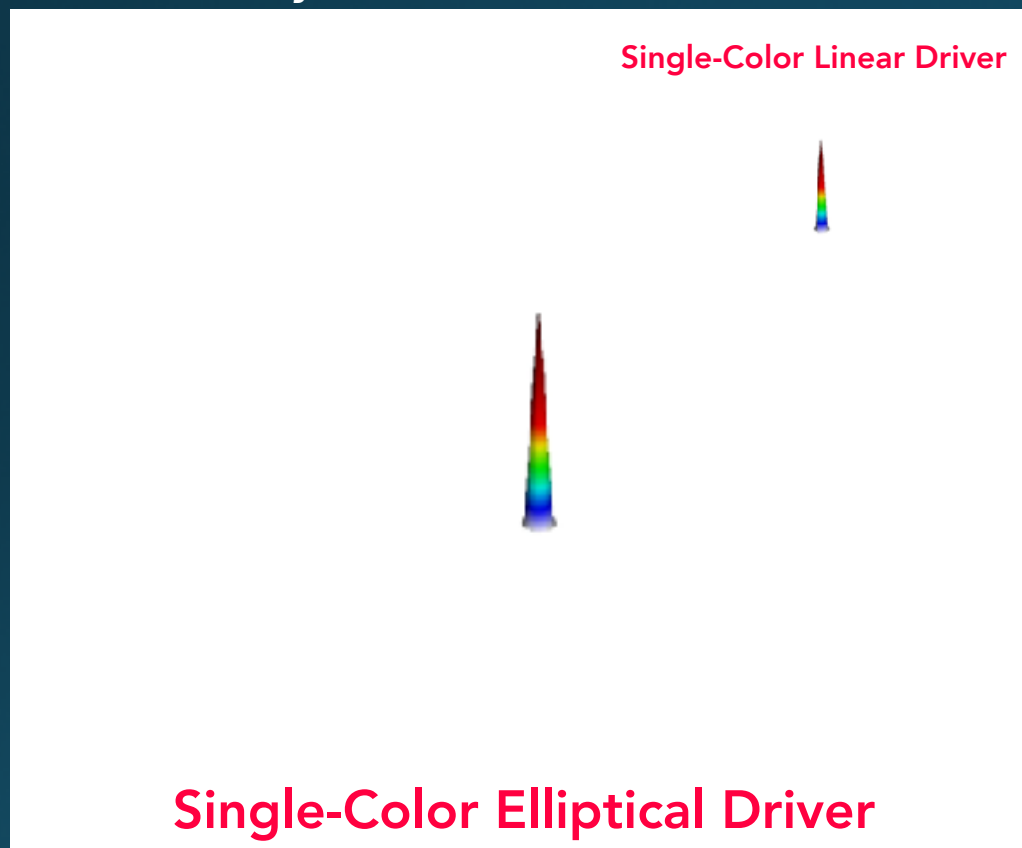
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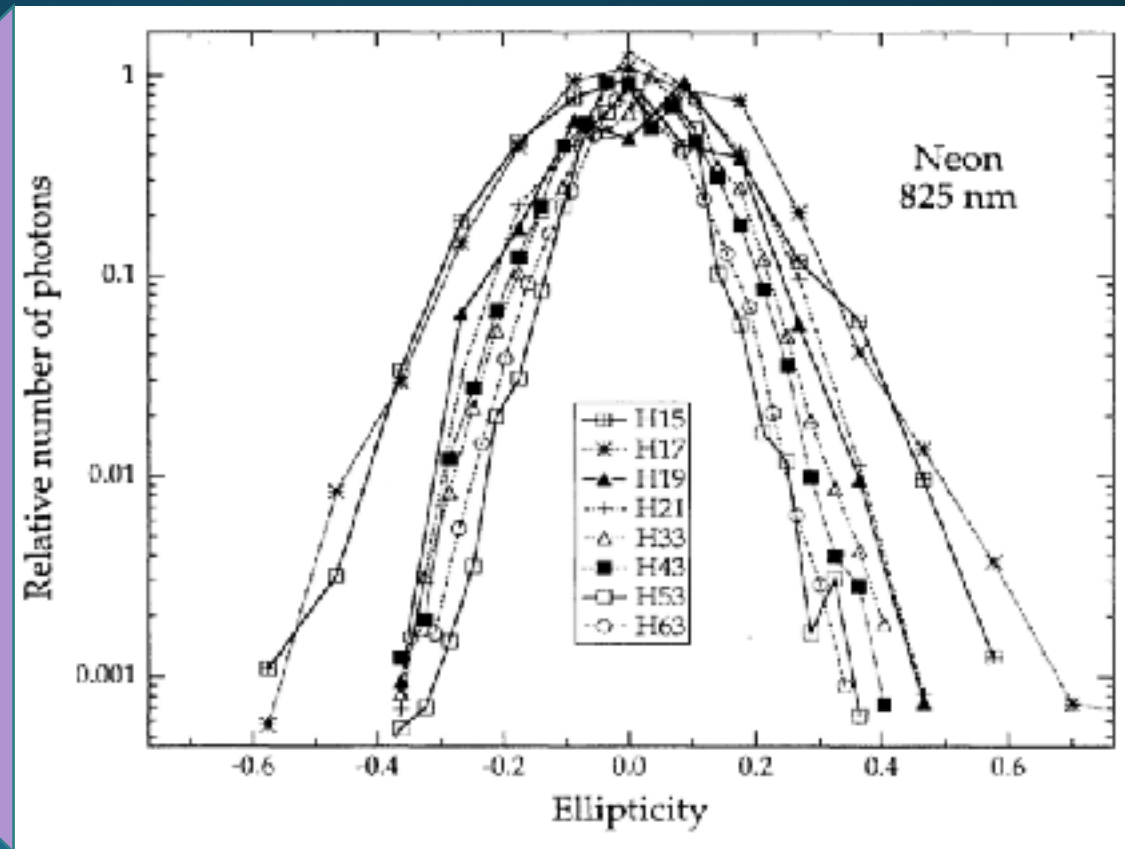
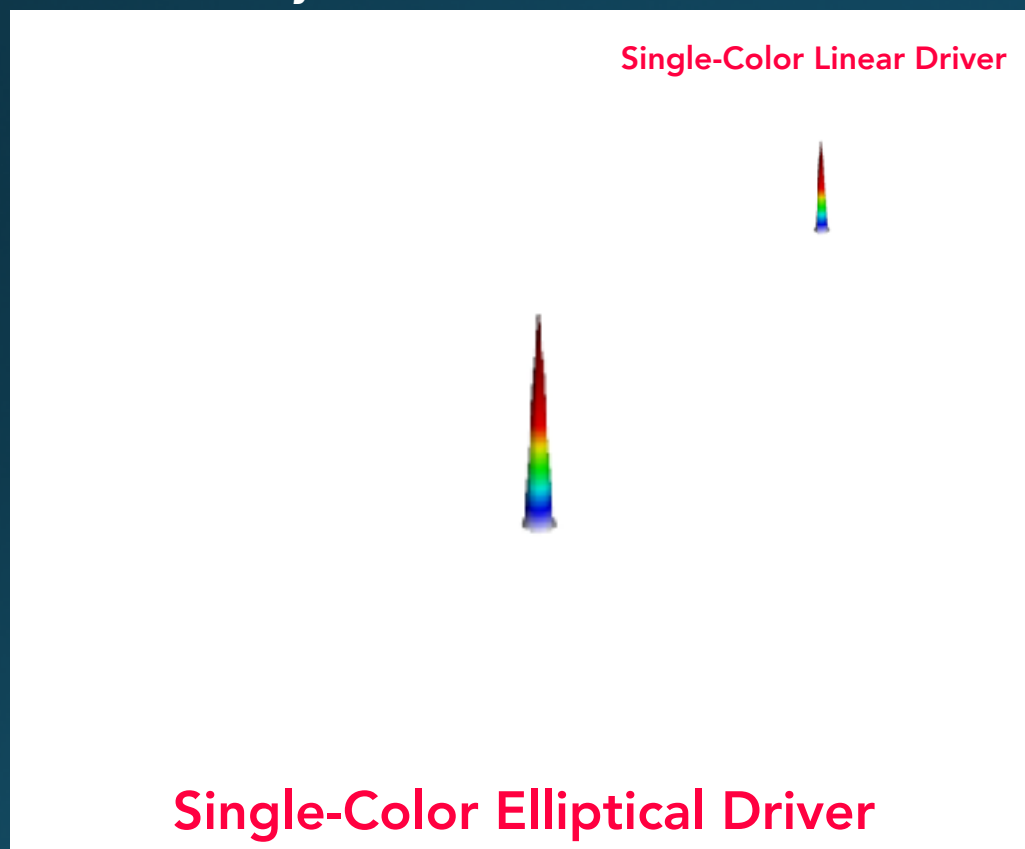
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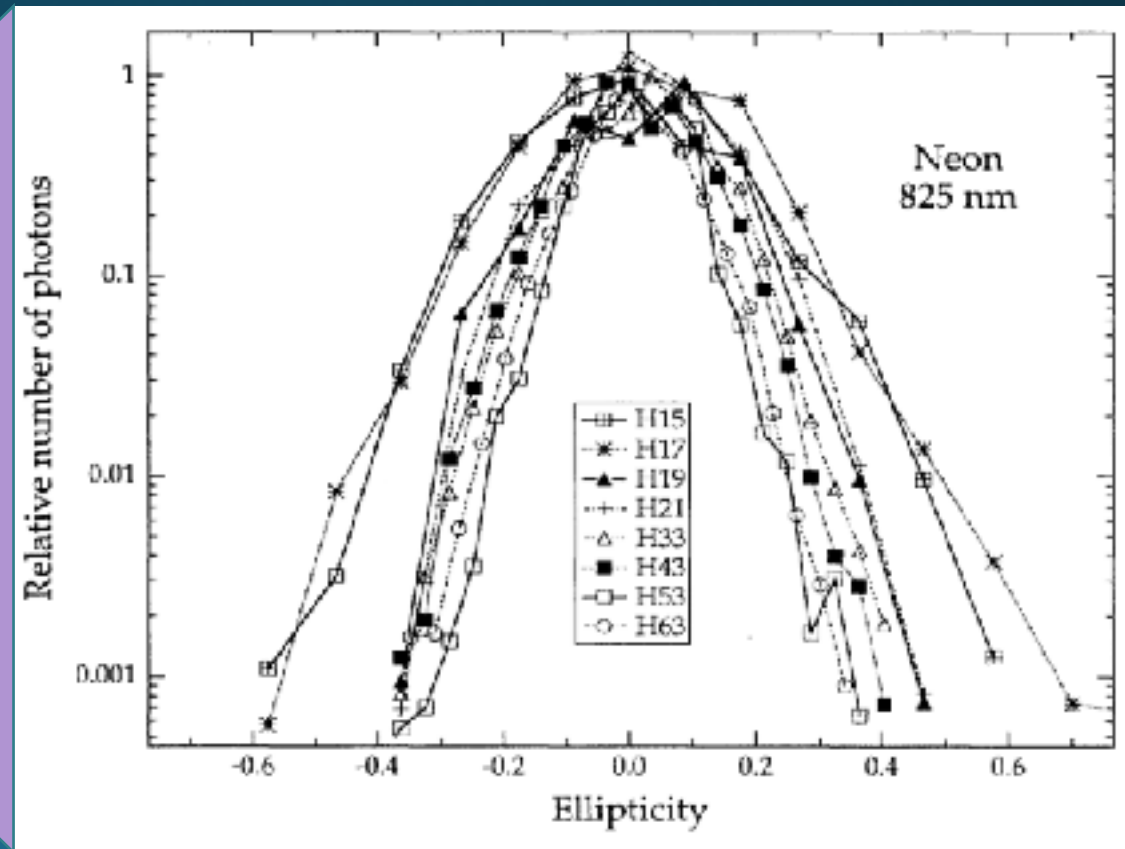
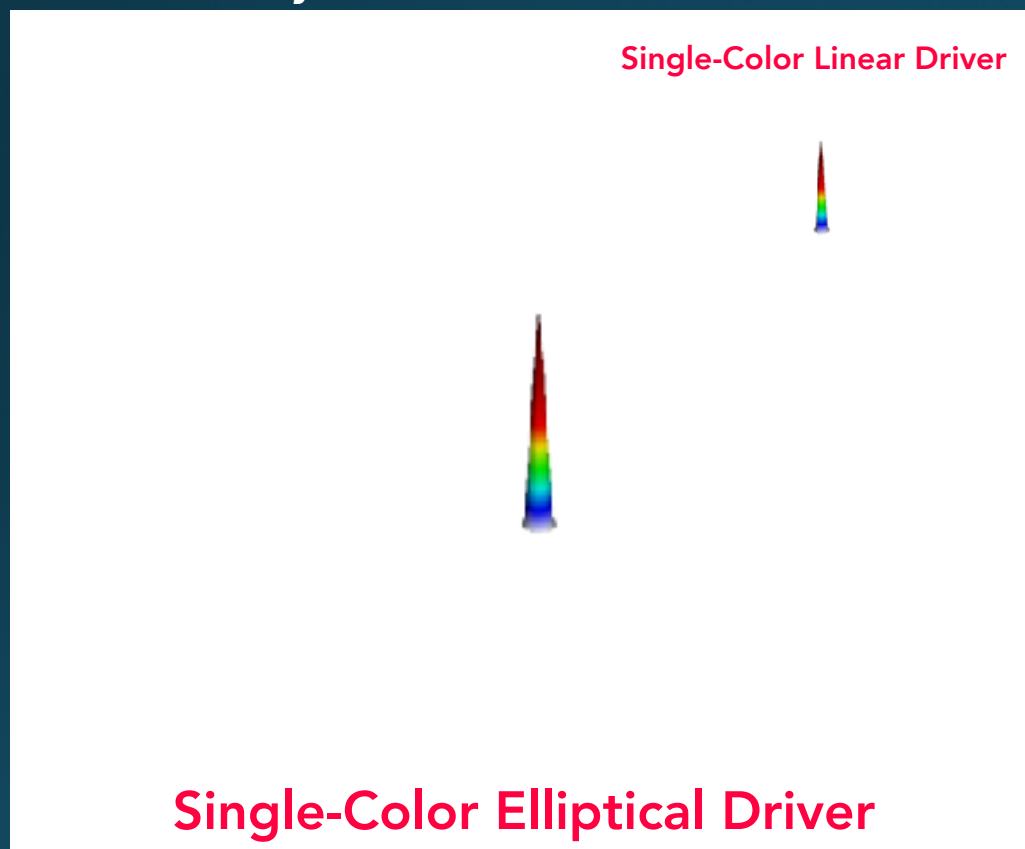
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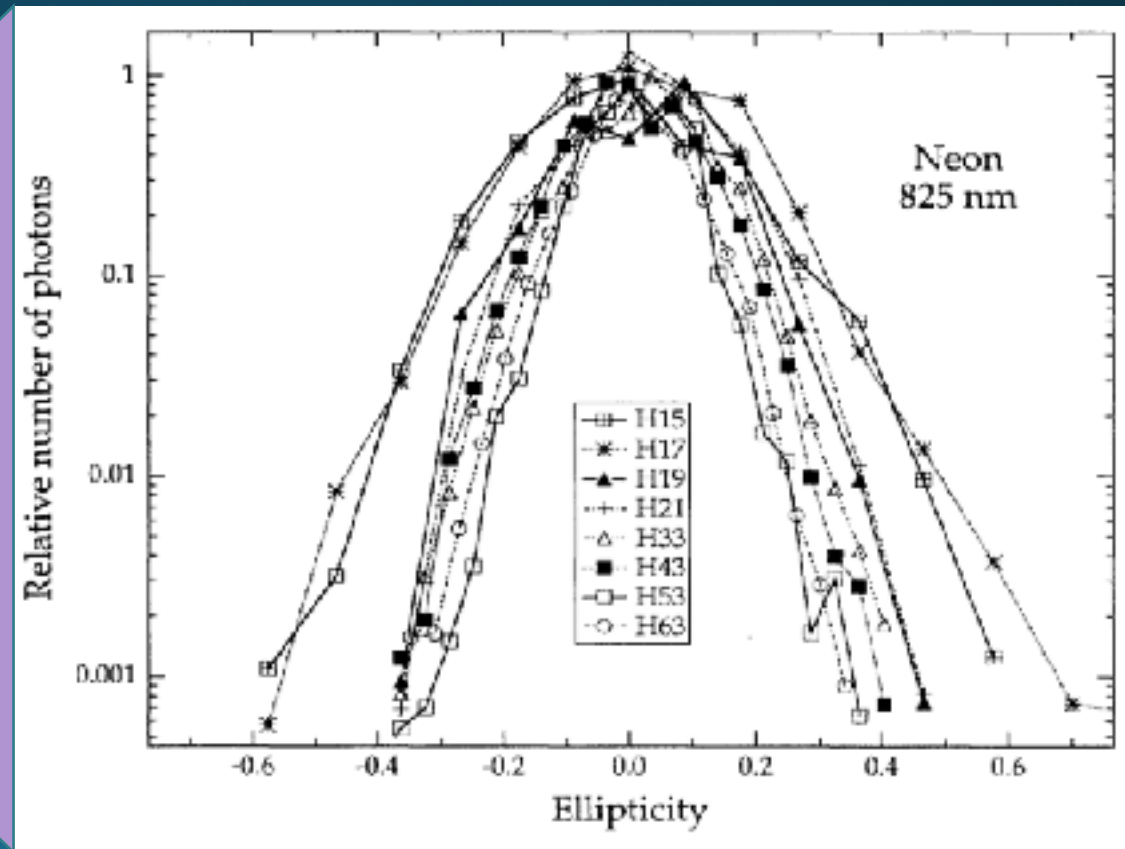
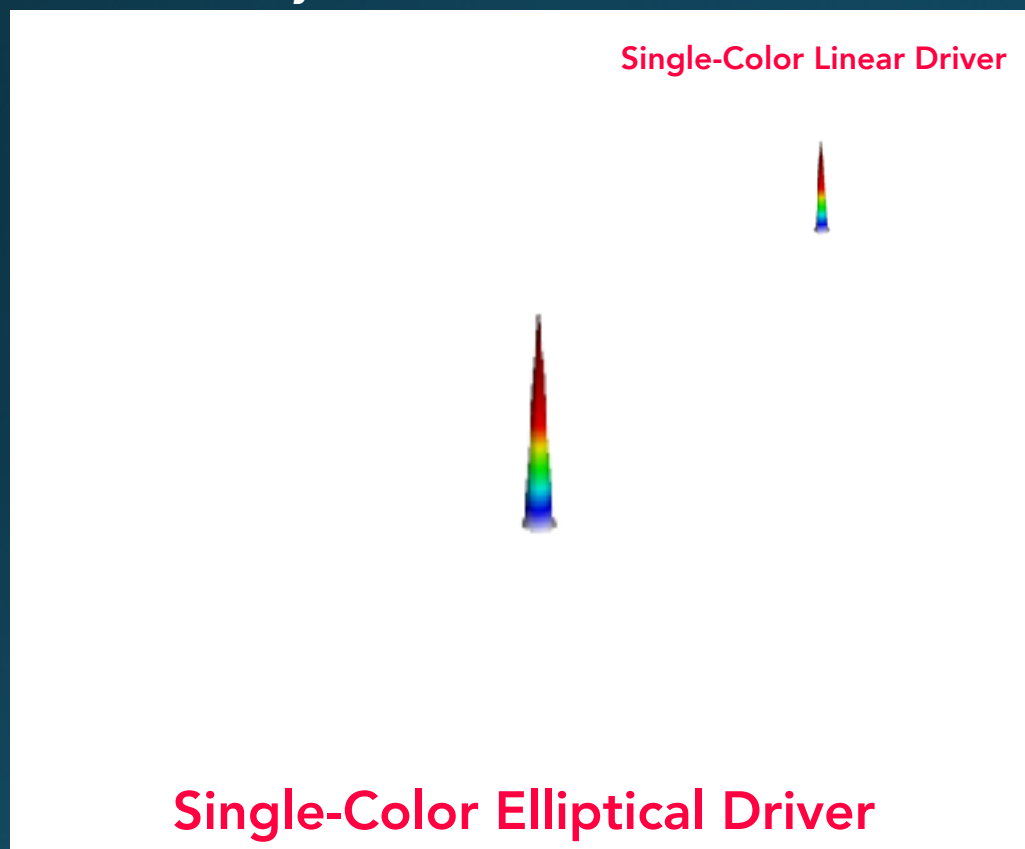
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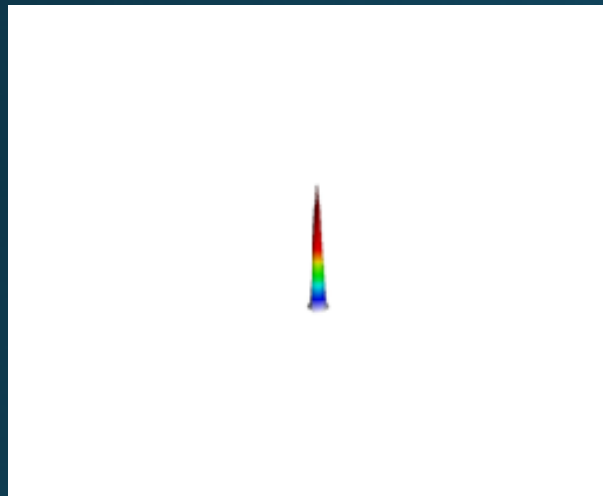


HHG in a Whole New Dimension: Reshaping the Frontier of Attosecond Science

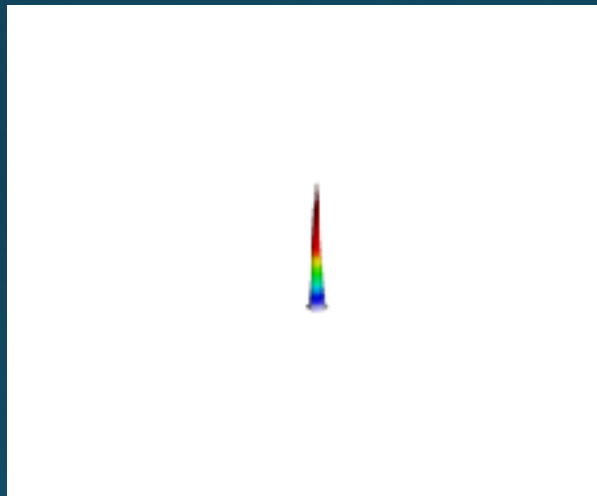


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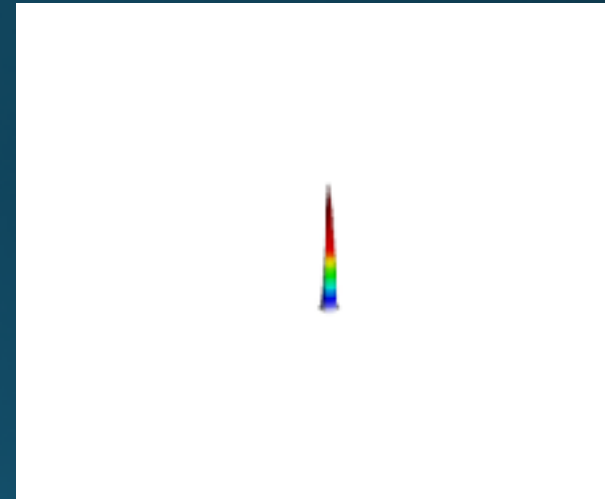
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One-Color Circular Driver



Two-Color Counter-Rotating



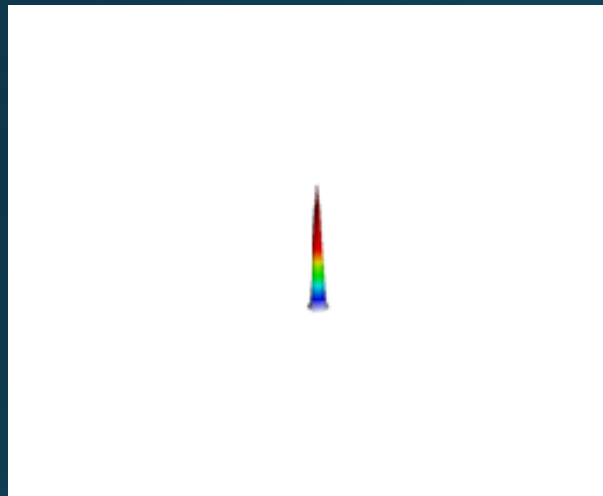


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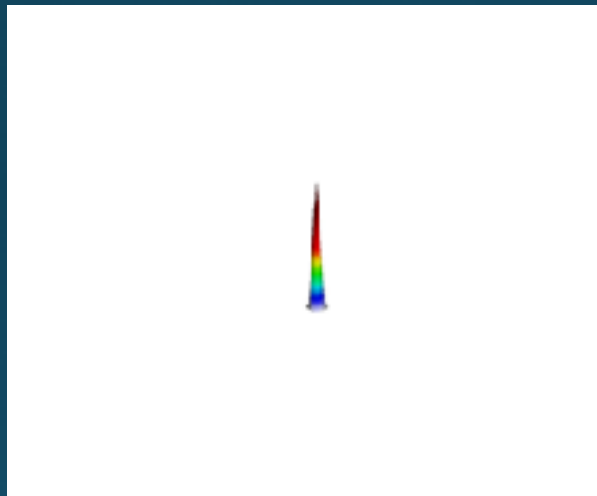


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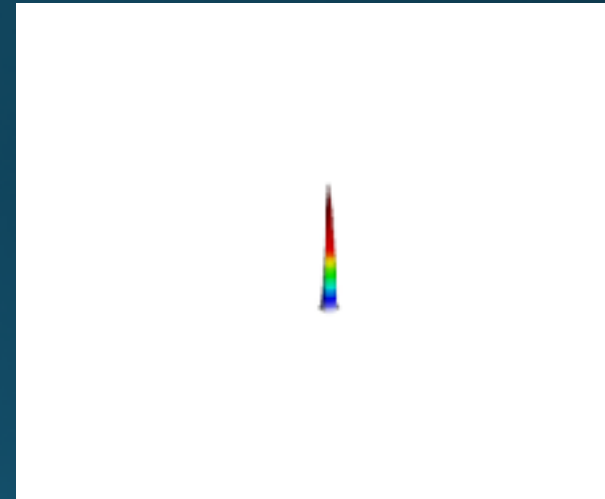
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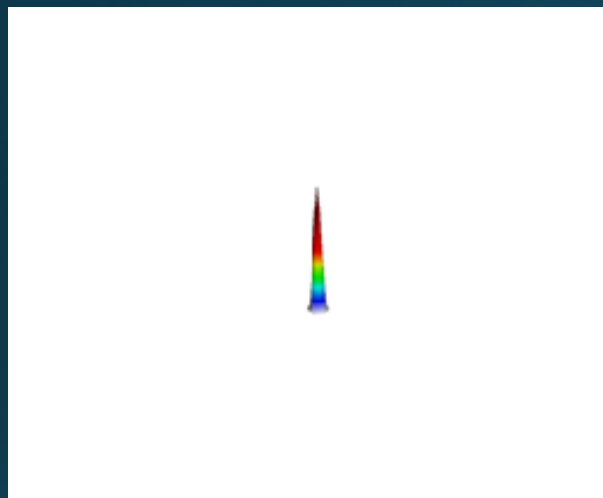


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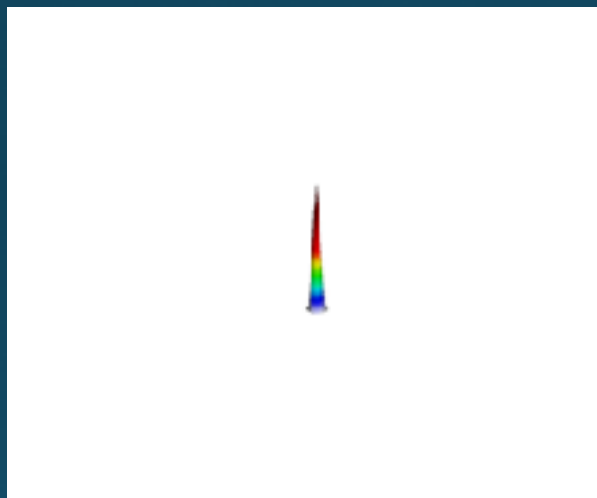


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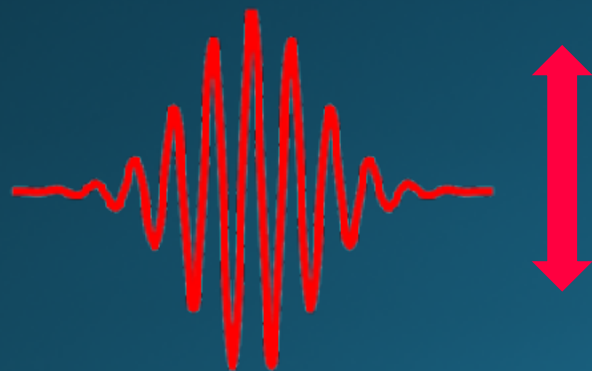
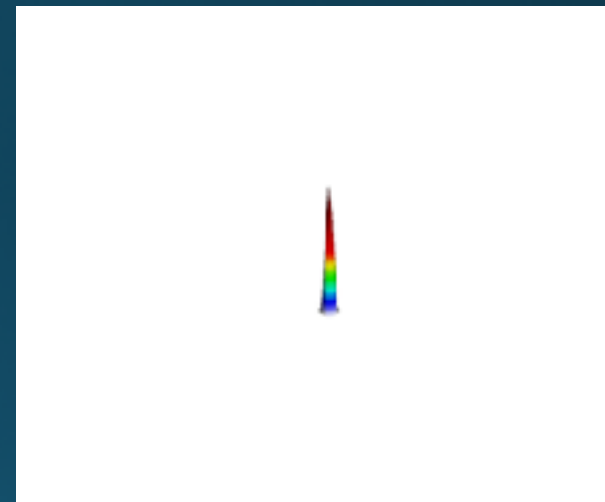
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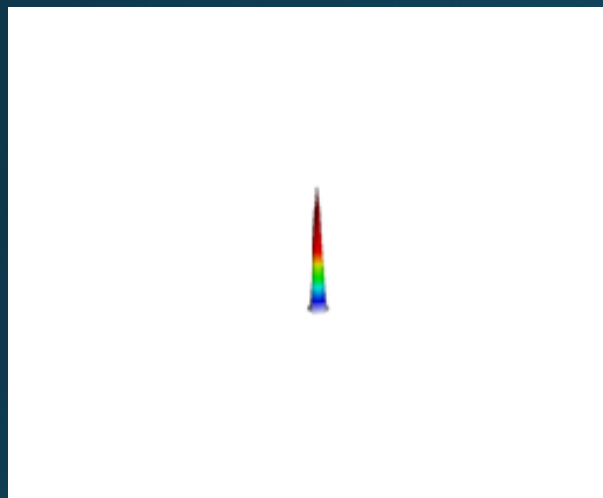


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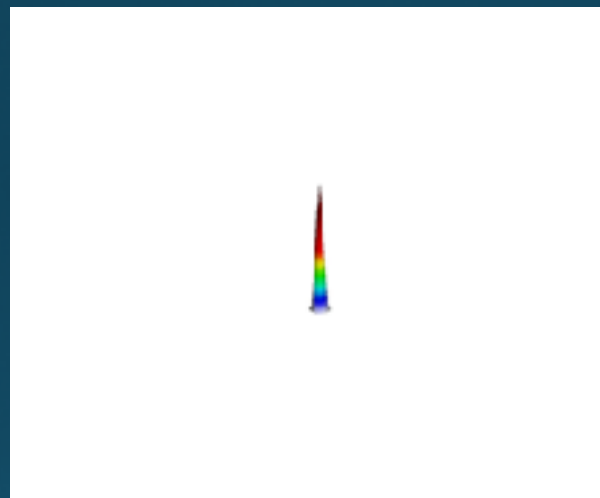


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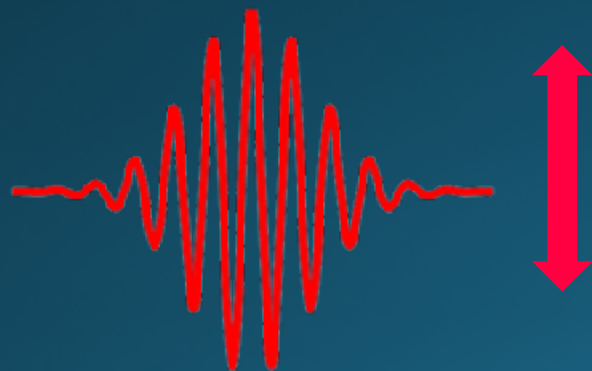
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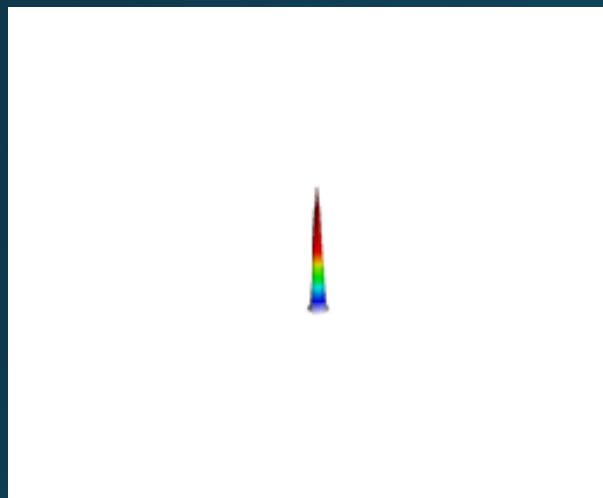


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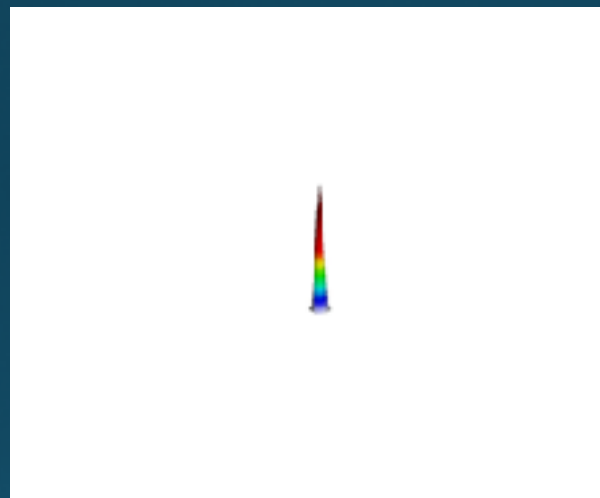


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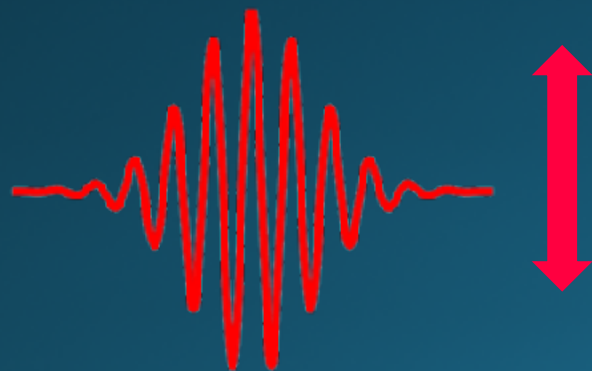
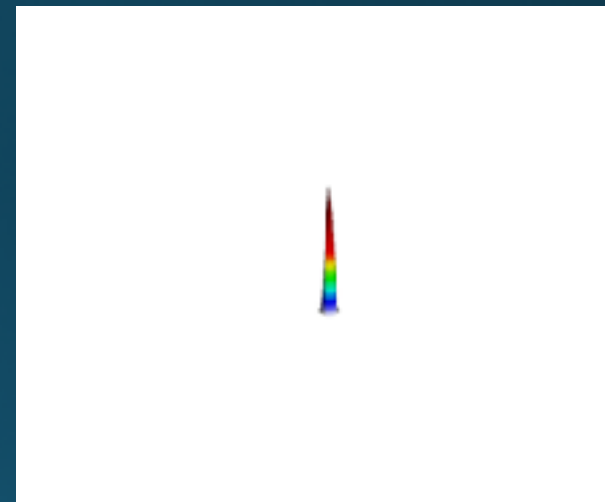
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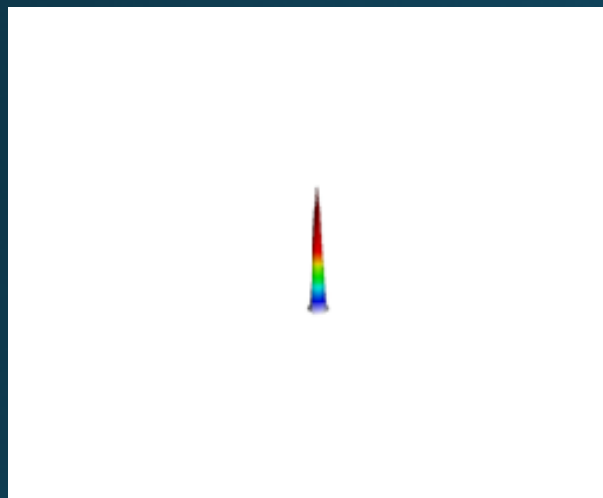


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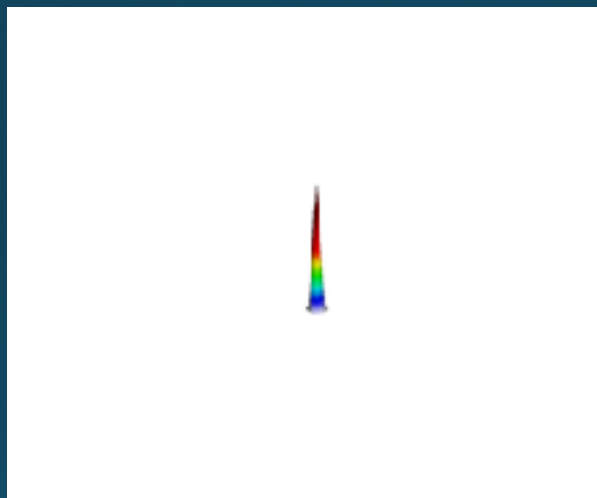


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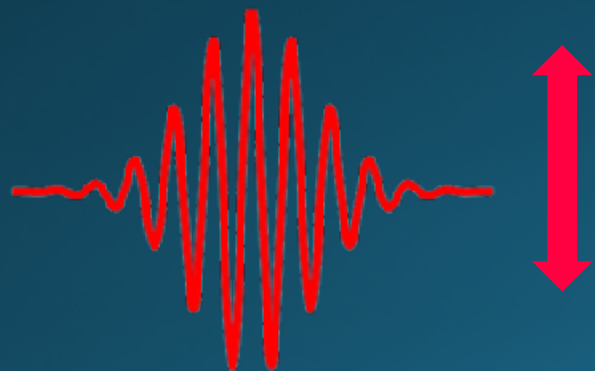
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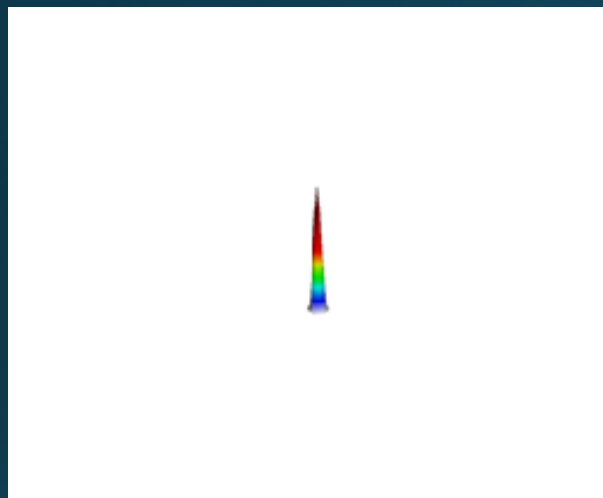


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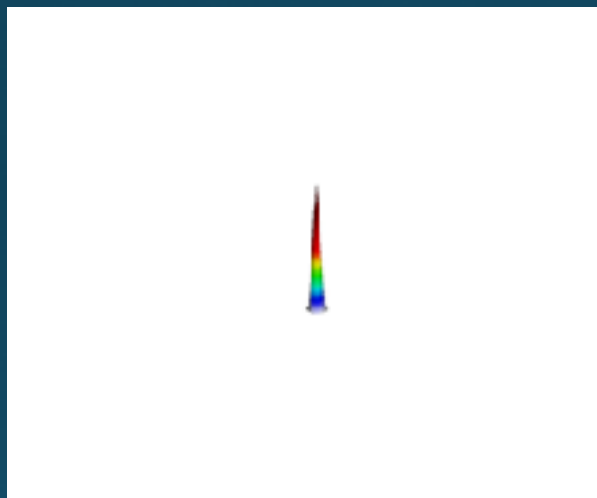


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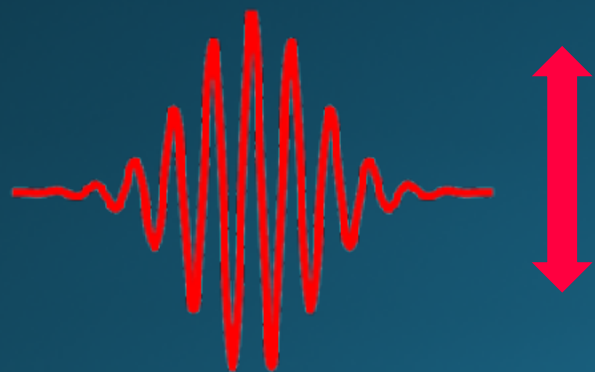
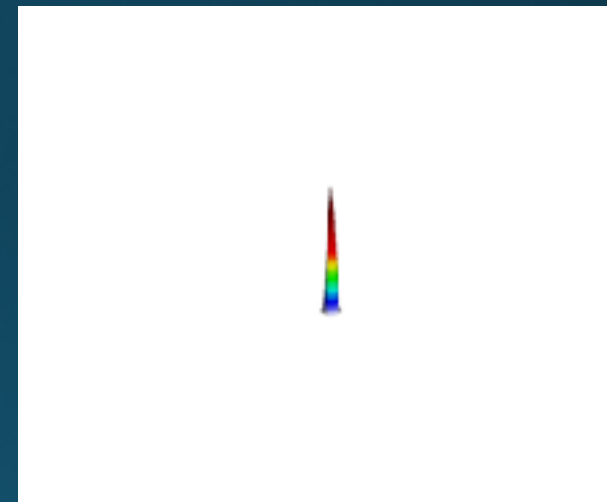
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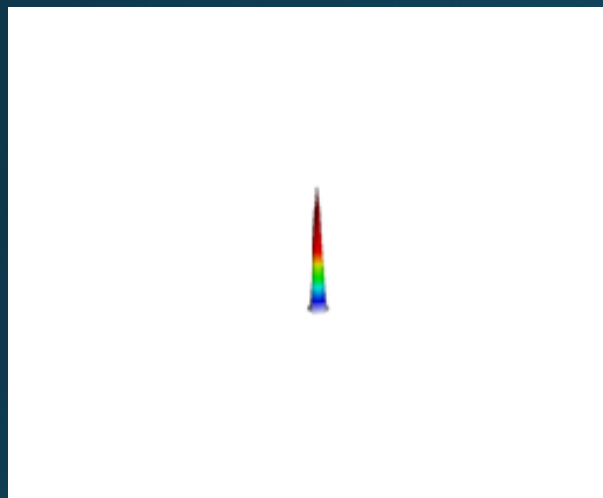


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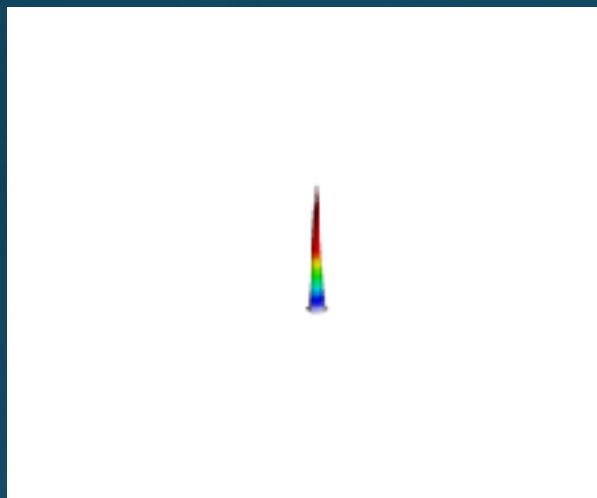


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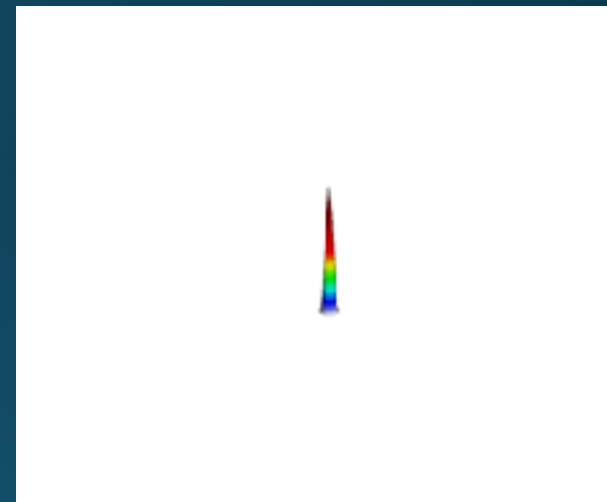
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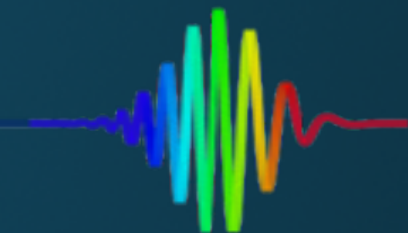
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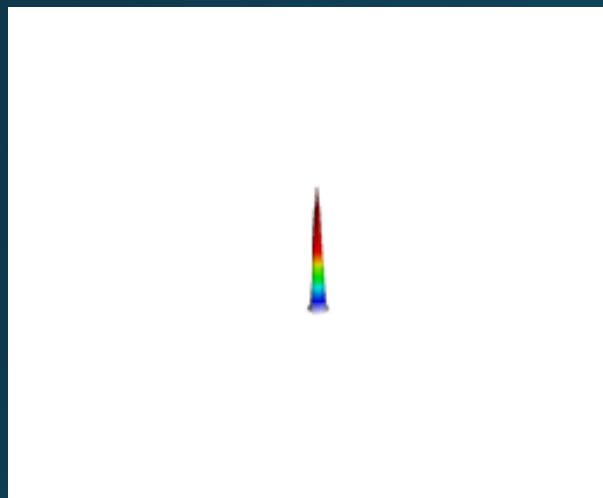


HHG in a Whole New Dimension: Reshaping the Frontier of Attosecond Science

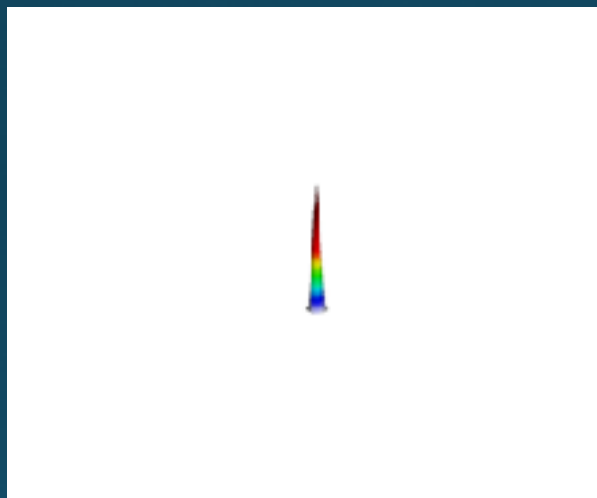


Videos: Courtesy of Carlos Hernandez-Garcia

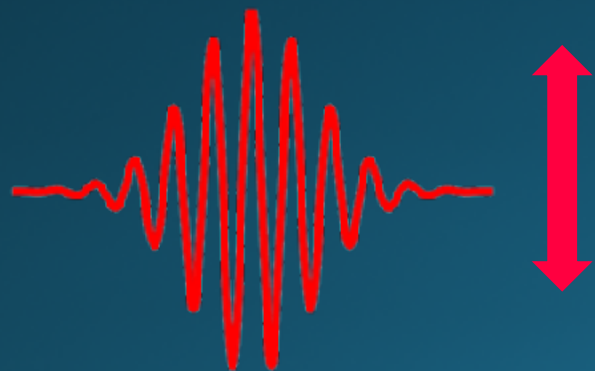
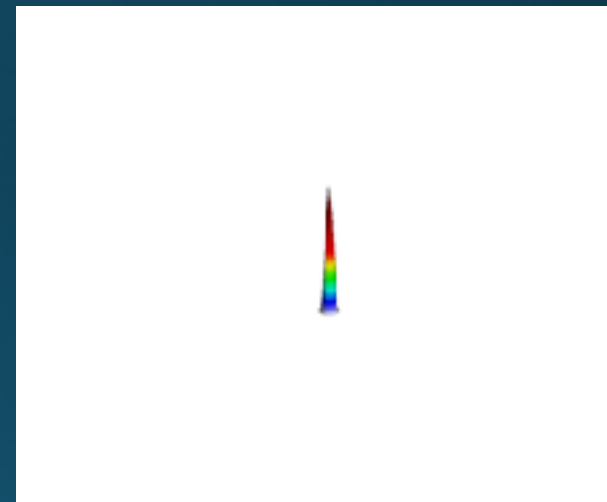
One Color Linear Driver



One-Color Circular Driver

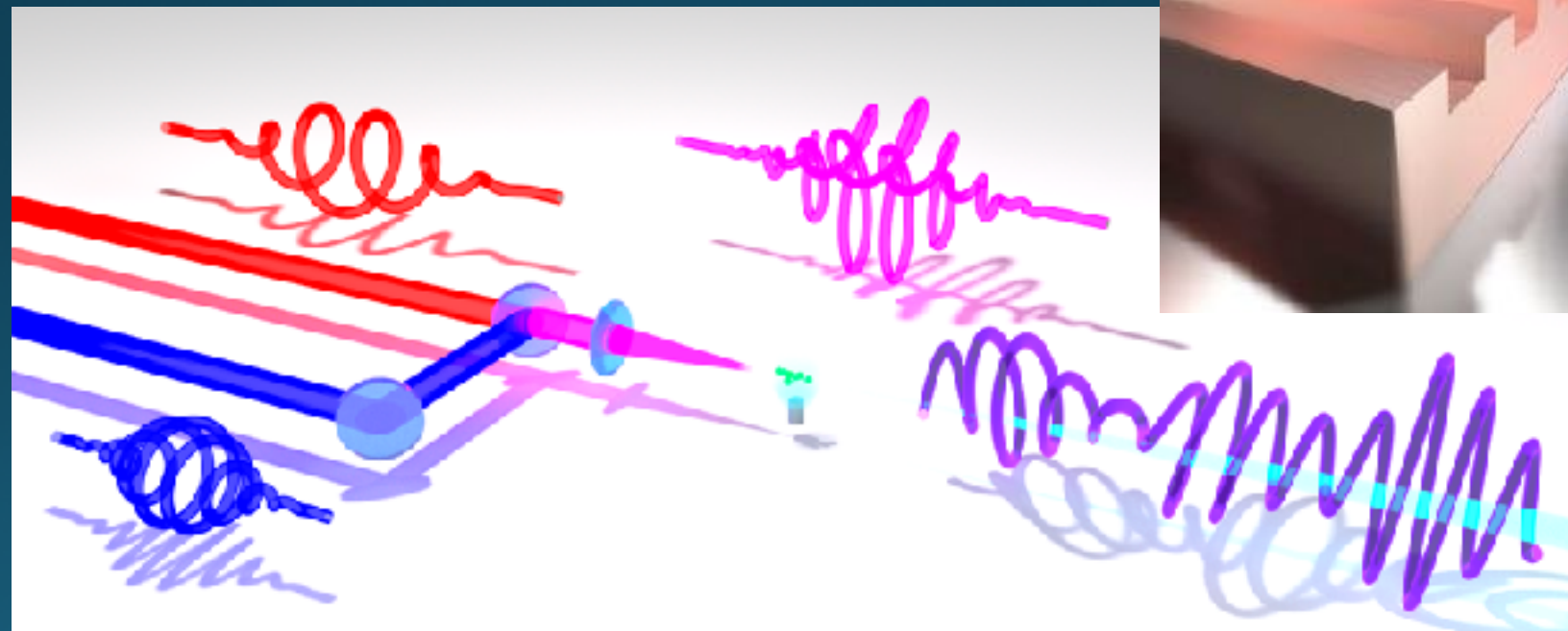
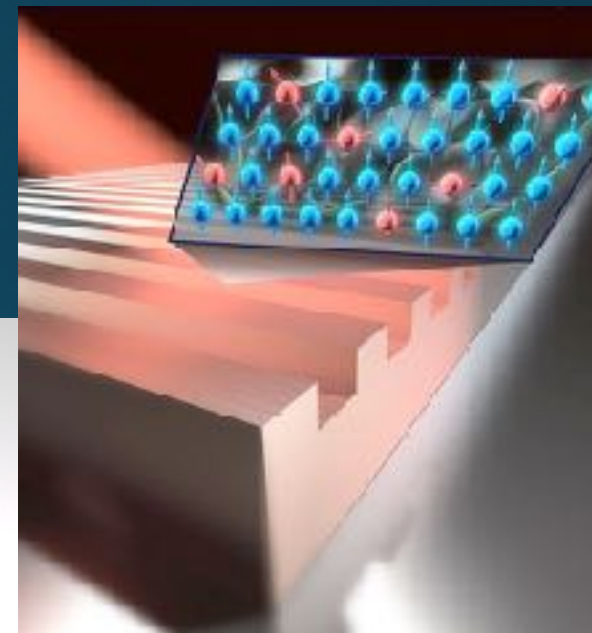
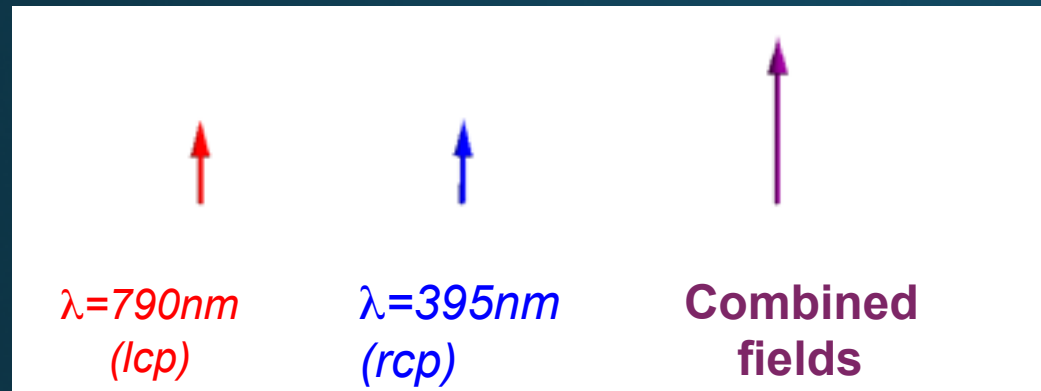


Two-Color Counter-Rotating





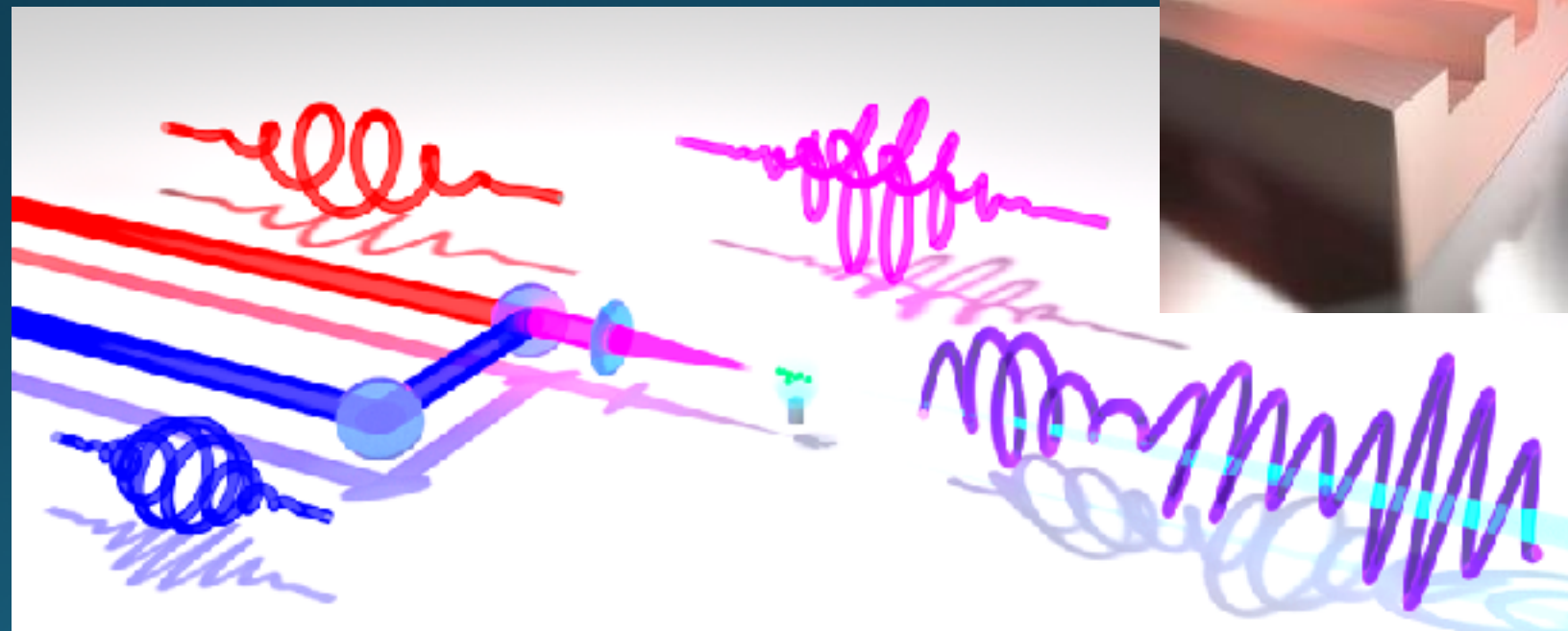
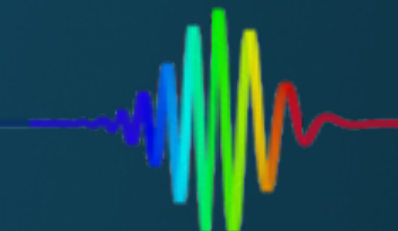
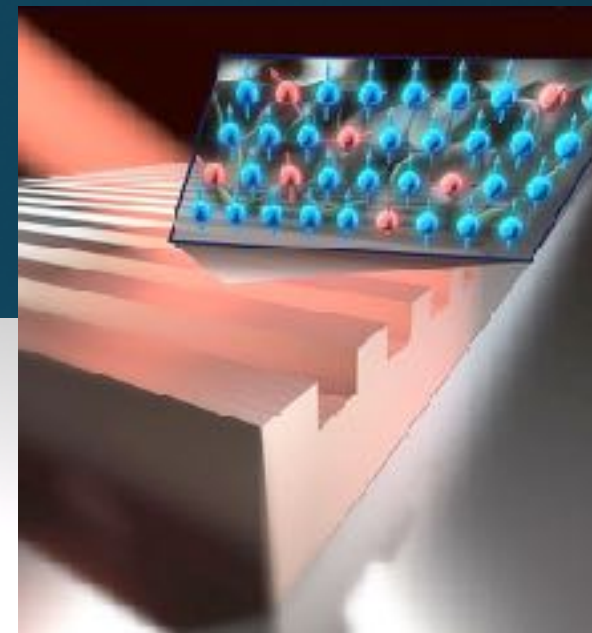
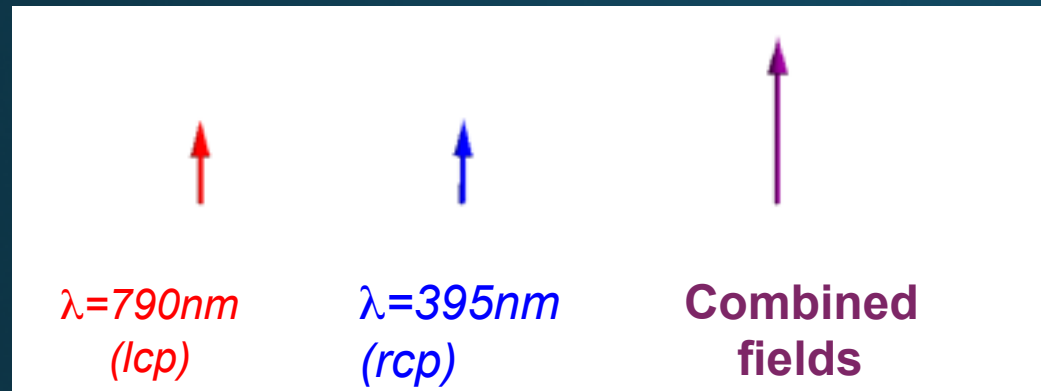
Circularly Polarized High Harmonic Generation (CPHHG): The Attosecond Blast(s) From the Past!



Eichmann, Phys. Rev. A. 51, 1995
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Fan, PNAS, 112, 2015
Dorney, PRL, 119, 2017
Many, many, more



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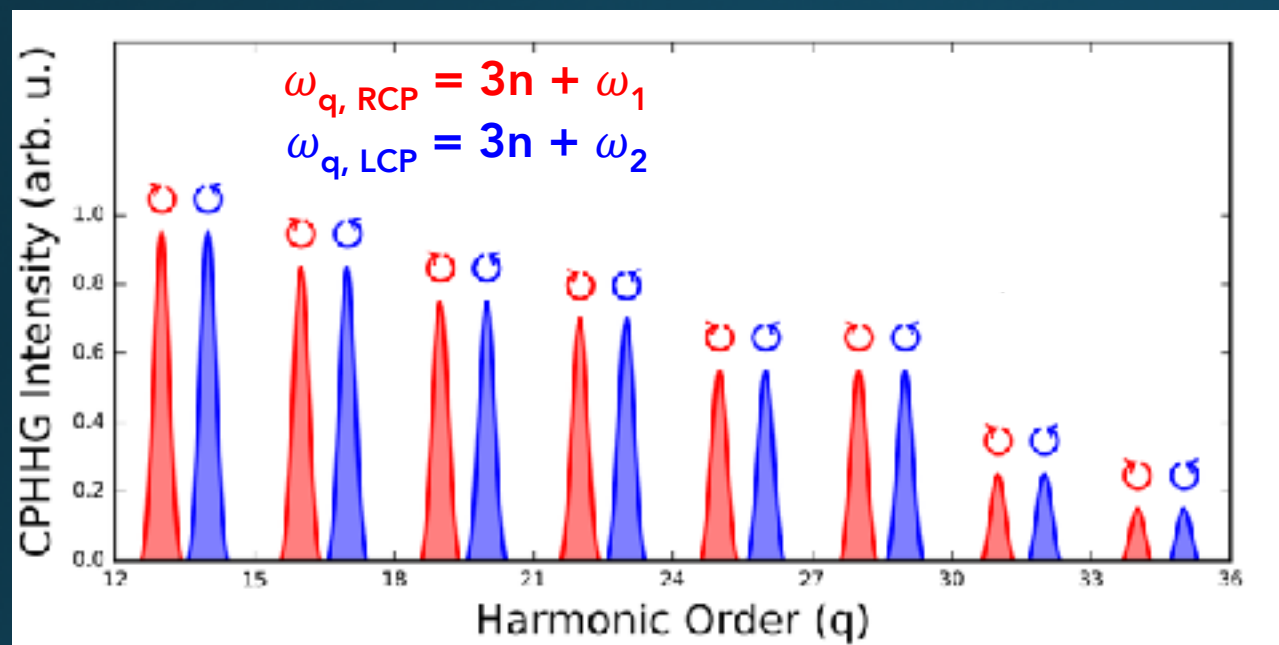
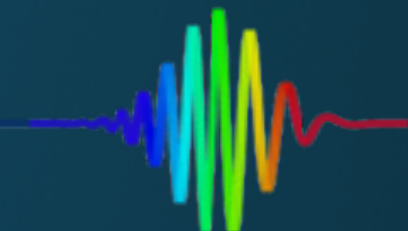


Circularly Polarized Harmonics, You Say?
Well, Surely We Have Circular Attosecond Pulses!



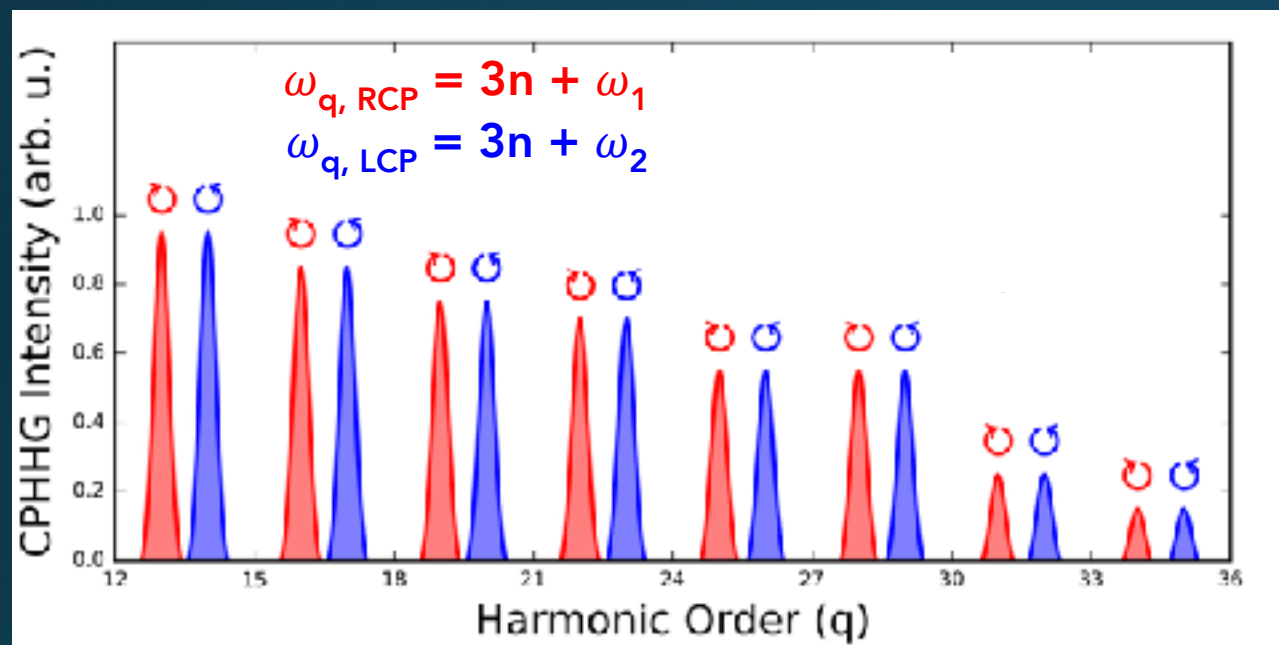
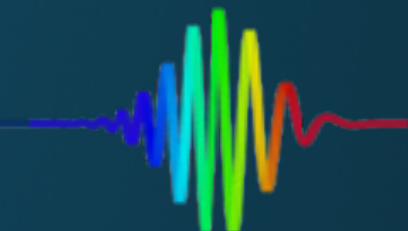
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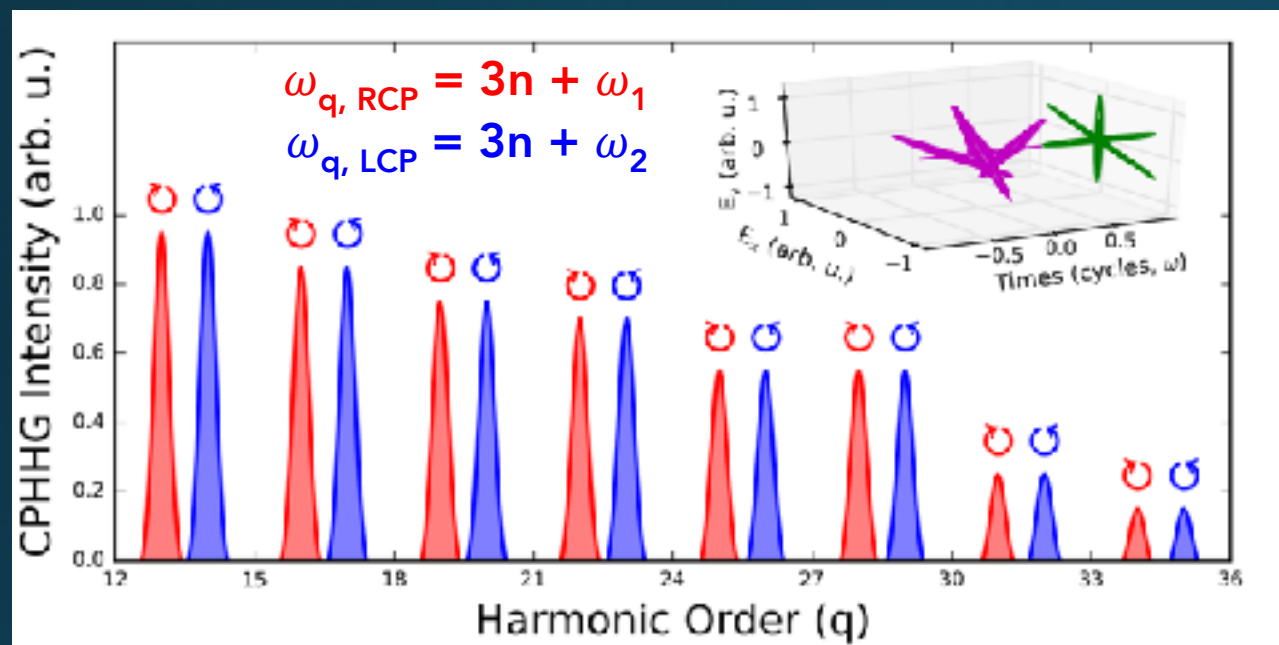
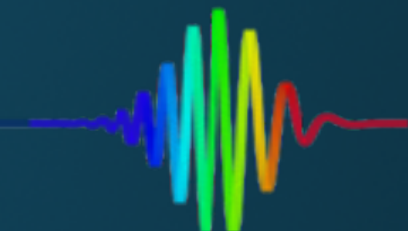
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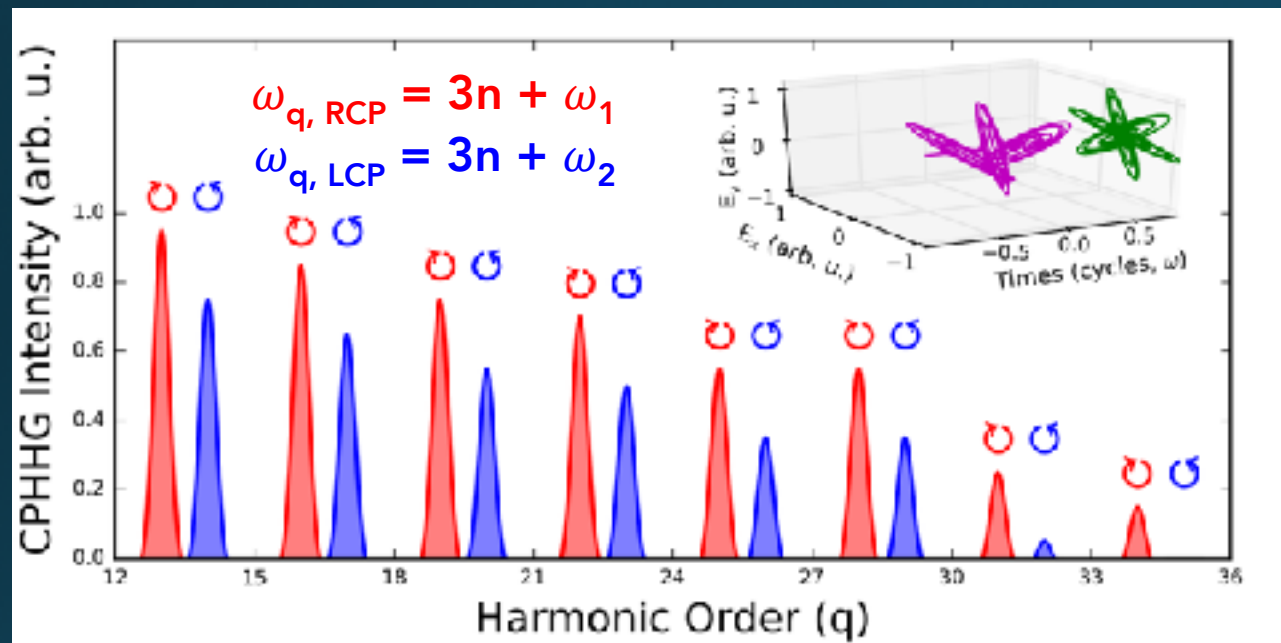
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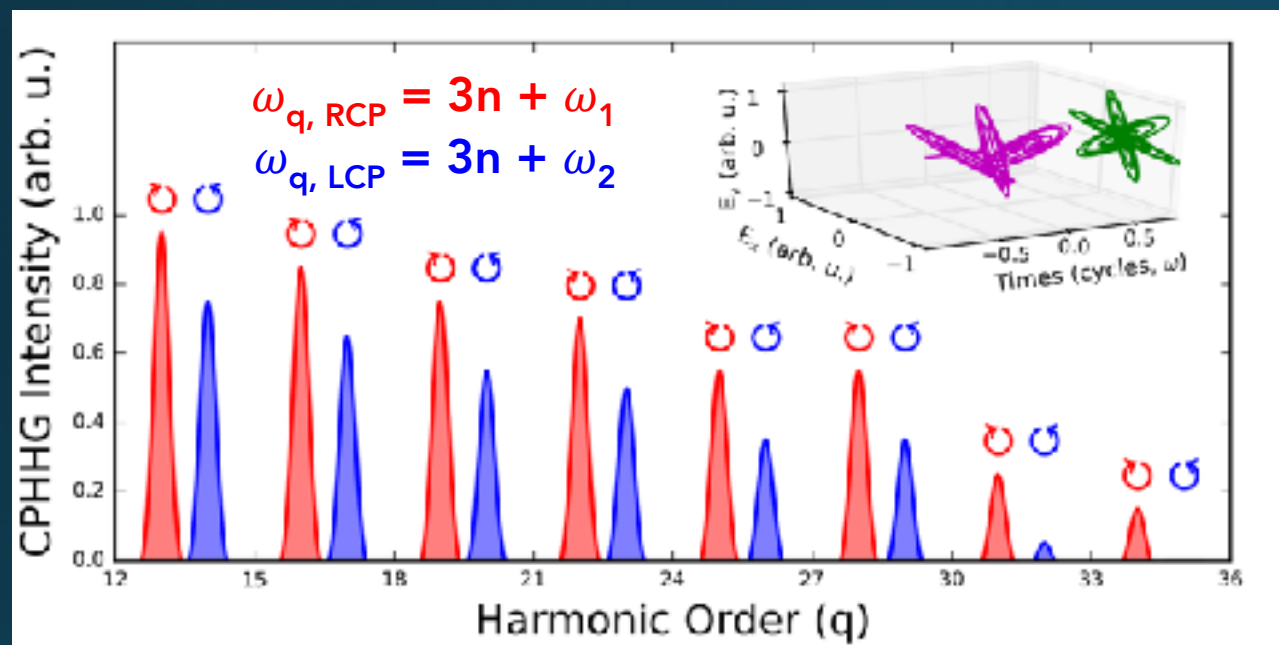


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Li, Opt. Quant. Electron. 49, 2017

Lerner, Opt. Lett. 42, 2017

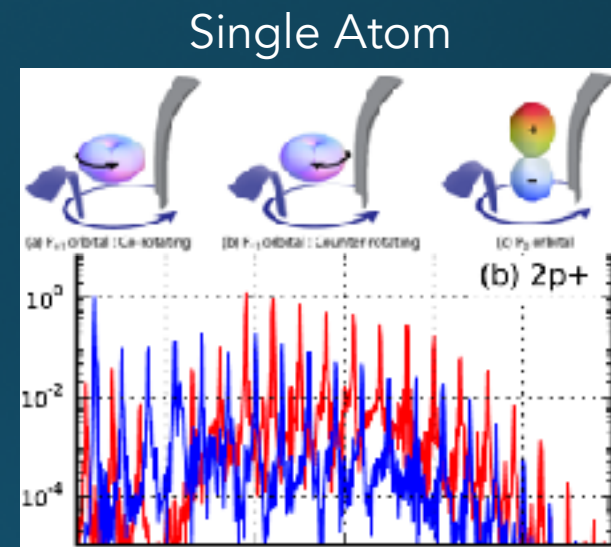
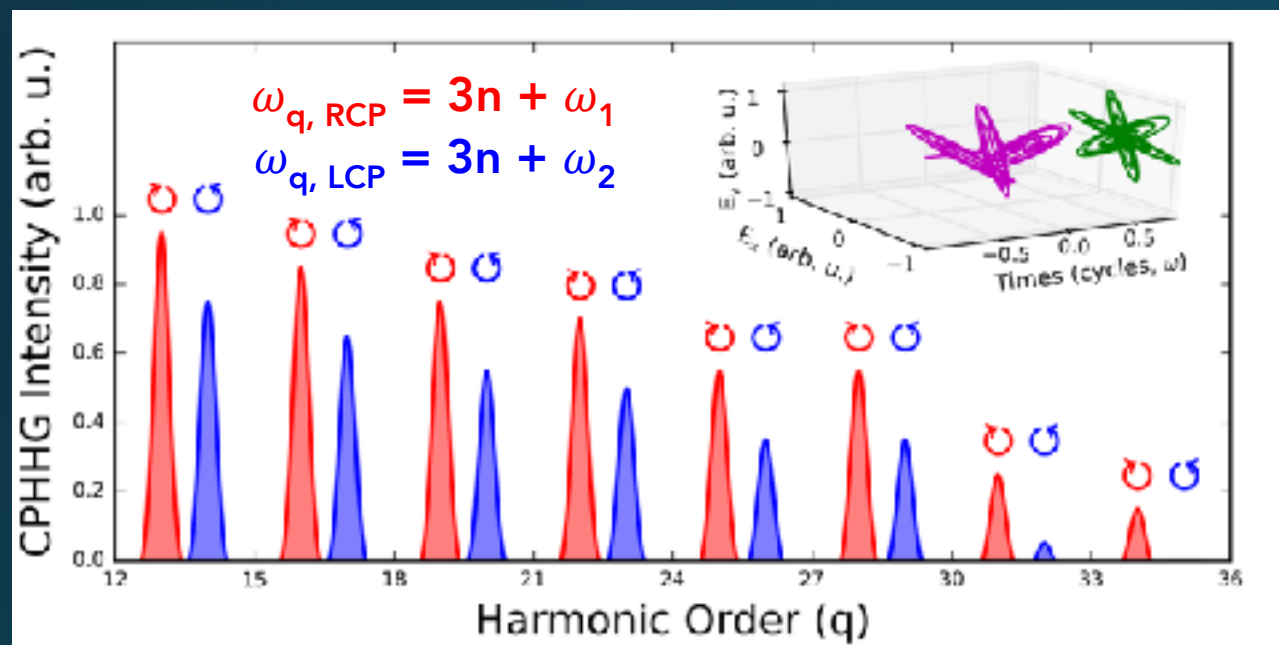
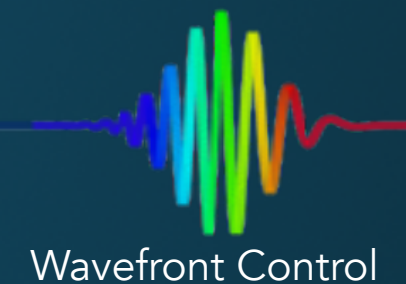
Skantzakis, Sci. Rep., 6, 2016

Yuan, Phys. Rev. Lett. 110, 2013

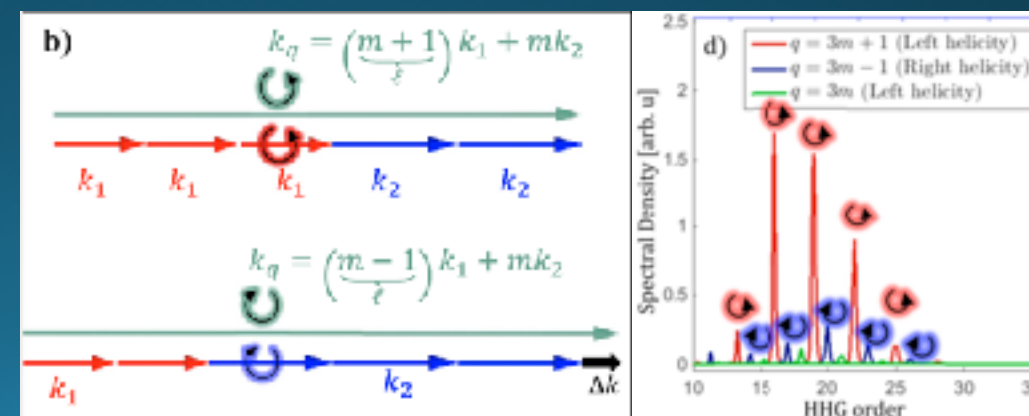
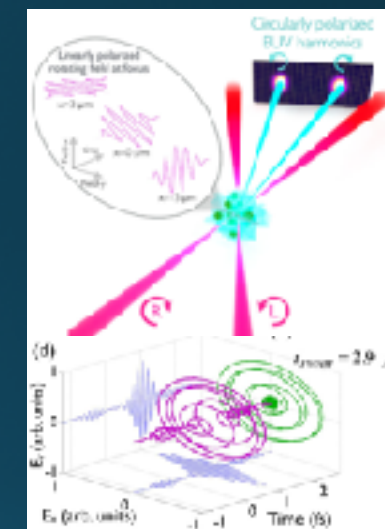
Zhang, Opt. Lett. 42, 2017

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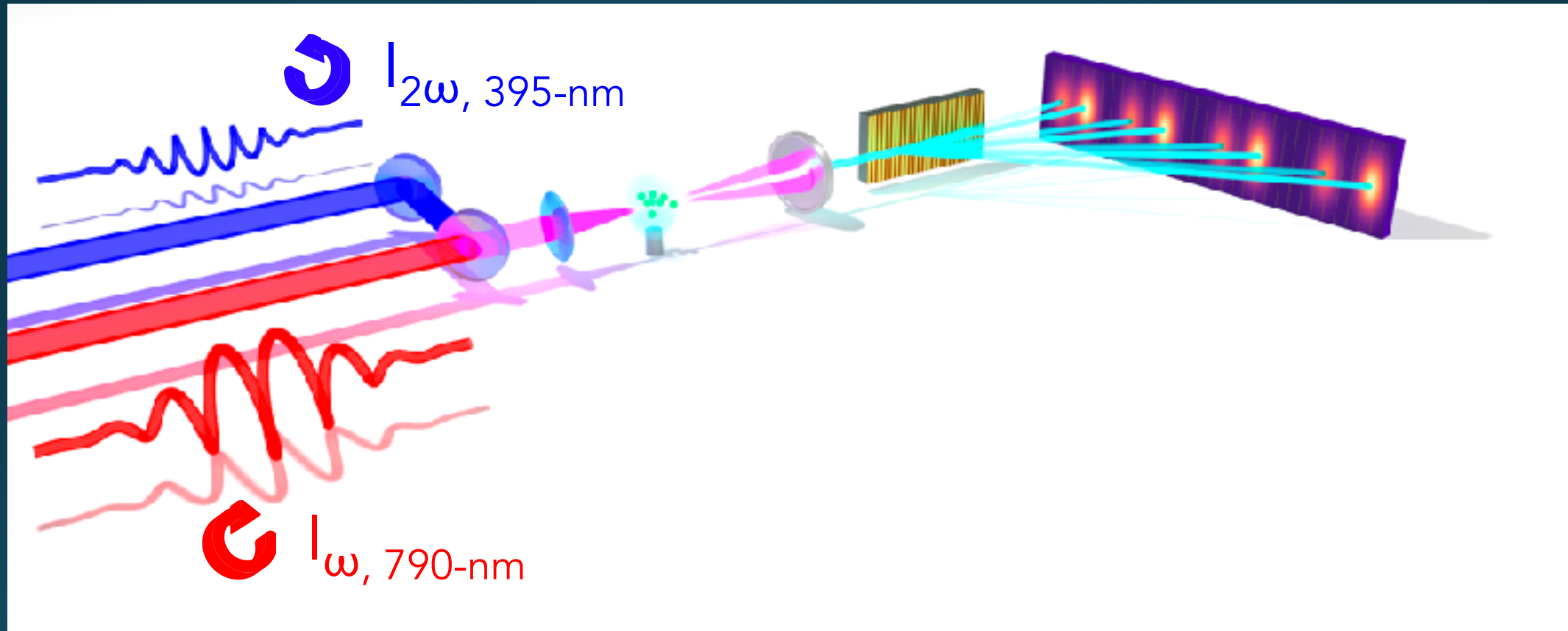
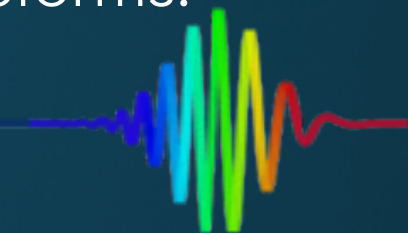


Macroscopic Control



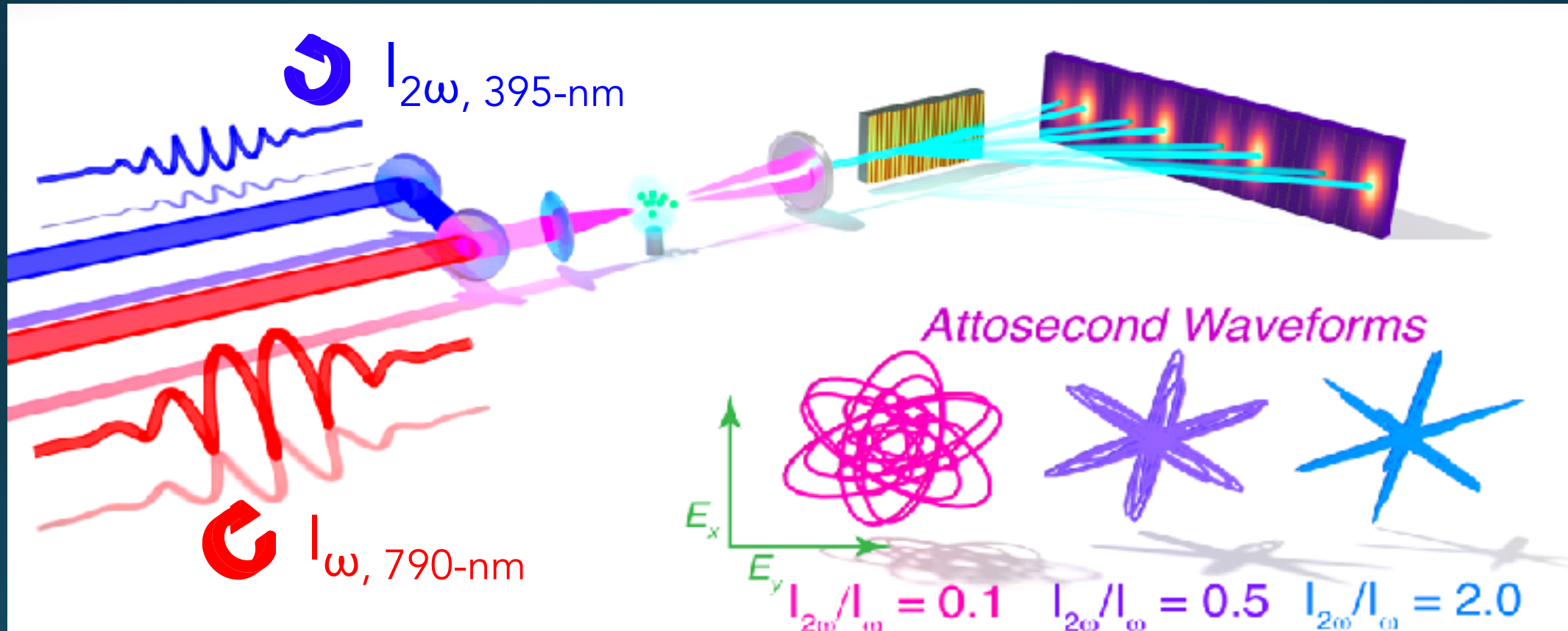
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Active Control Over the Polarization of High-Harmonic Waveforms: Production of Elliptically Polarized Attosecond Pulse Trains!



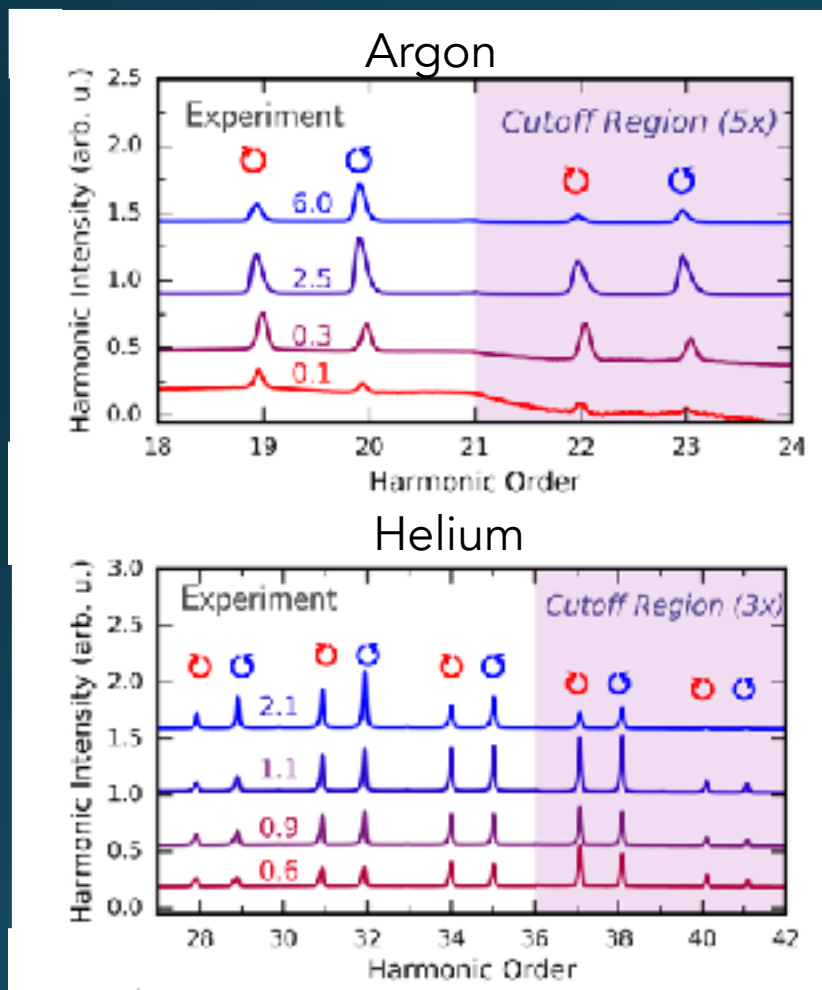
Dorney et. al., Phys. Rev. Lett., 119, 2017

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Dorney et. al., Phys. Rev. Lett., 119, 2017

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



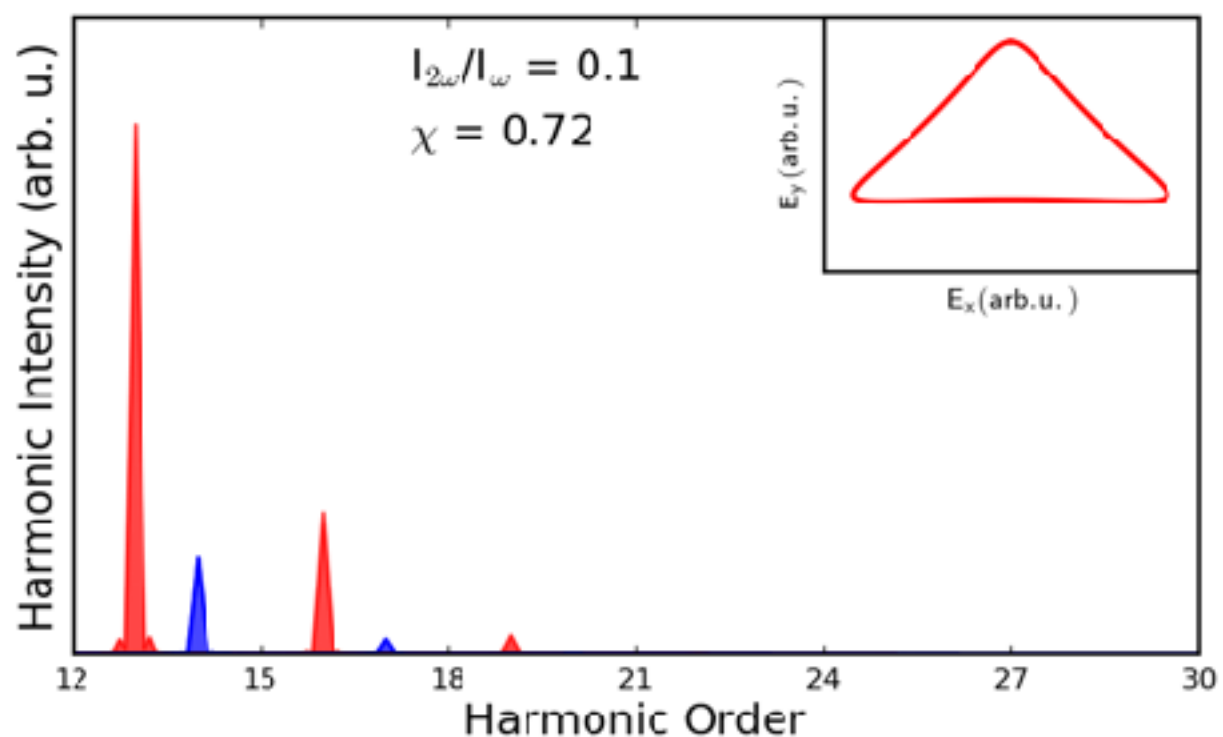
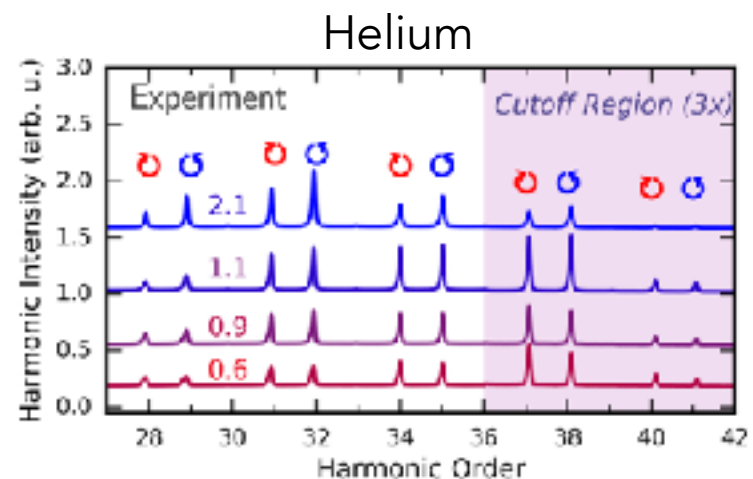
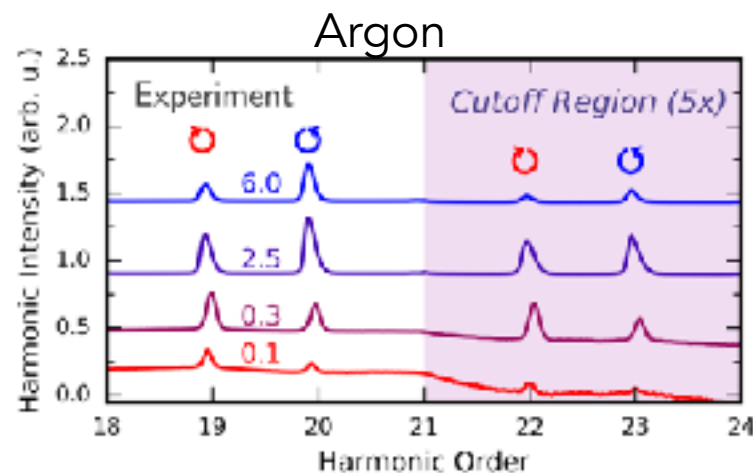
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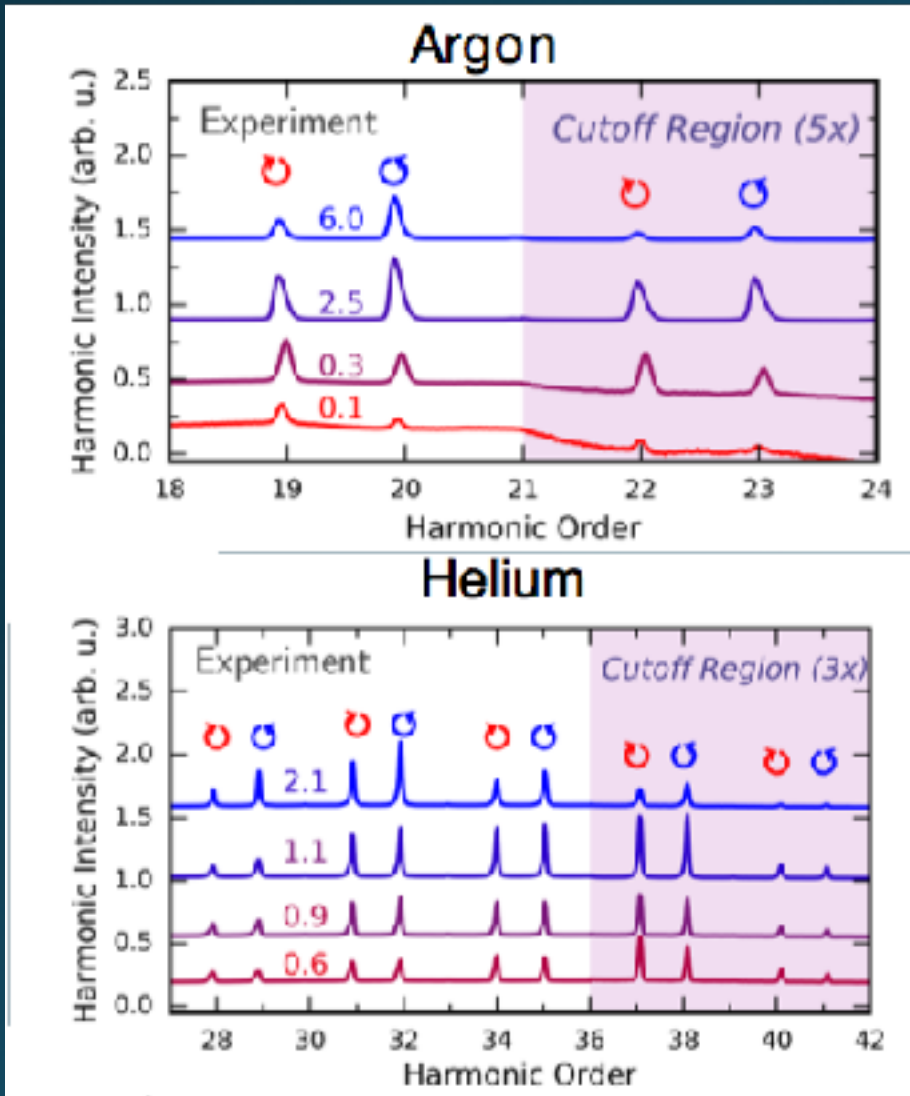
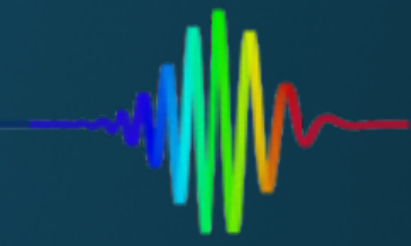


$$\chi = \frac{(I_{RCP} - I_{LCP})}{(I_{RCP} + I_{LCP})}$$

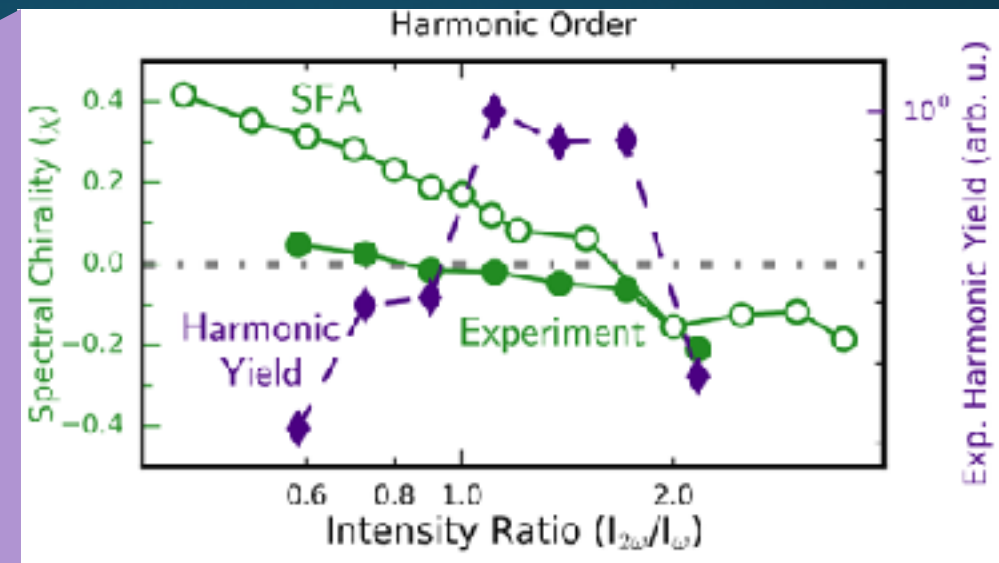
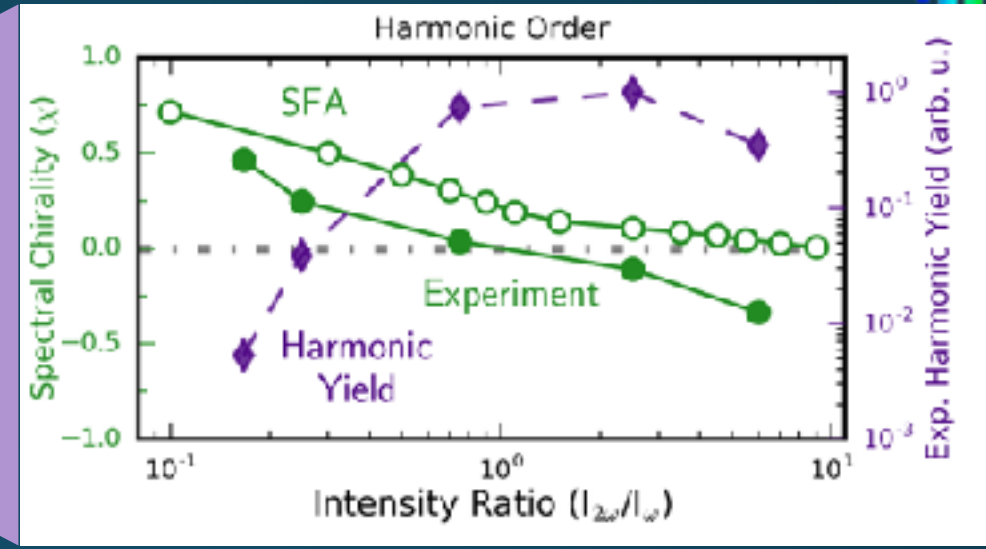
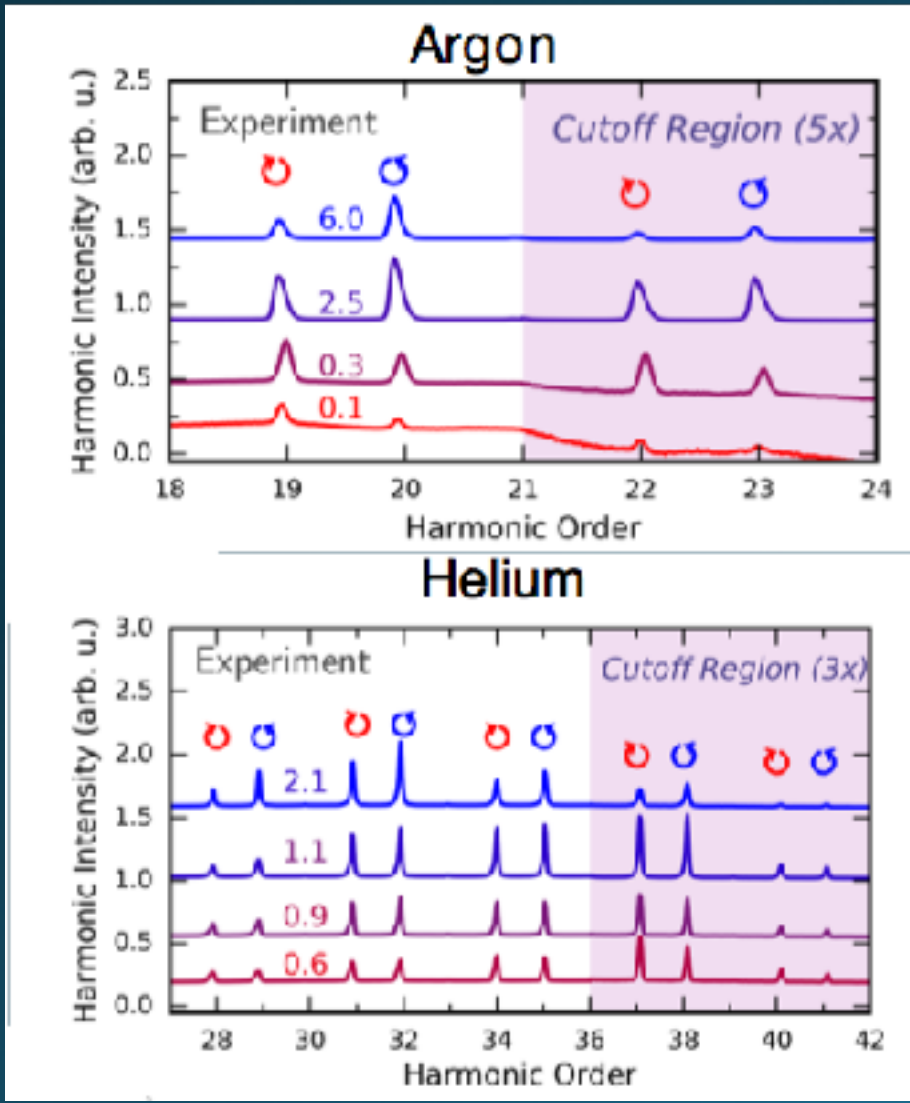
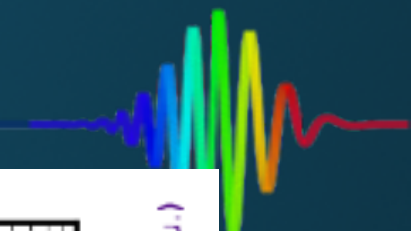
SFA Simulation in Ar



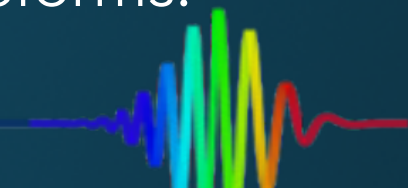
Dorney et. al., Phys. Rev. Lett., 119, 2017



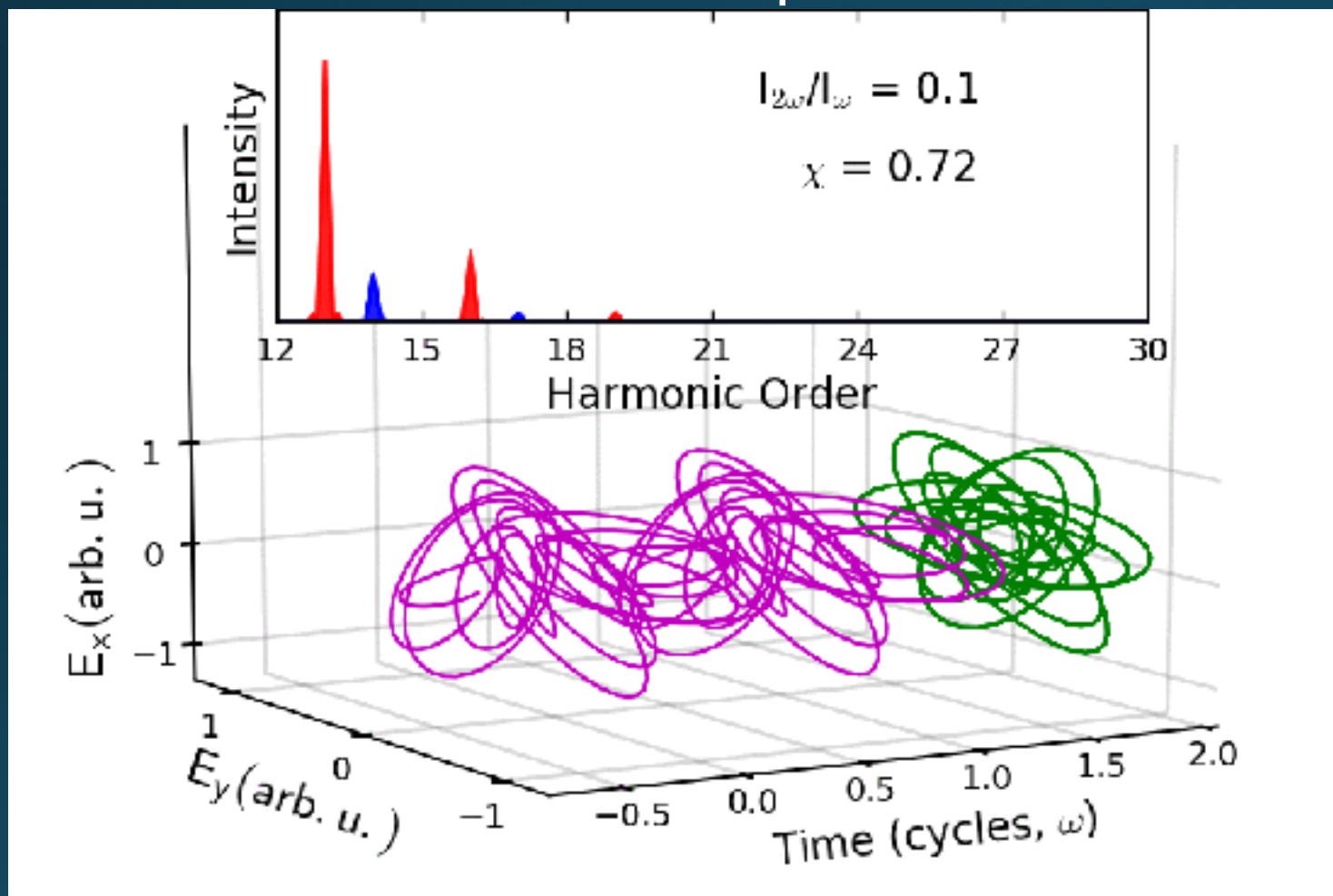
Helicity-Selective CPHHG: Frequency-Invariant Chiral Control and Preservation of Spectral Polarization



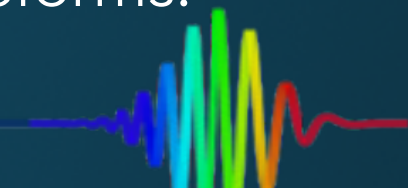
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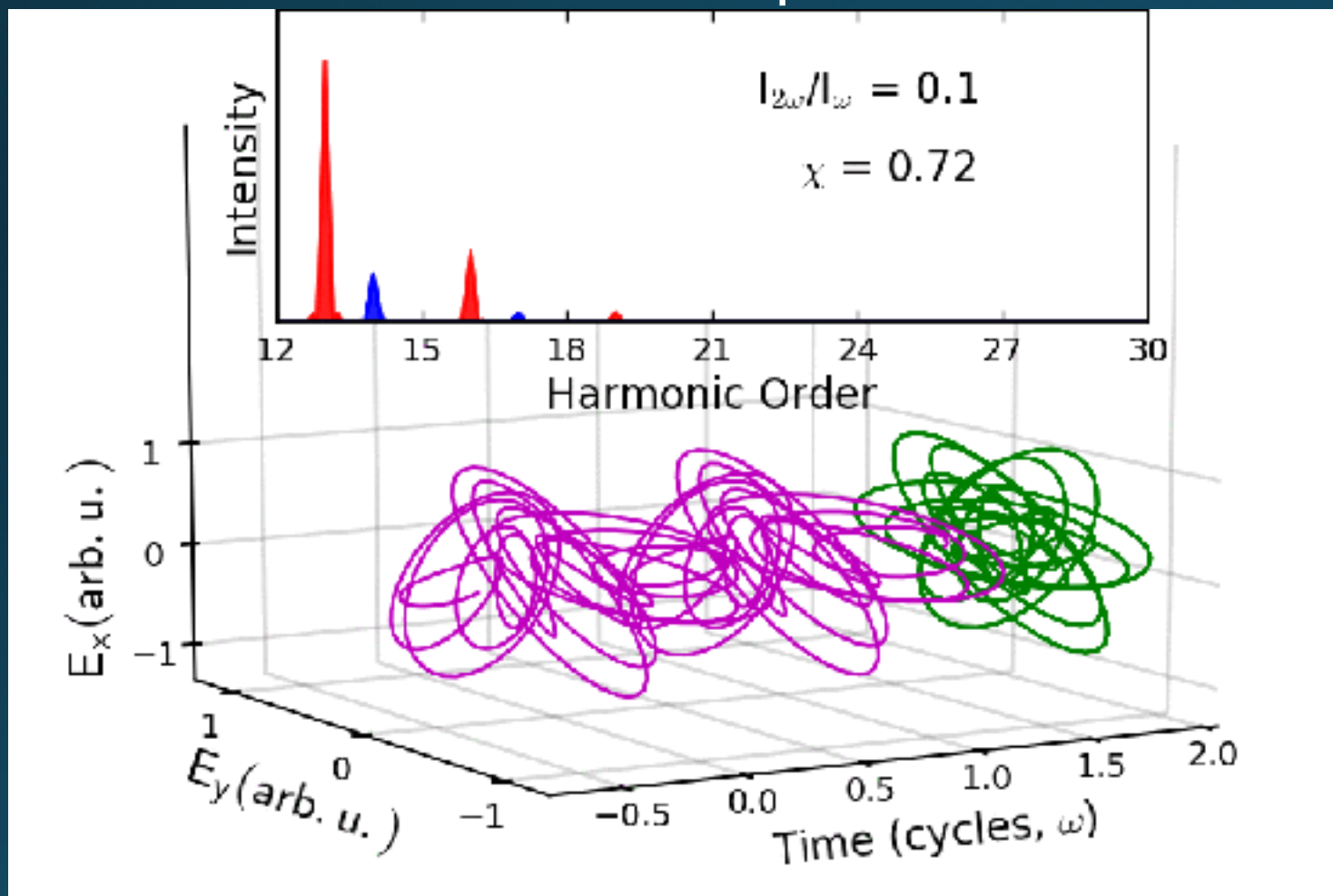
- First demonstration of real-time polarization control of attosecond pulse trains in CPHHG!



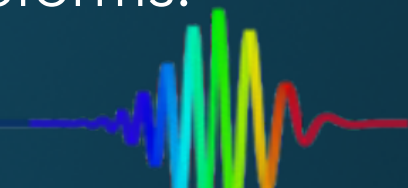
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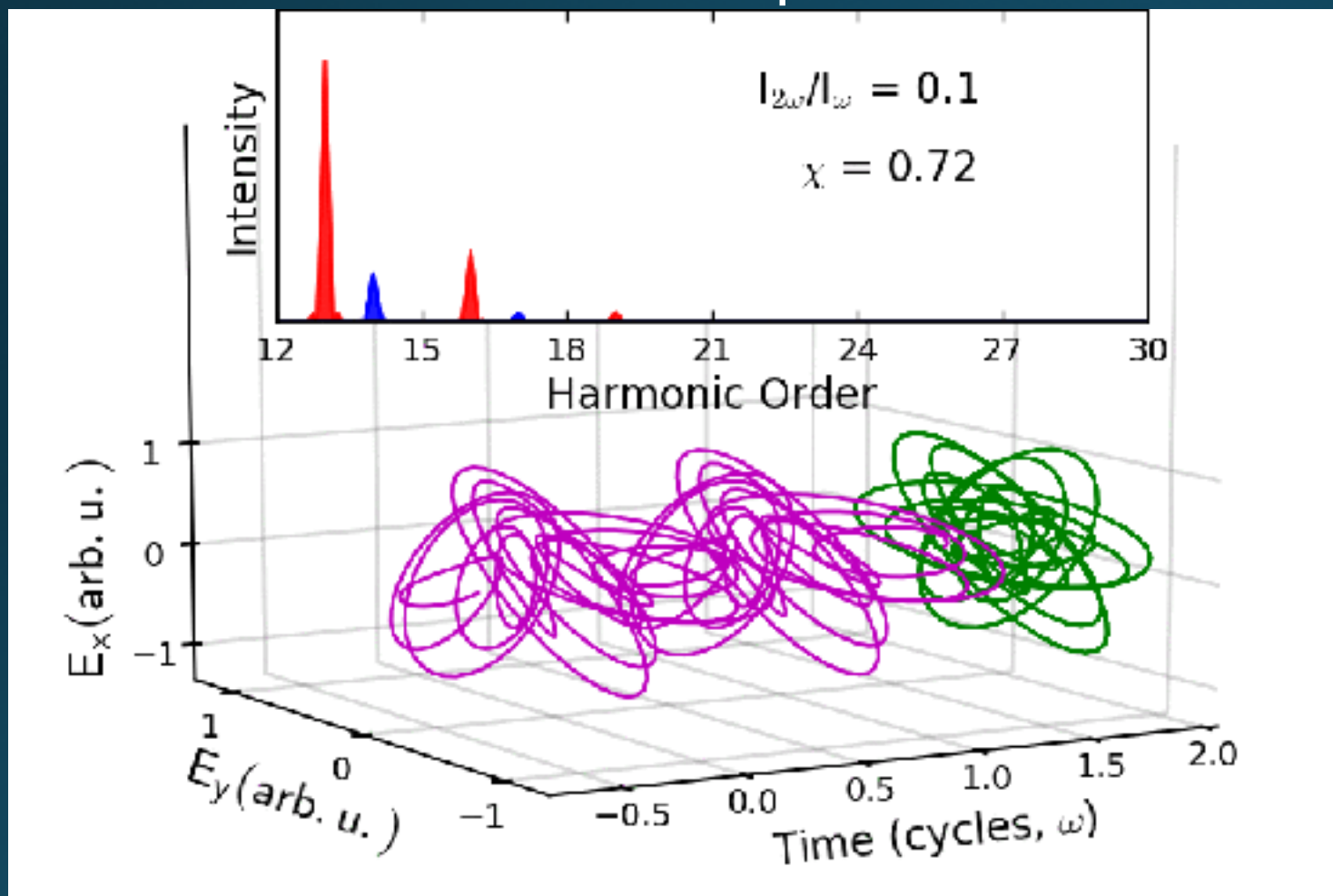
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Neufeild, et al., arXiv:1709.06261, 2017

Instantaneous Optical Chirality

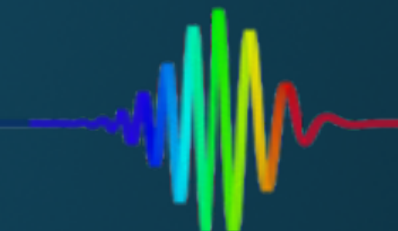
$$C_P = \frac{1}{c_0^2} \dot{\phi}(t) I(t)$$

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



Dorney et al., Phys. Rev. Lett., 119, 2017

Pisanty et al., Phys. Rev. A, 90, 2014

Li, et al., arXiv:1702.04084, 2017



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., 119, 2017

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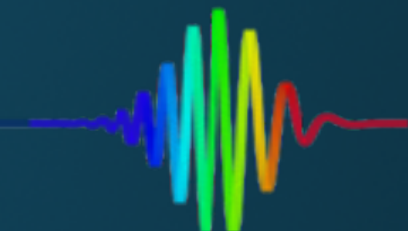
$$I_q \propto P(\Omega) \propto \sum_{i=0}^{\infty} p_1^{|n_1^i|} p_2^{|n_2^i|} \quad p_{\omega} = \frac{I_{\omega}}{I_{\omega} + I_{2\omega}} = \frac{1}{1 + I_{ratio}} \quad p_{2\omega} = \frac{I_{2\omega}}{I_{\omega} + I_{2\omega}} = \frac{1}{1 + 1/I_{ratio}}$$

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n = channel number

s = number of "emitted" photons

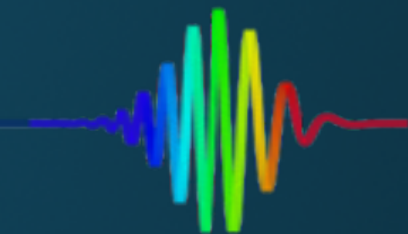
Channel	$H_{19, RCP} (7\omega_1 + 6\omega_2)$	
(n, s)	Total Photons	Statistical Scaling
(6, 0)	$7\omega_1 + 6\omega_2$	$p_{1\omega}$
(6, 1)	$7\omega_1 + 8\omega_2$	$p_{2\omega}$
(6, 2)	$9\omega_1 + 10\omega_2$	$p_{2\omega}$
(6, 3)	$11\omega_1 + 12\omega_2$	$p_{2\omega}$

Dorney et al., Phys. Rev. Lett., 119, 2017

Pisanty et al., Phys. Rev. A, 90, 2014

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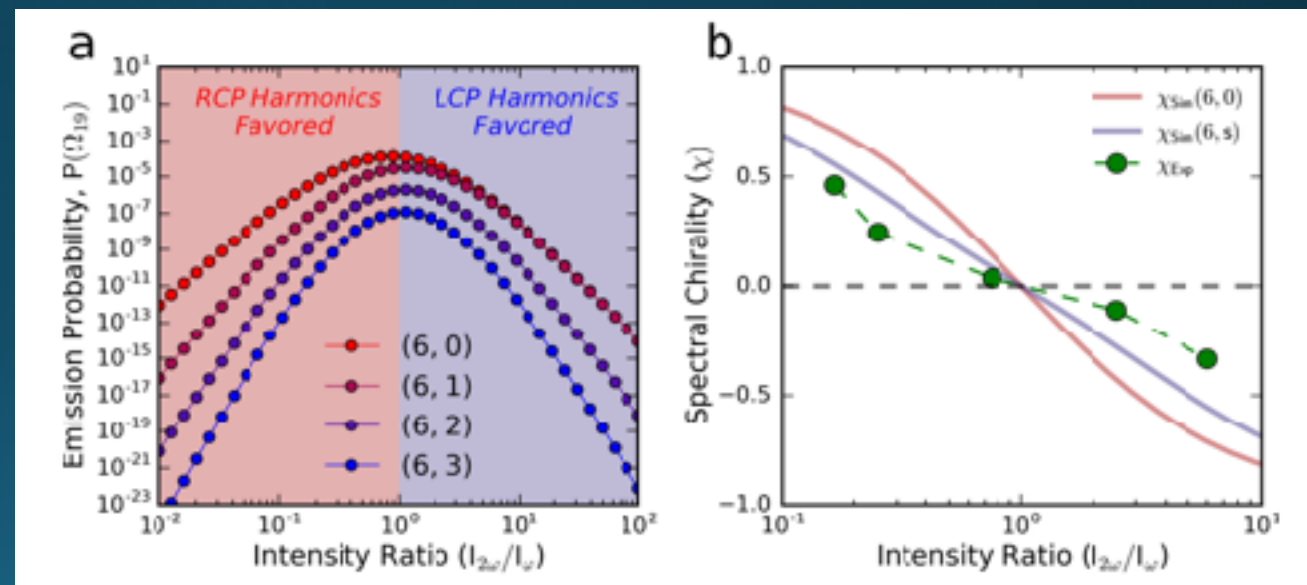


$$I_q \propto P(\Omega) \propto \sum_{i=0}^{\infty} p_1^{|n_1^i|} p_2^{|n_2^i|} \quad p_{\omega} = \frac{I_{\omega}}{I_{\omega} + I_{2\omega}} = \frac{1}{1 + I_{ratio}} \quad p_{2\omega} = \frac{I_{2\omega}}{I_{\omega} + I_{2\omega}} = \frac{1}{1 + 1/I_{ratio}}$$

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Dorney et al., Phys. Rev. Lett., 119, 2017

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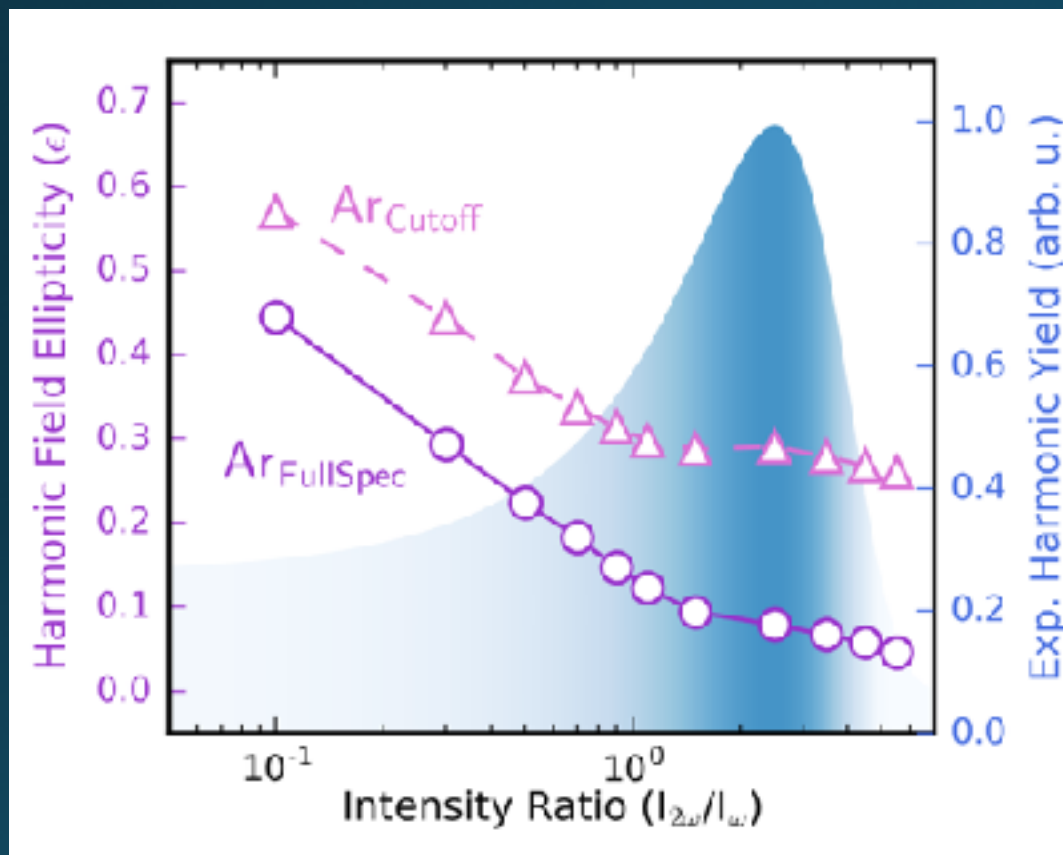
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Attosecond Polarization Control at a Price...



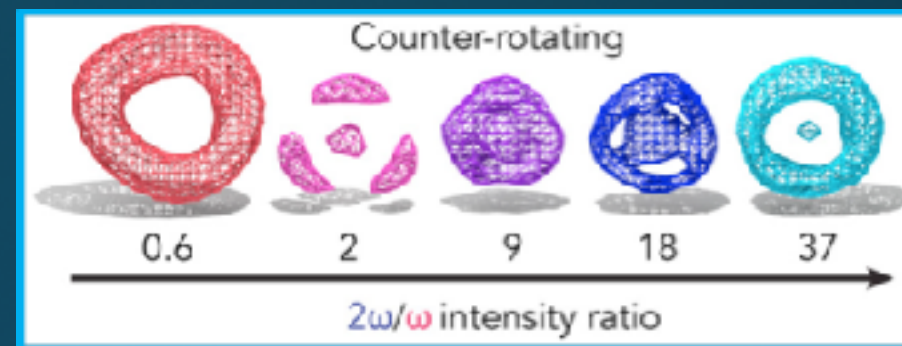
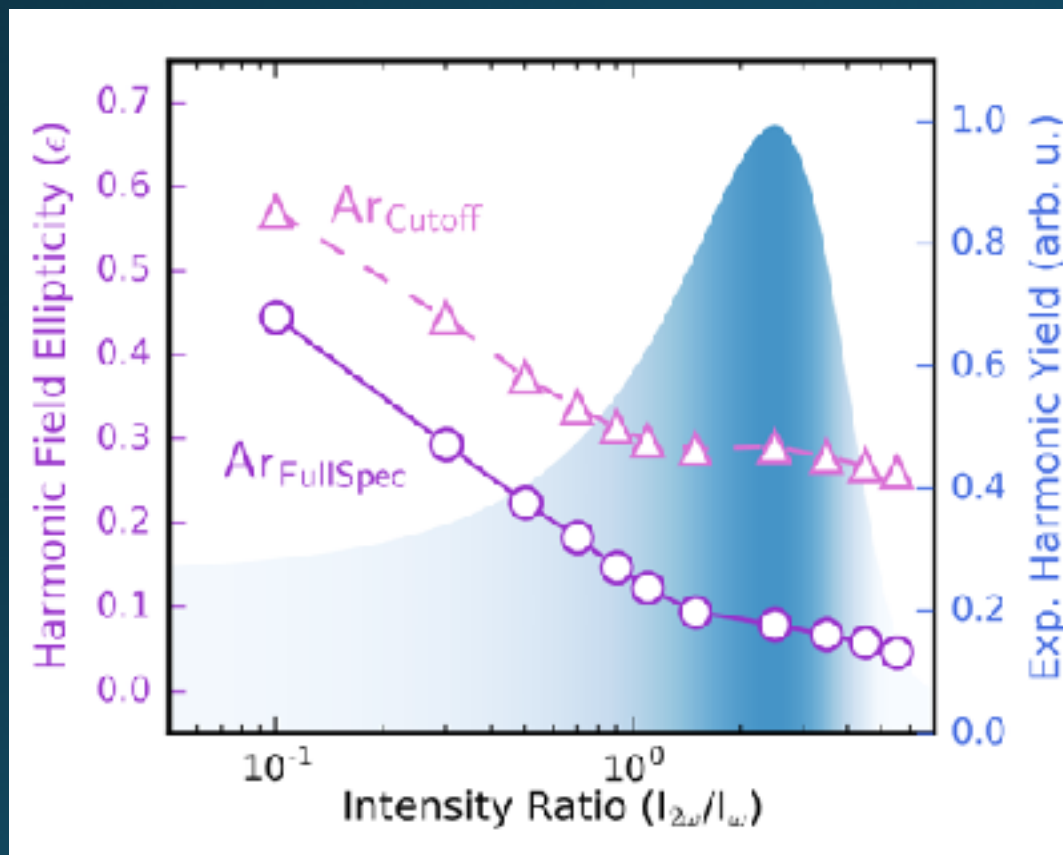
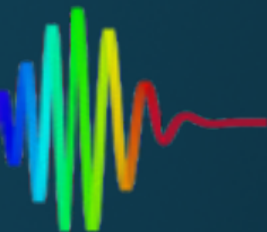
Mancuso, Phys. Rev. A., 93, 2016
Dorney et. al., Phys. Rev. Lett., 119, 2017



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Dorney et. al., Phys. Rev. Lett., 119, 2017

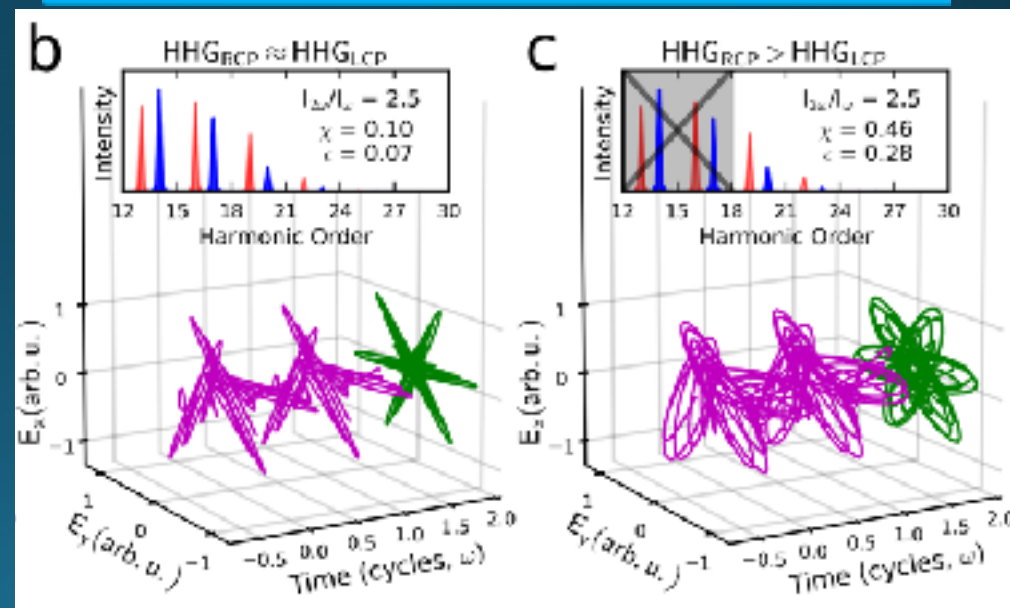
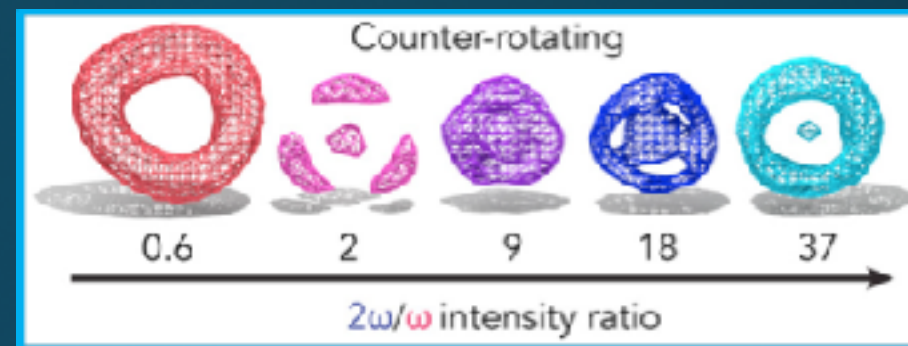
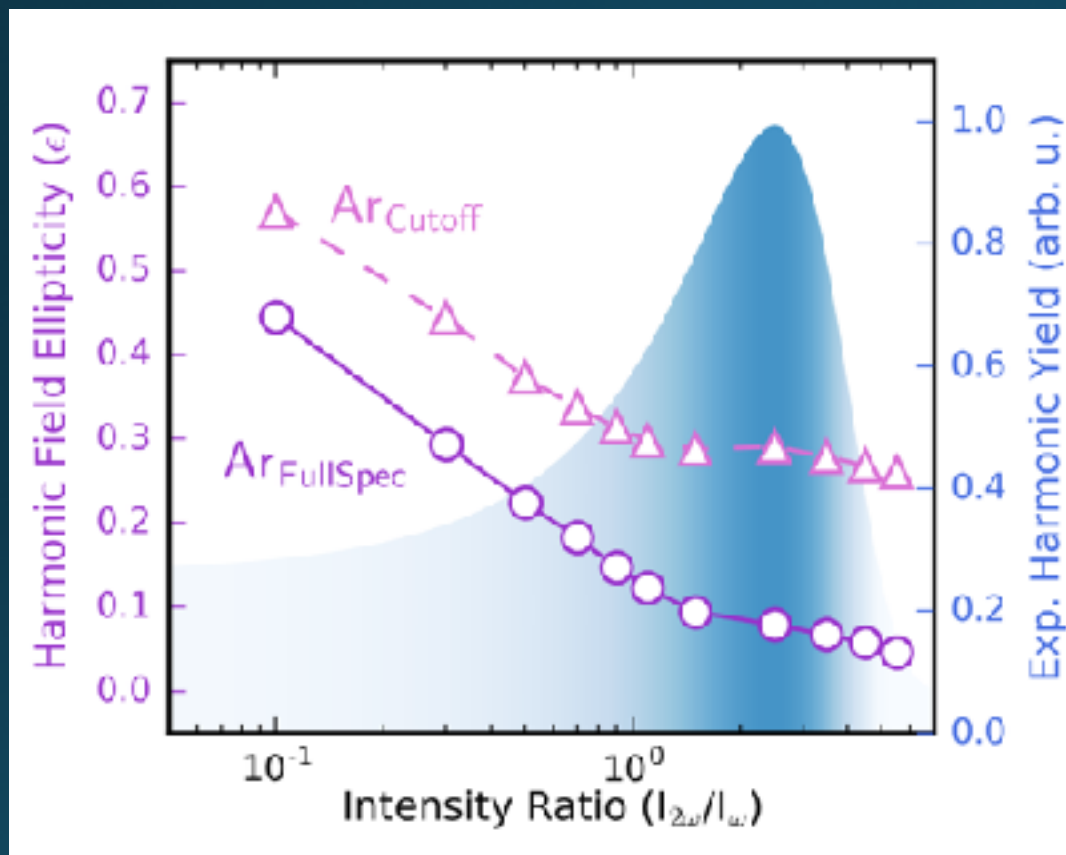
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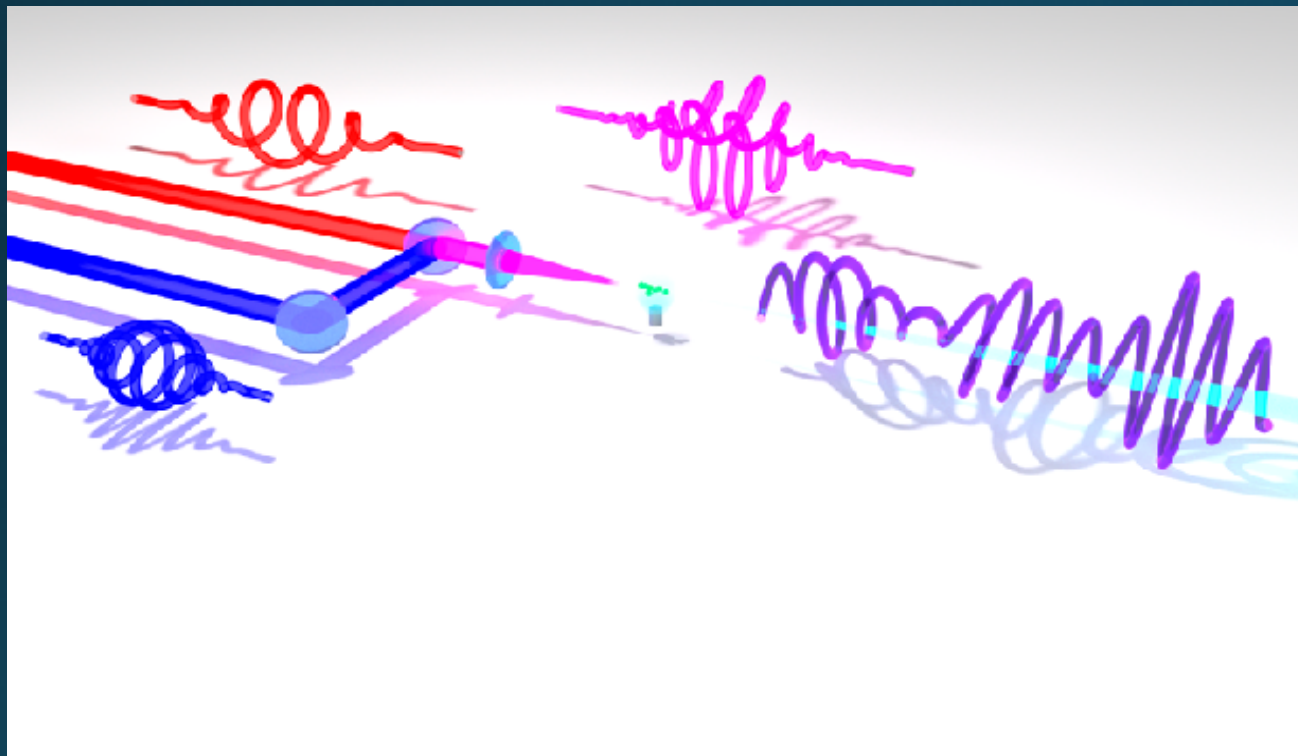
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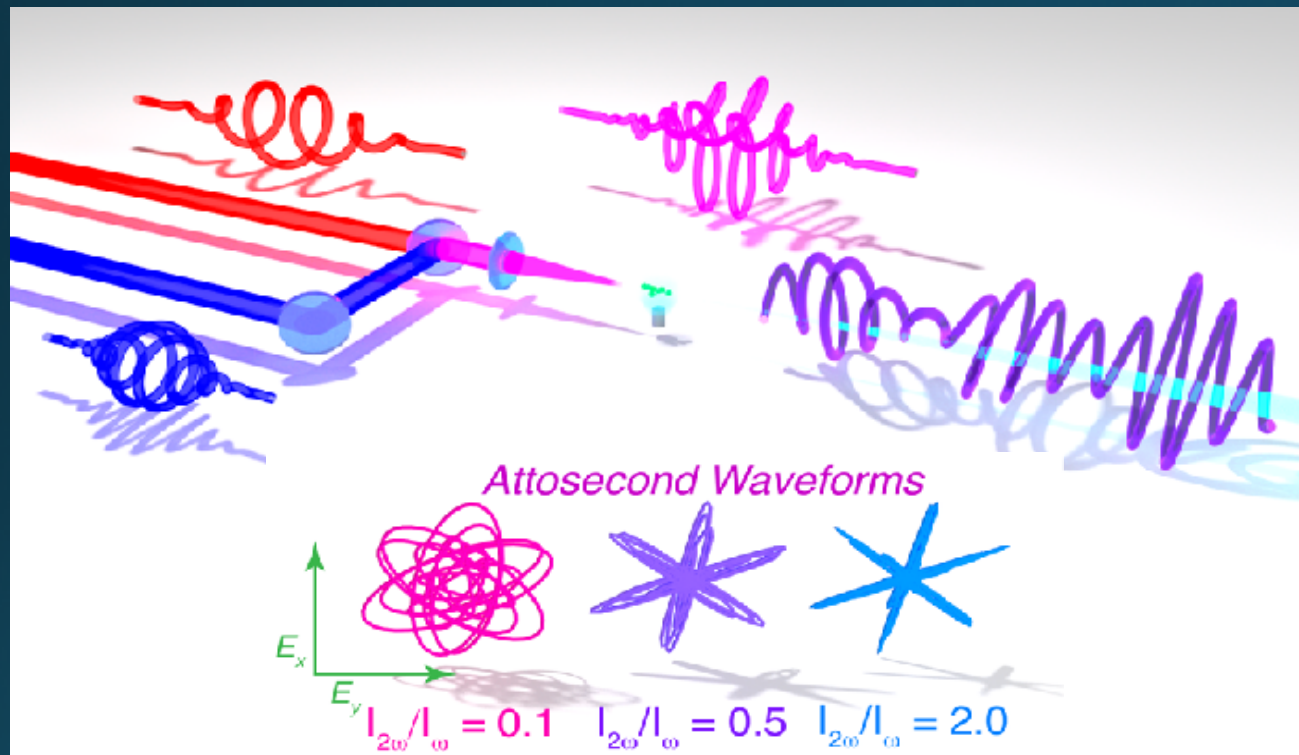
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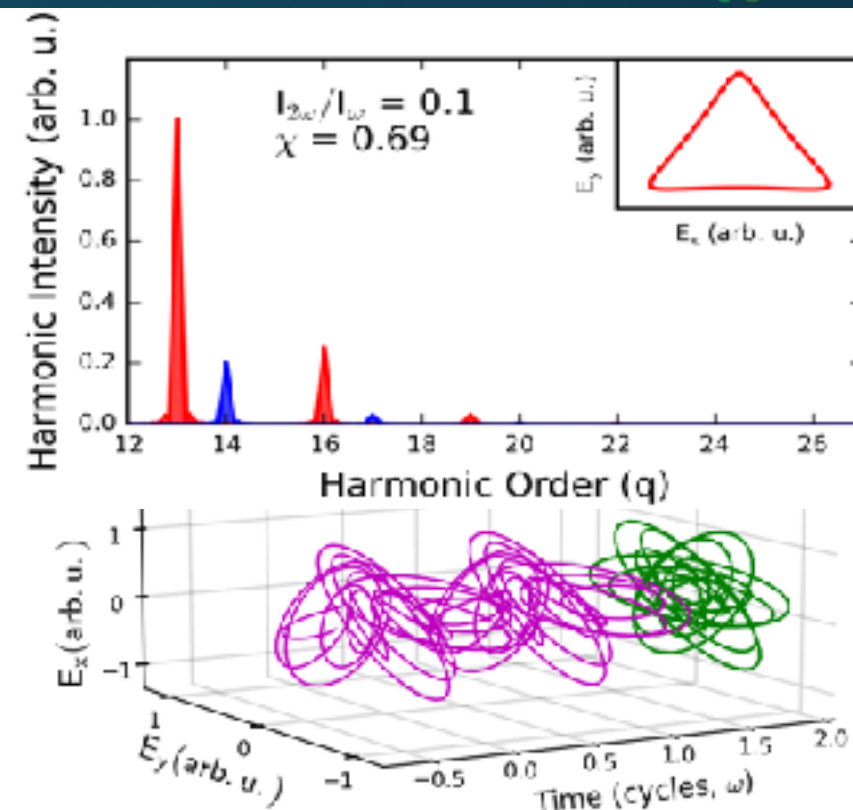
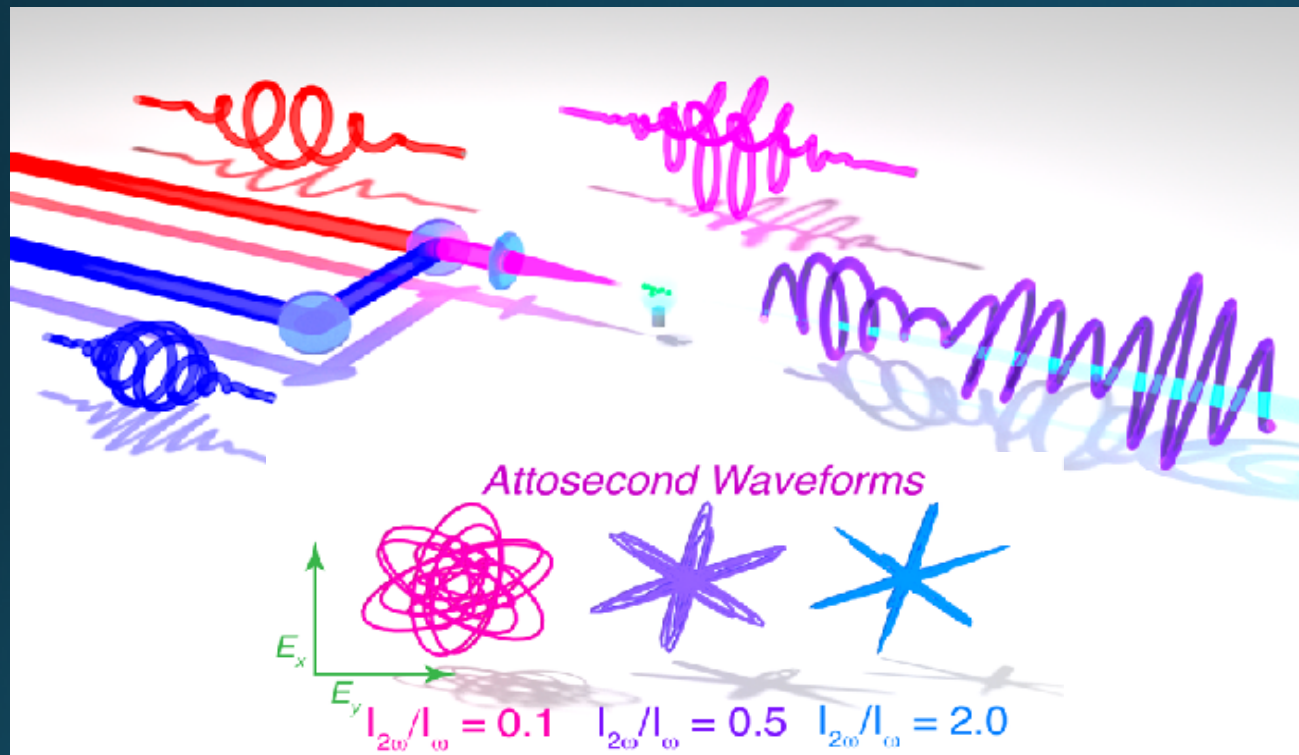
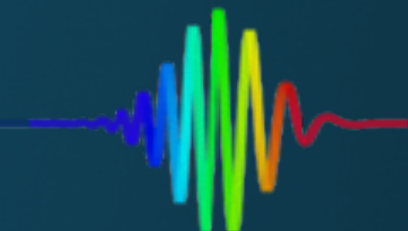
Dorney et. al., Phys. Rev. Lett., 118, 2017

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

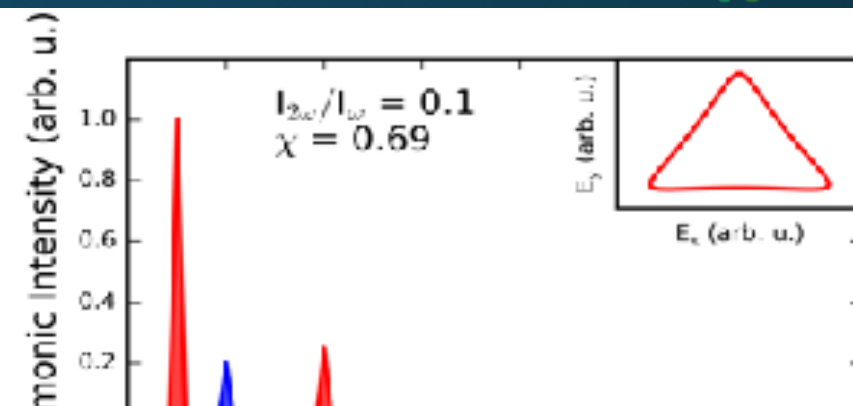
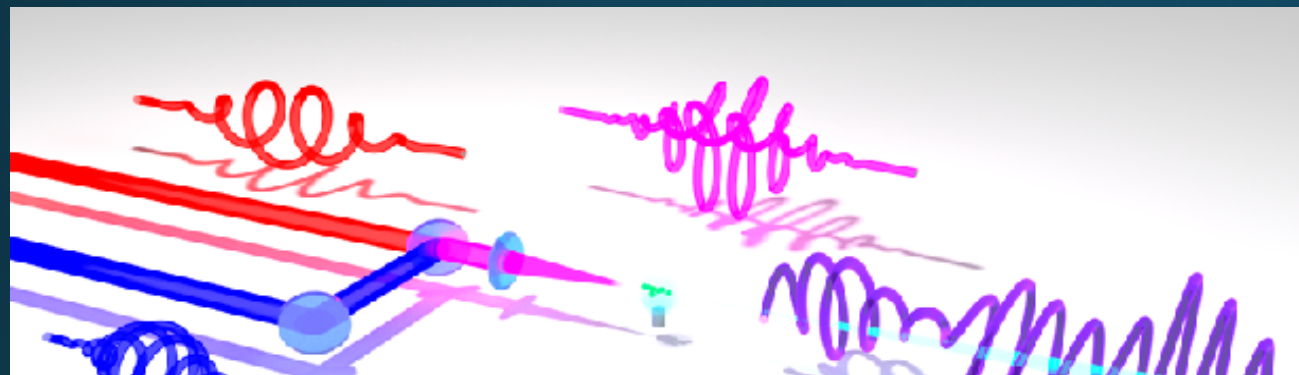


Dorney et. al., Phys. Rev. Lett., 118, 2017

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PRL 119, 063201 (2017)

PHYSICAL REVIEW LETTERS

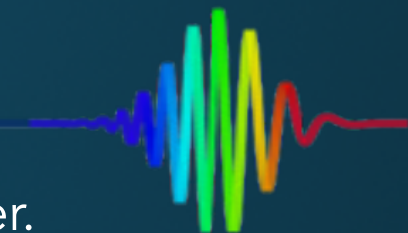
week ending
11 AUGUST 2017

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

Kevin M. Dorney,^{1,*} Jennifer L. Ellis,¹ Carlos Hernández-García,² Daniel D. Hickstein,¹ Christopher A. Mancuso,¹ Nathan Brooks,¹ Tingting Fan,¹ Guangyu Fan,³ Dmitriy Zusin,¹ Christian Gentry,¹ Patrik Grychtol,¹ Henry C. Kapteyn,¹ and Margaret M. Murnane¹



CPHHG and Its Younger Sibling: Bicircular High-Harmonic Spectroscopy (BHHS)!

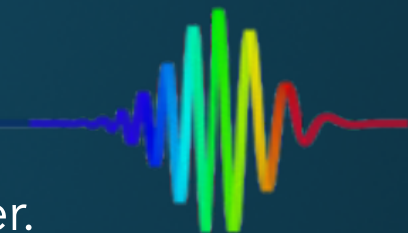


- CPHHG vs BHHS; two sides of the same coin, yet often far removed from each other.

Baykusheva, PRL, 116, 2016

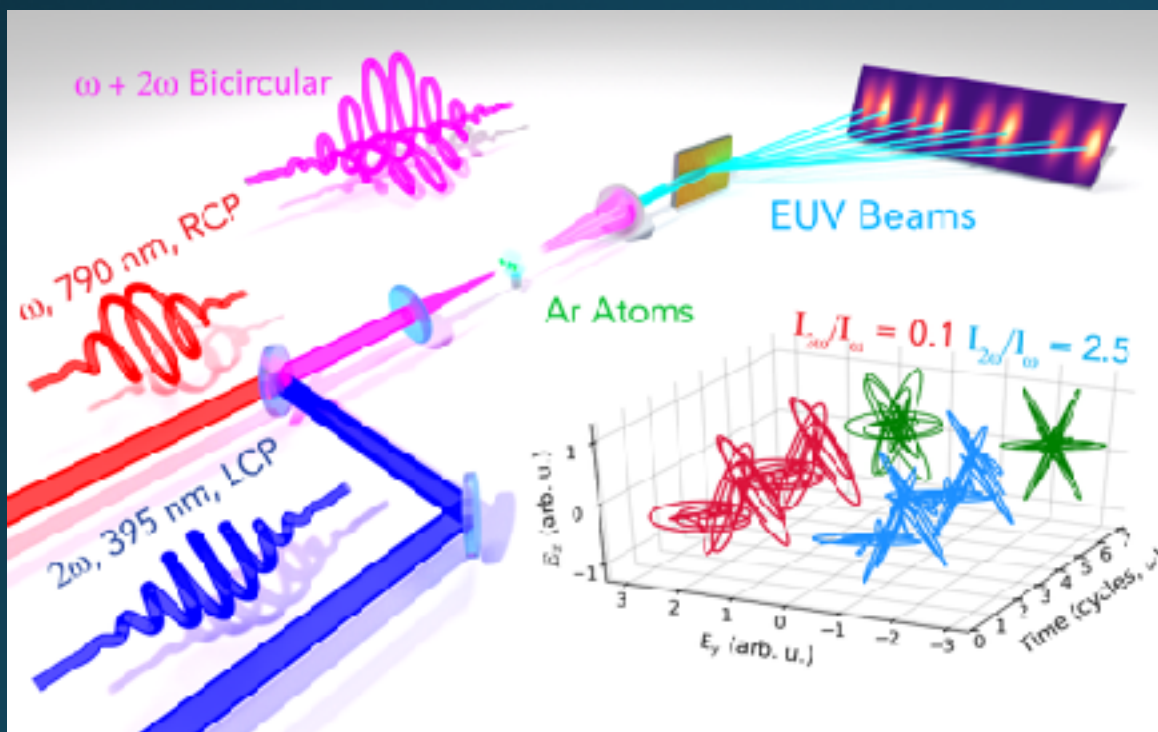
Dorney, PRL 119, 2017

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CPHHG: EUV Light Pulses for Spectroscopy



Baykusheva, PRL, 116, 2016

Dorney, PRL 119, 2017

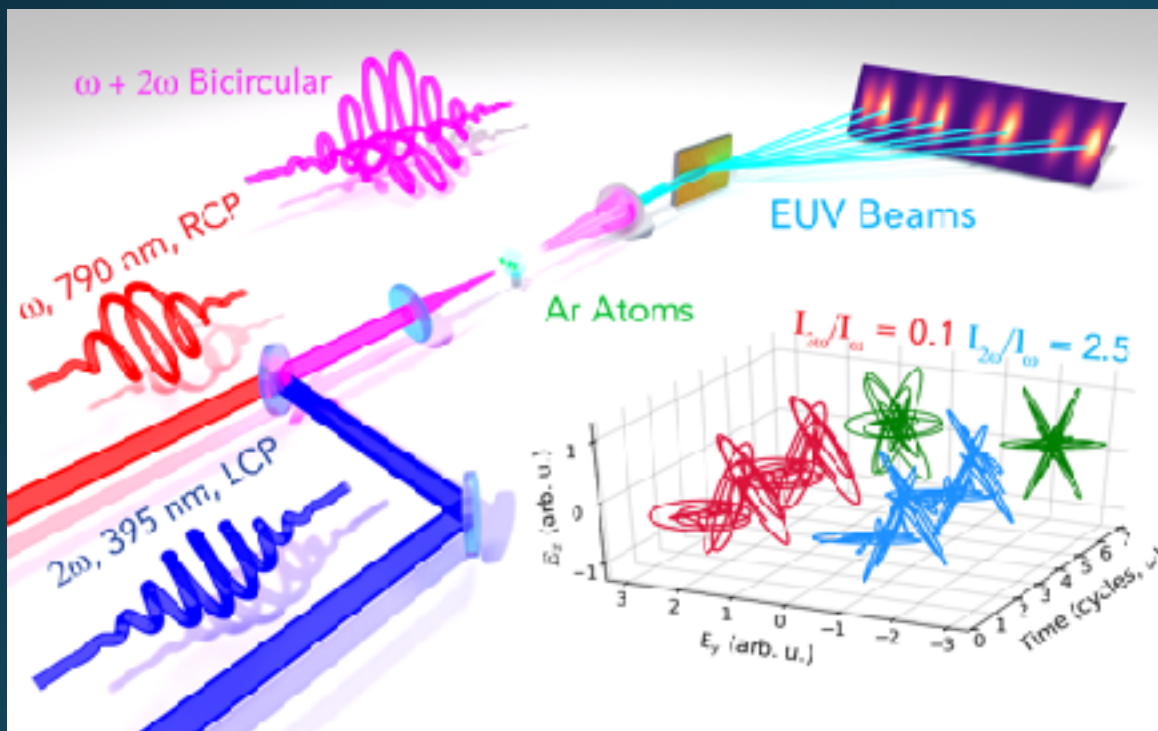
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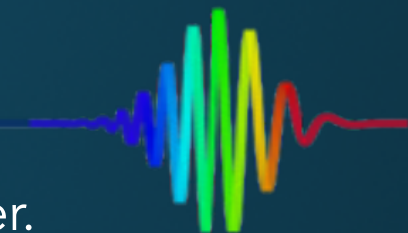
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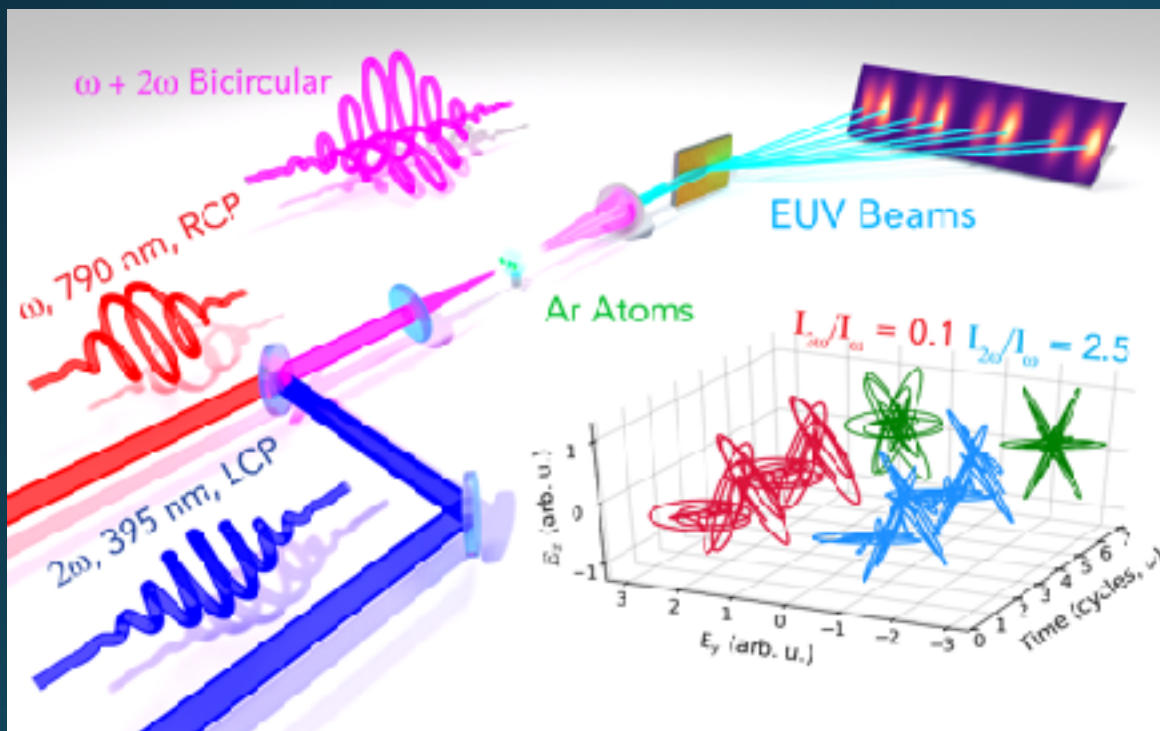
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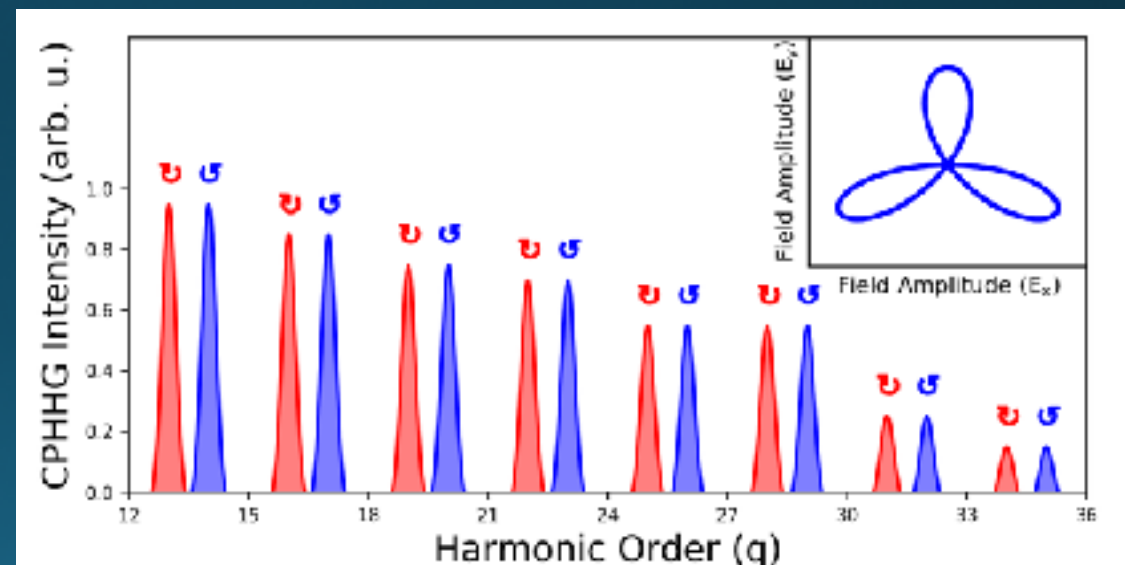


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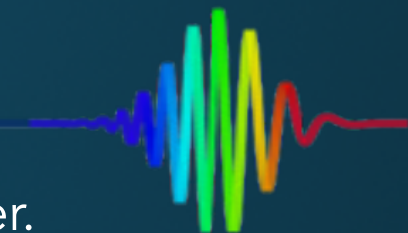
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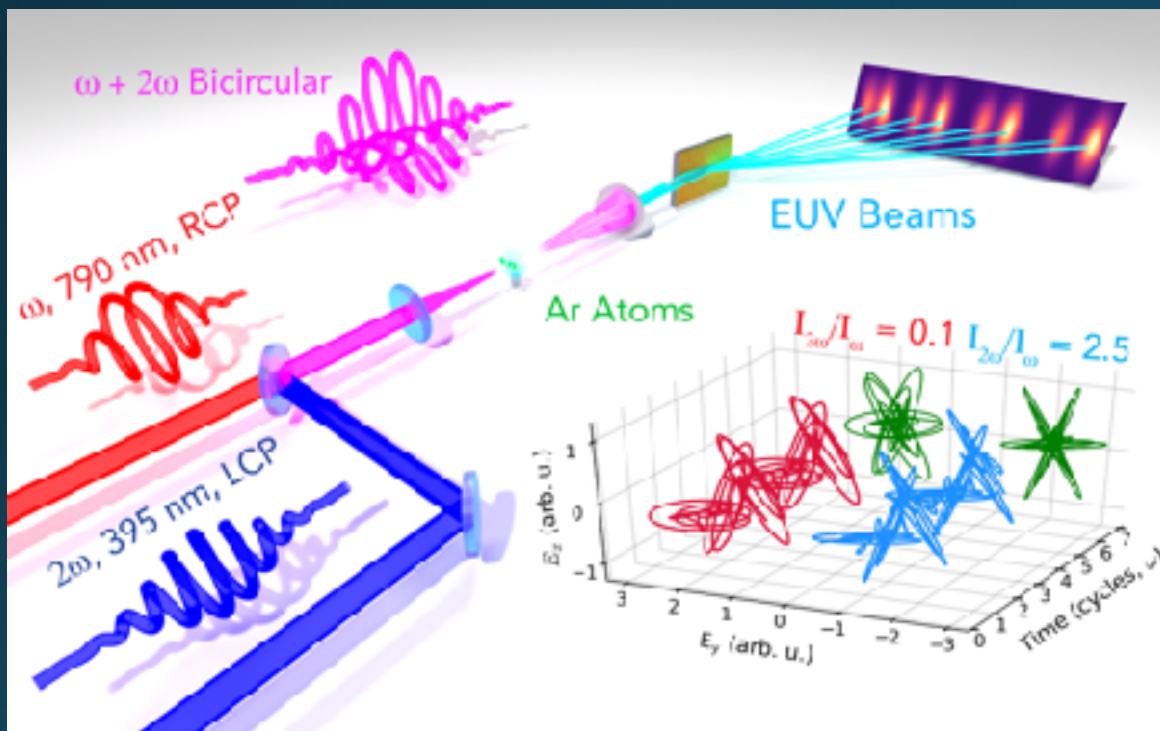


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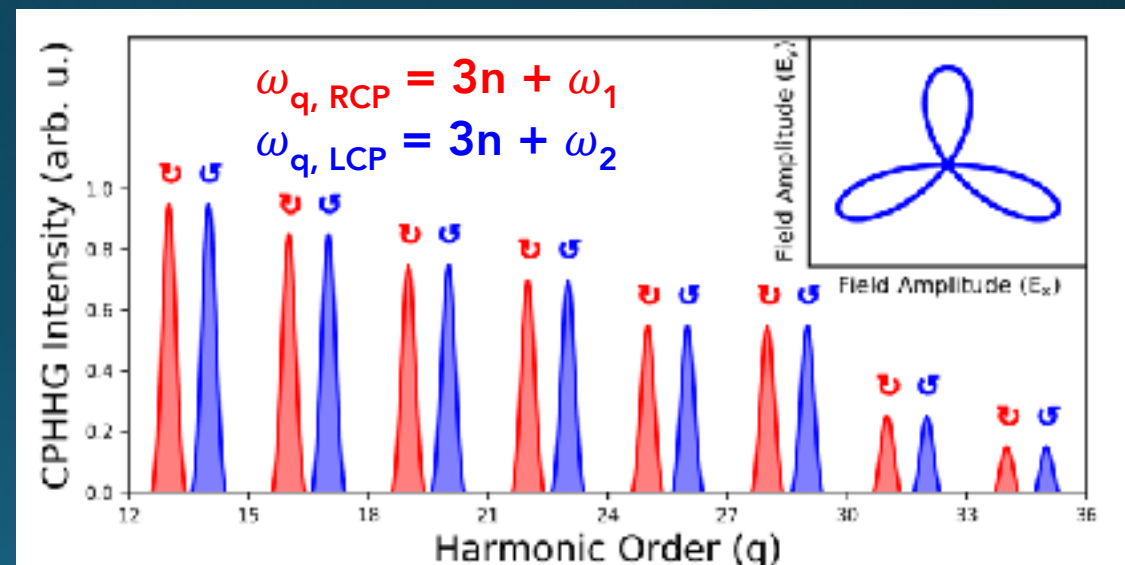


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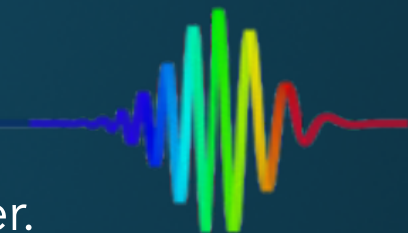
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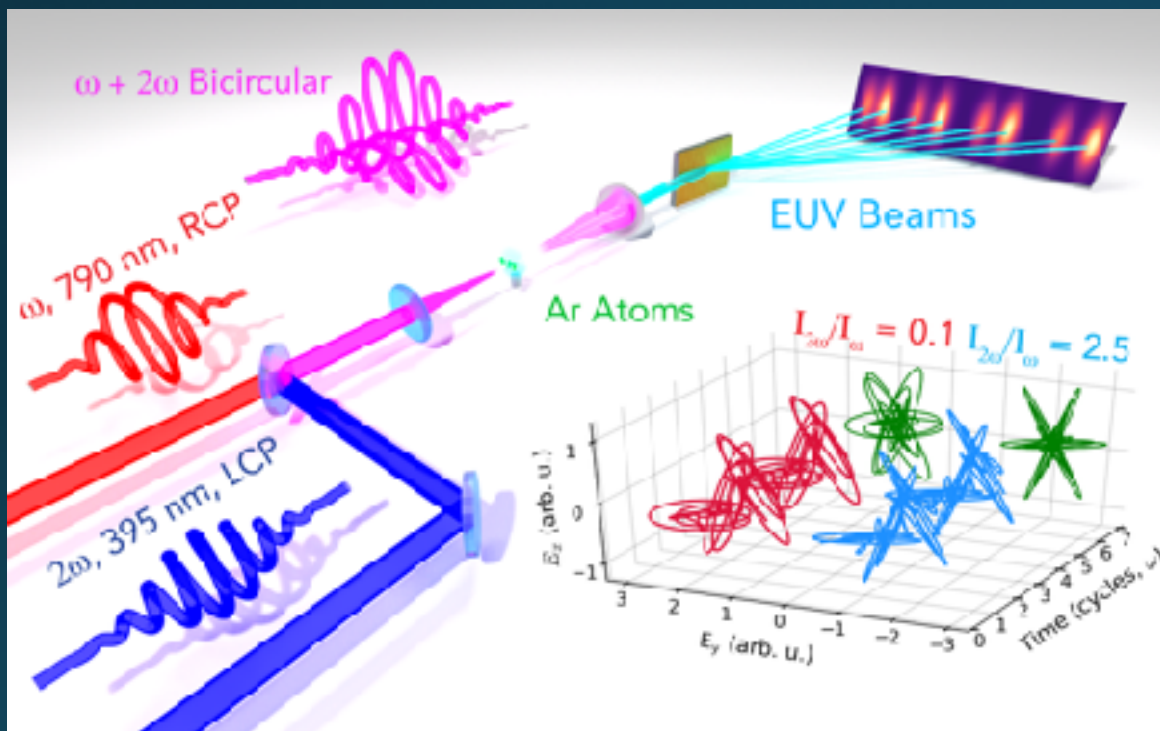
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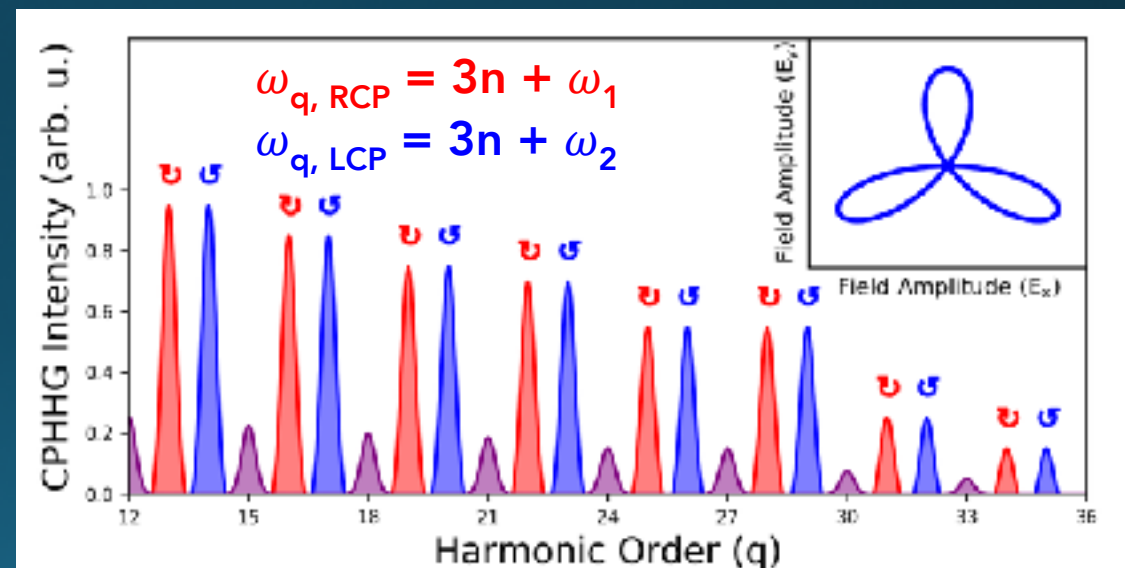


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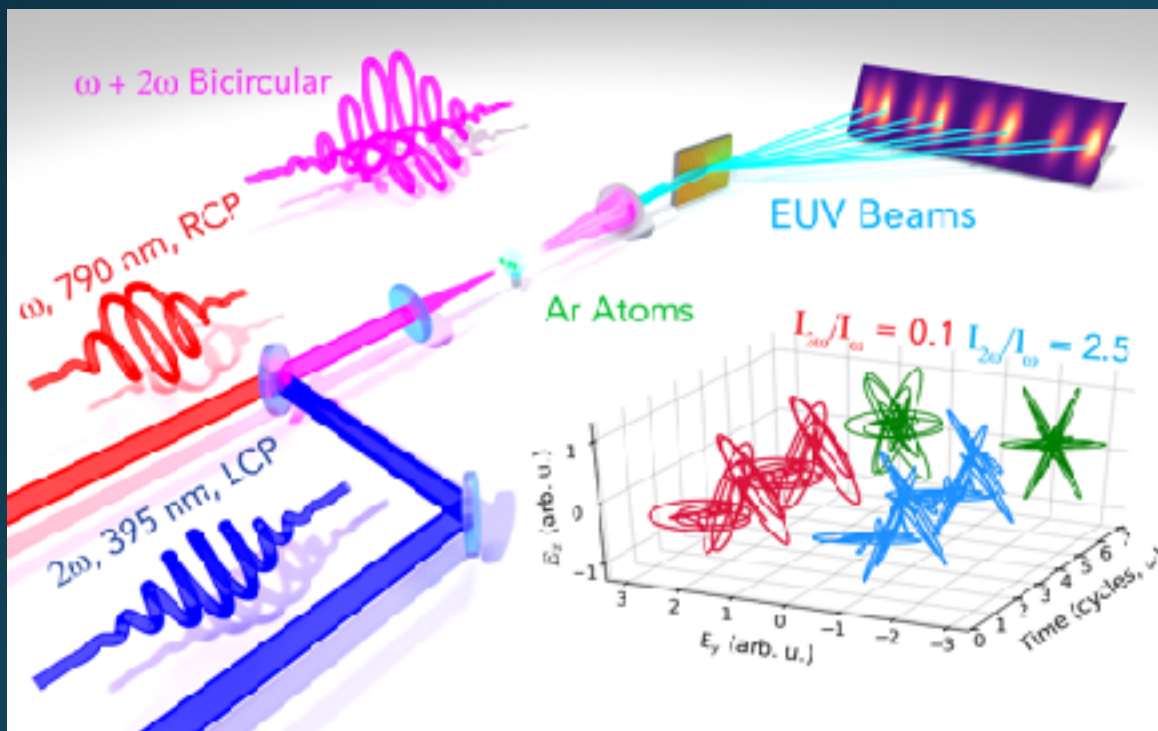
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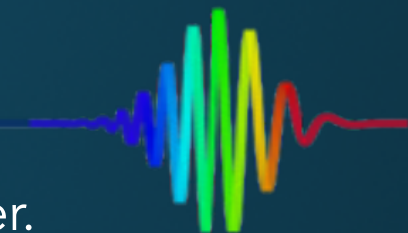
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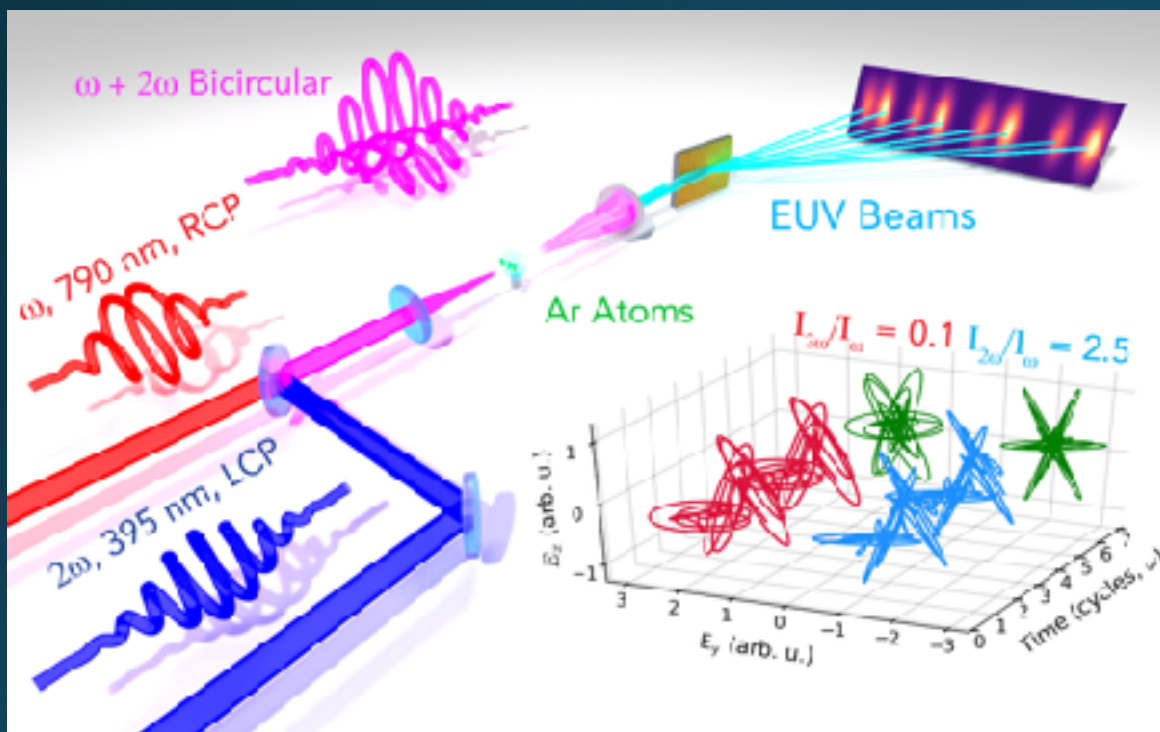
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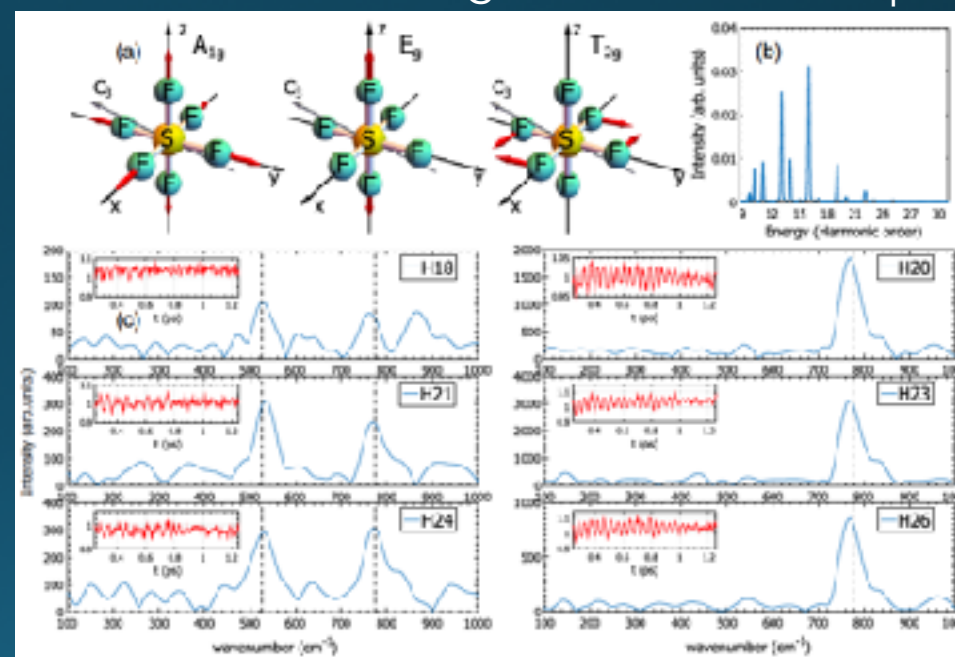


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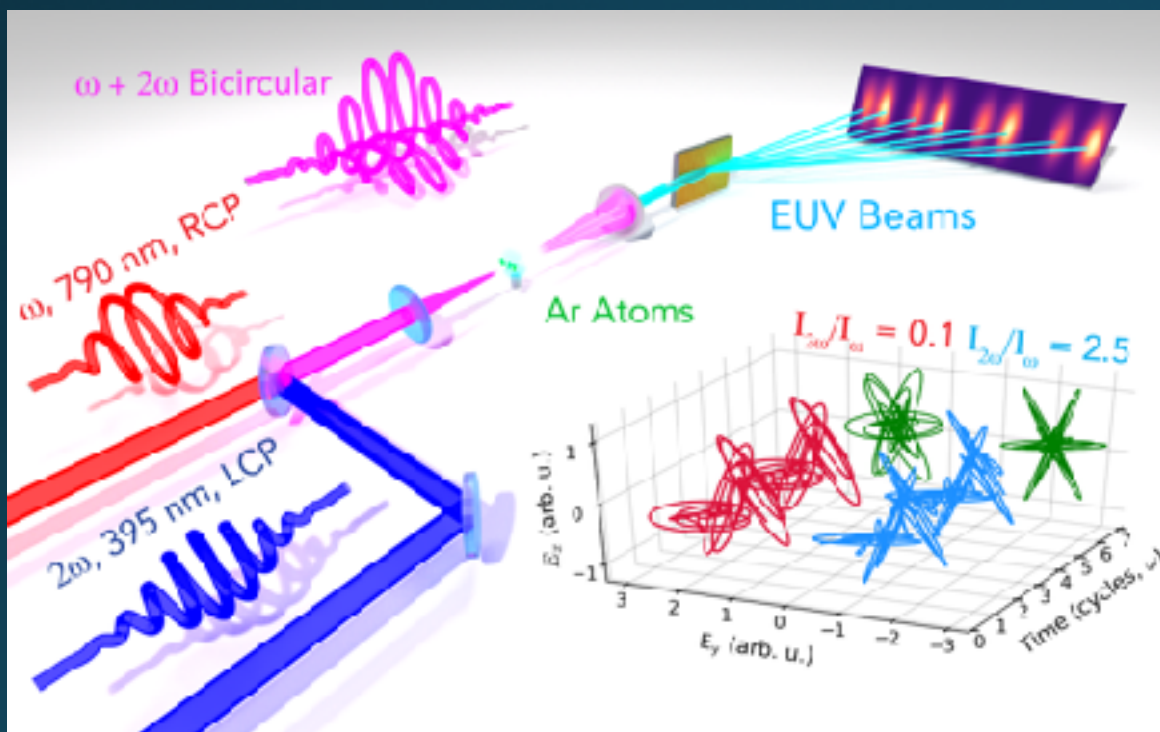
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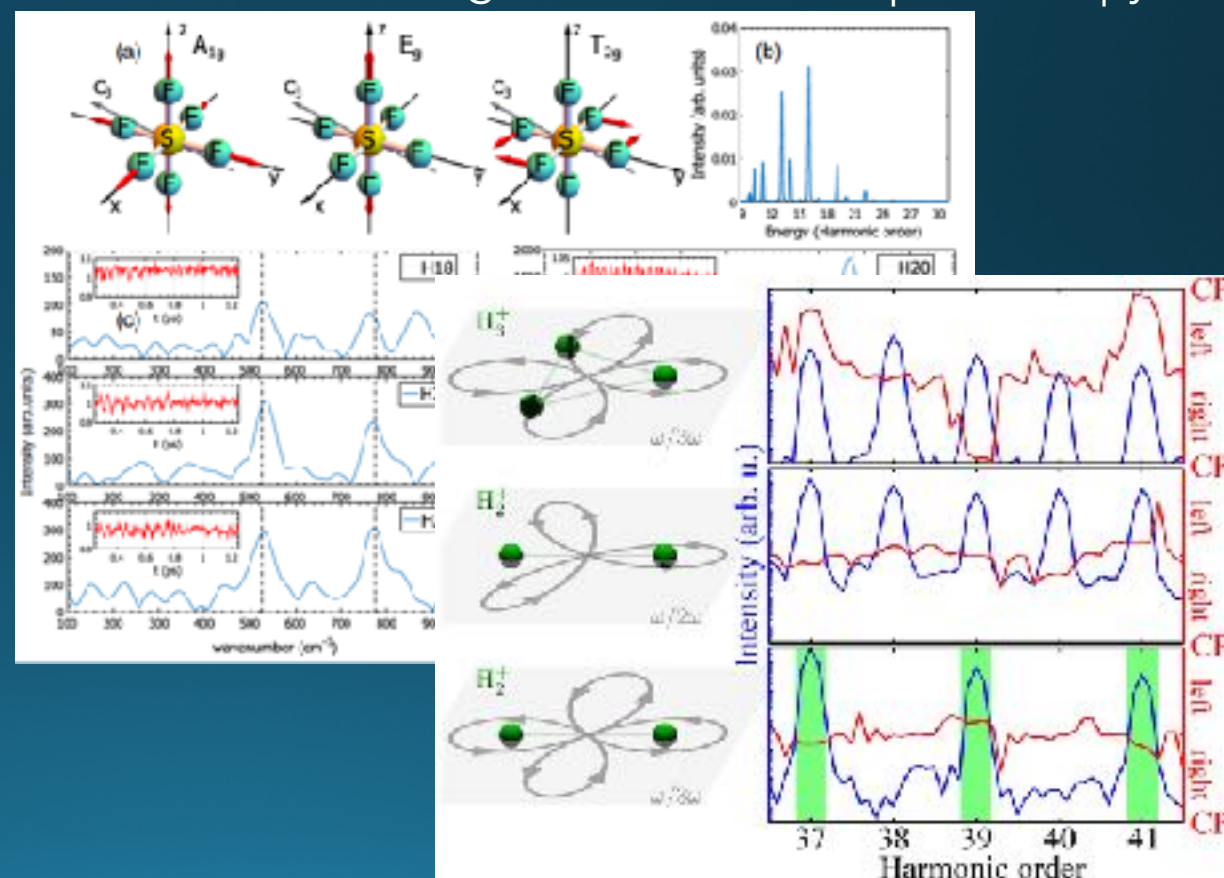
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Bicircular High-Harmonic Spectroscopy: A Dramatic Response to Electronic Structure

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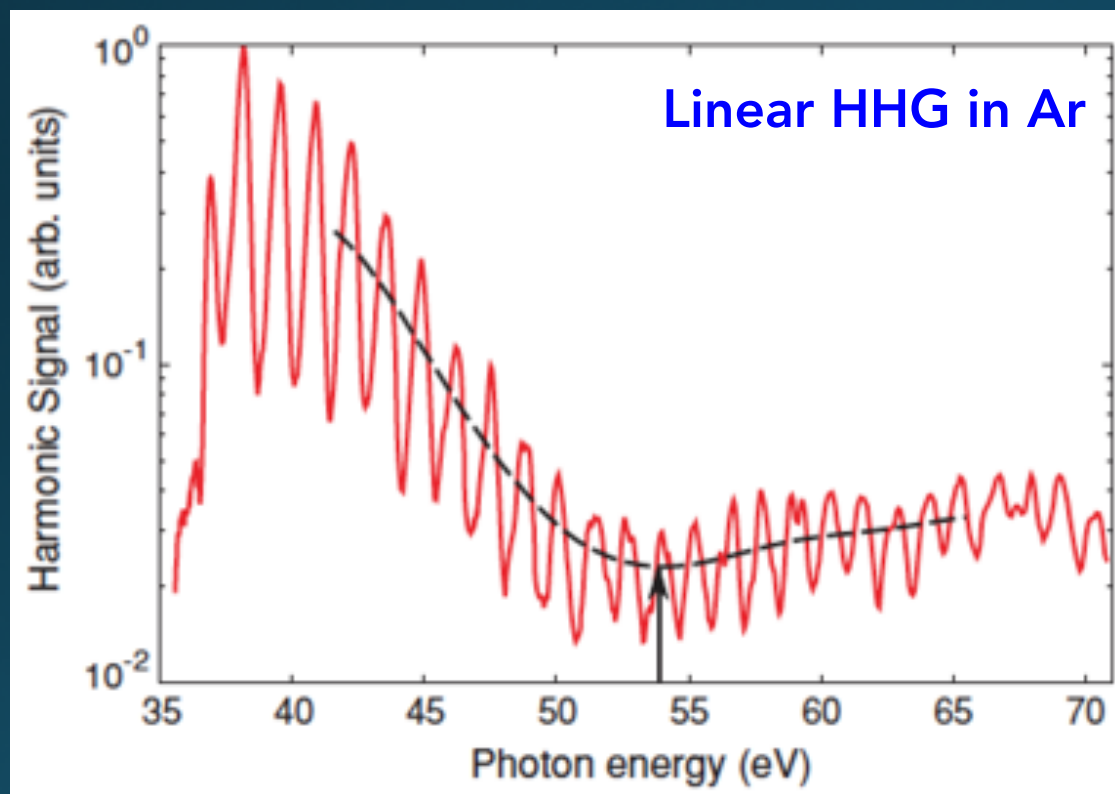


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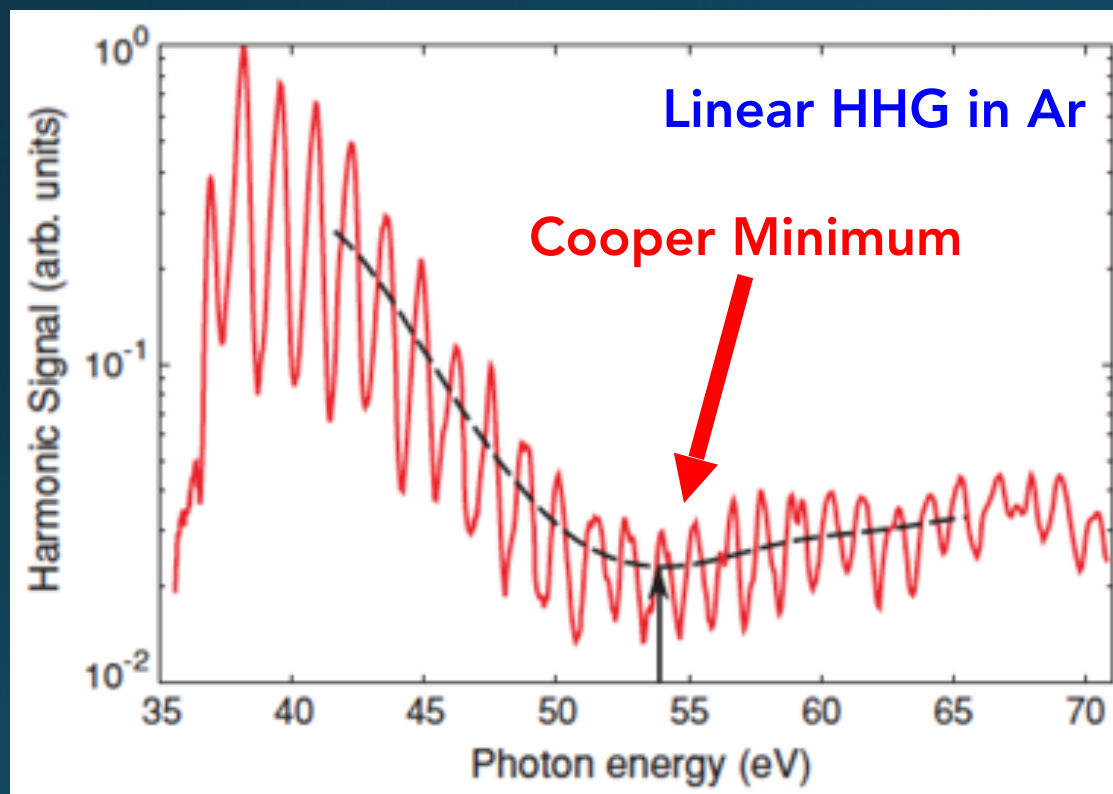
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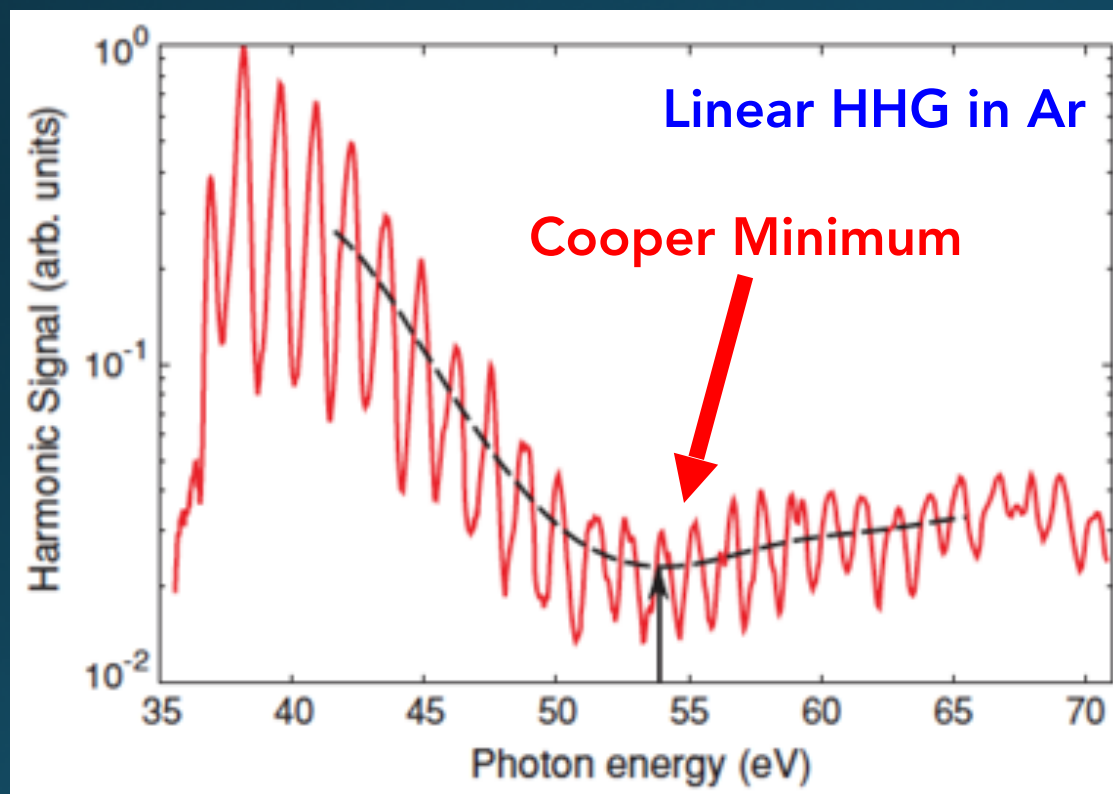


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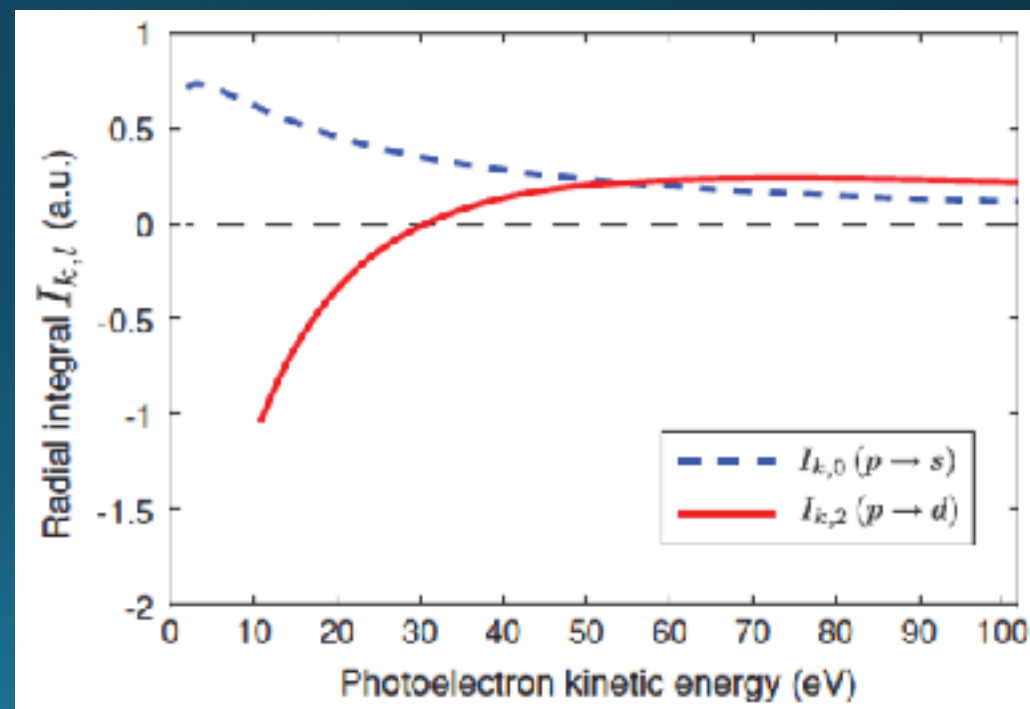
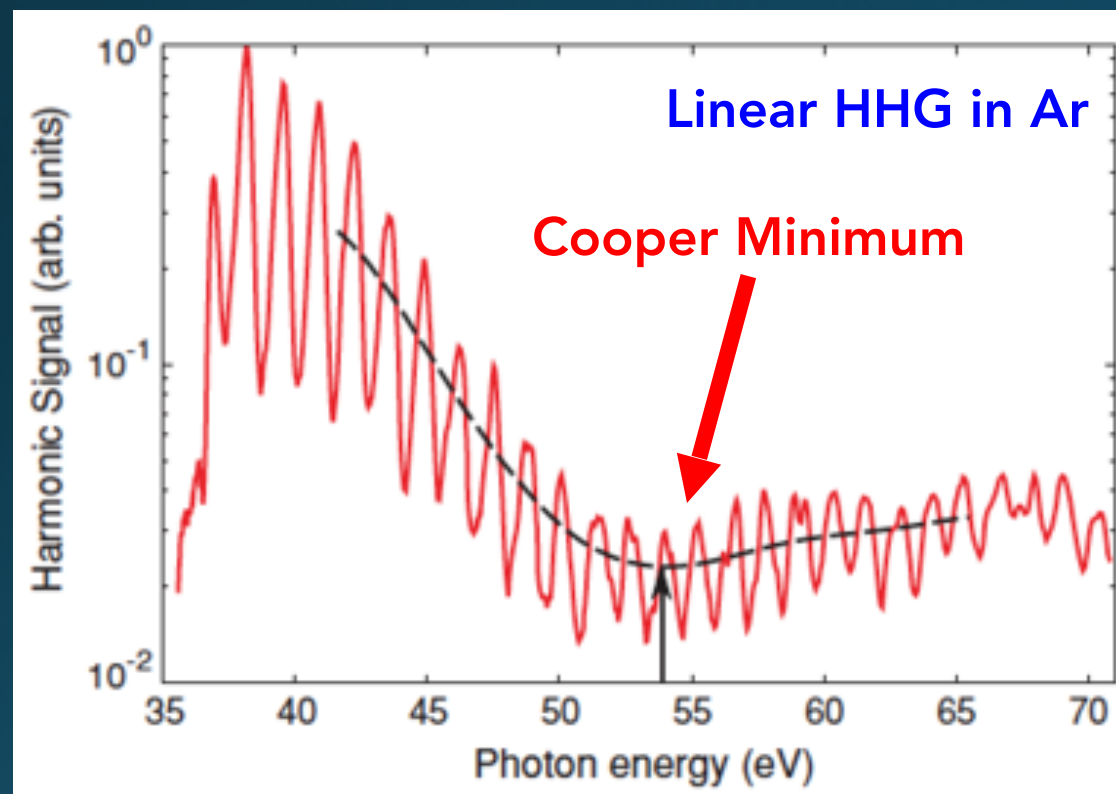


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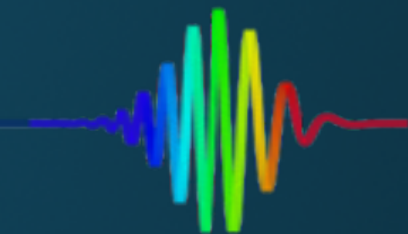
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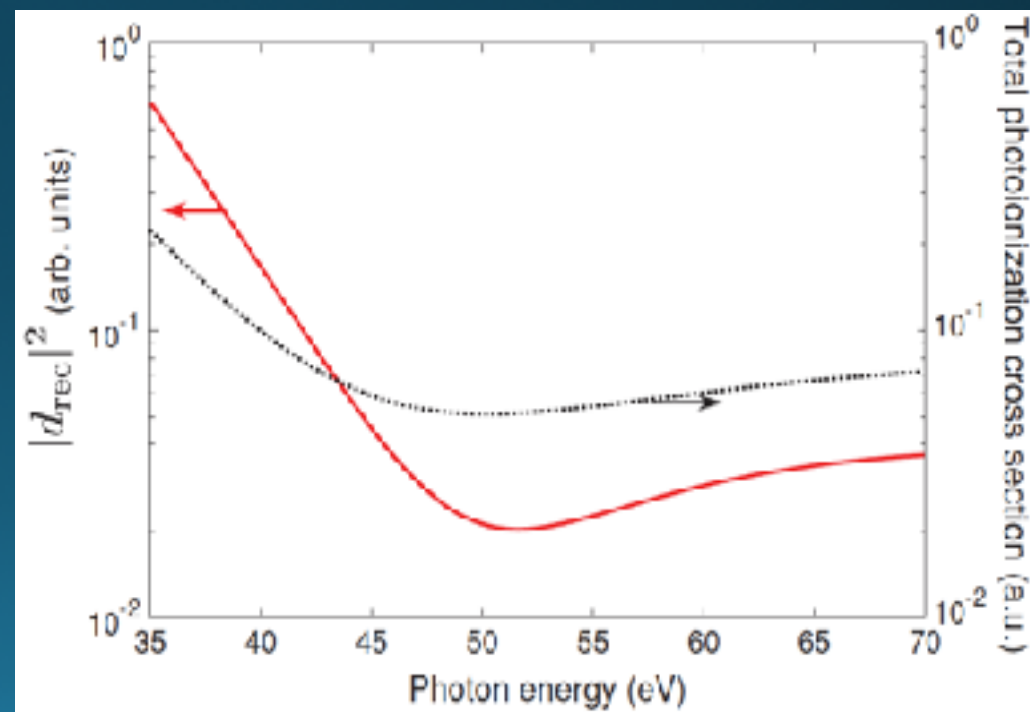
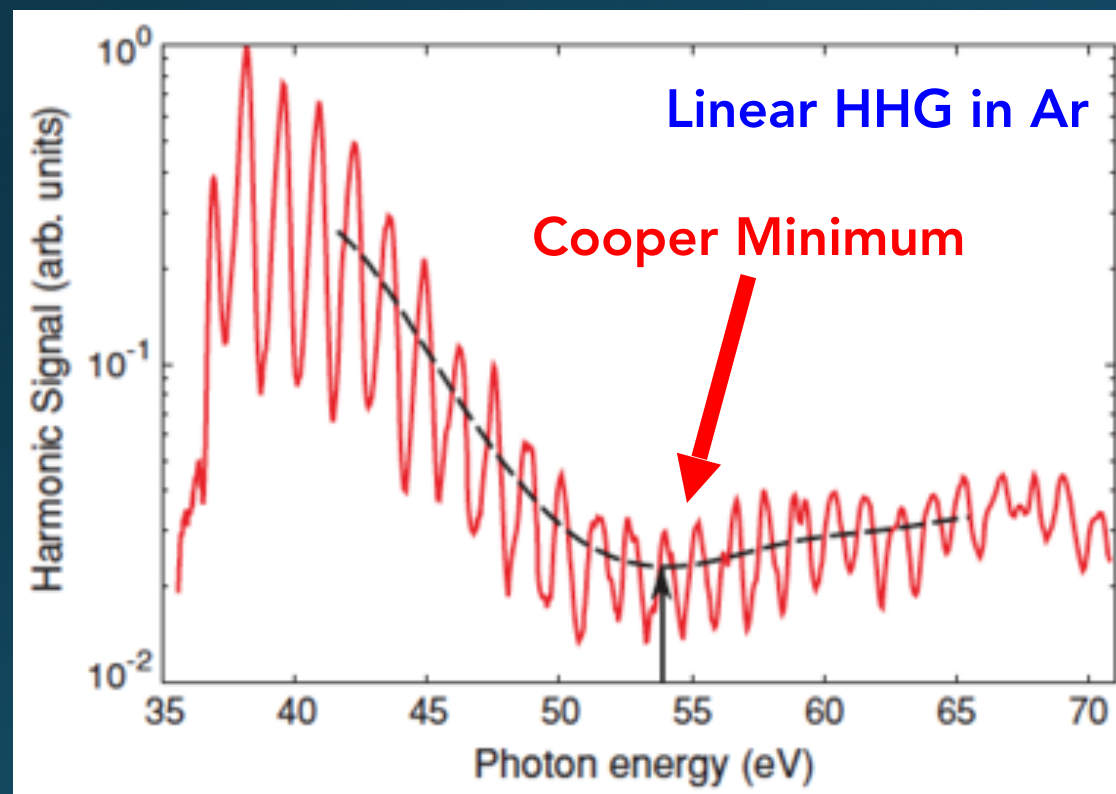


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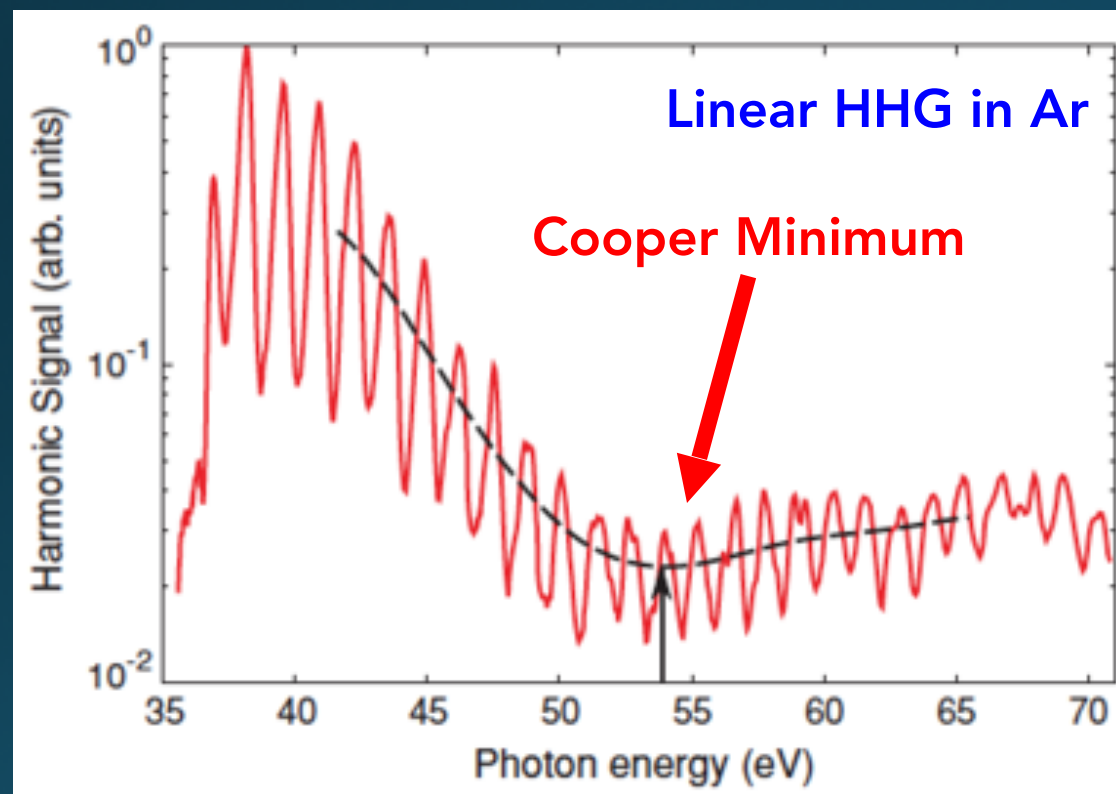


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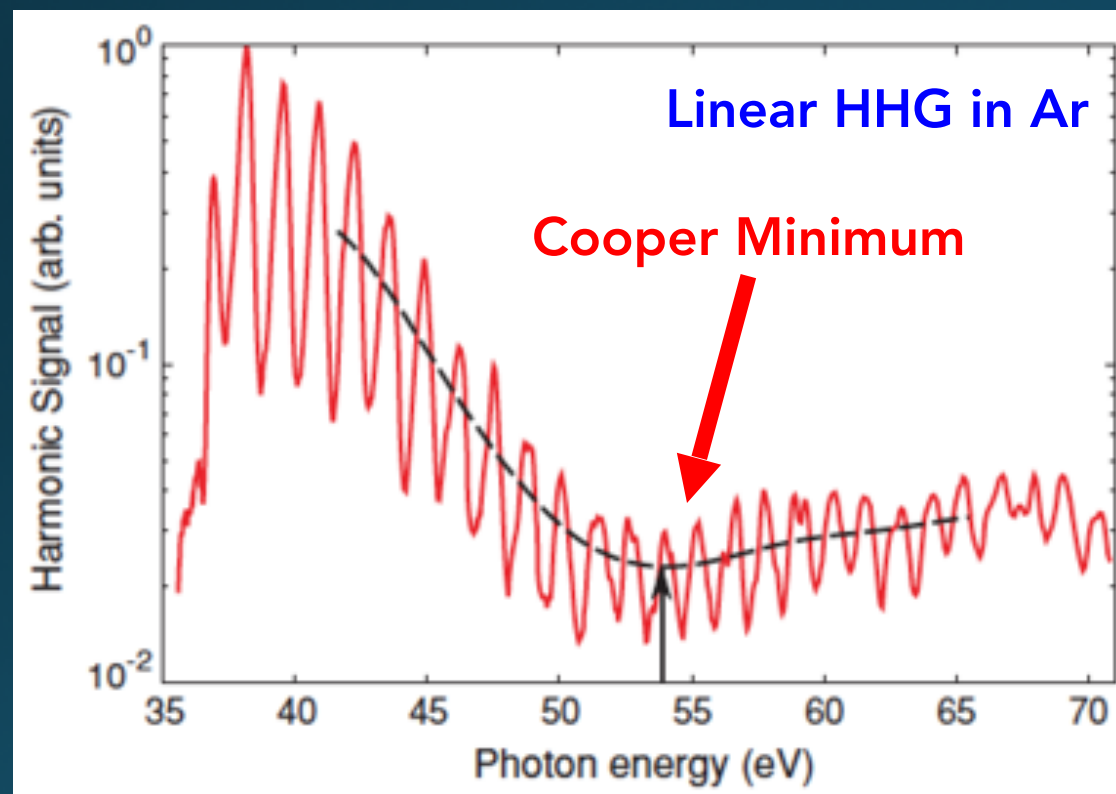
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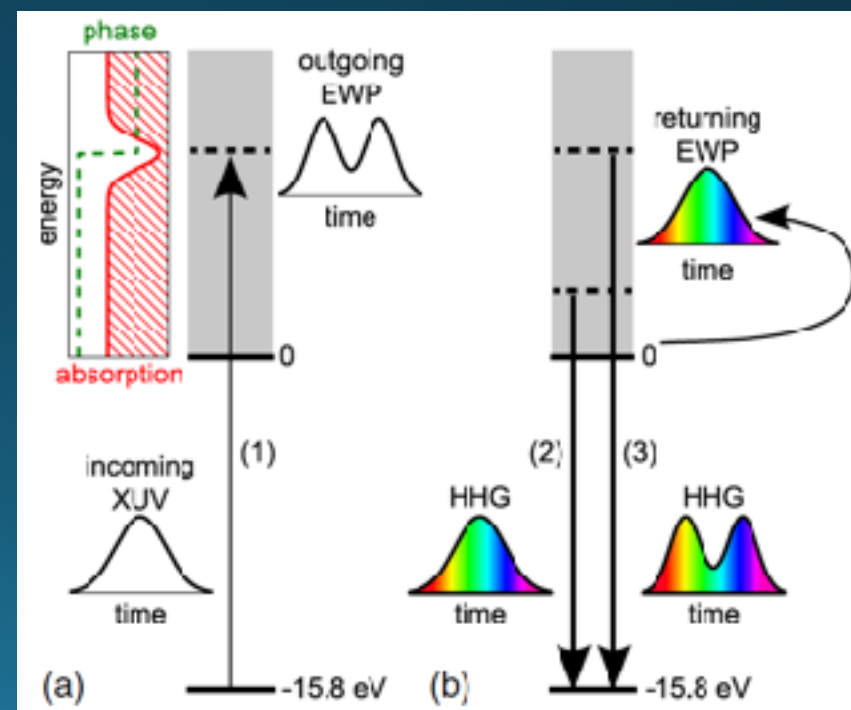
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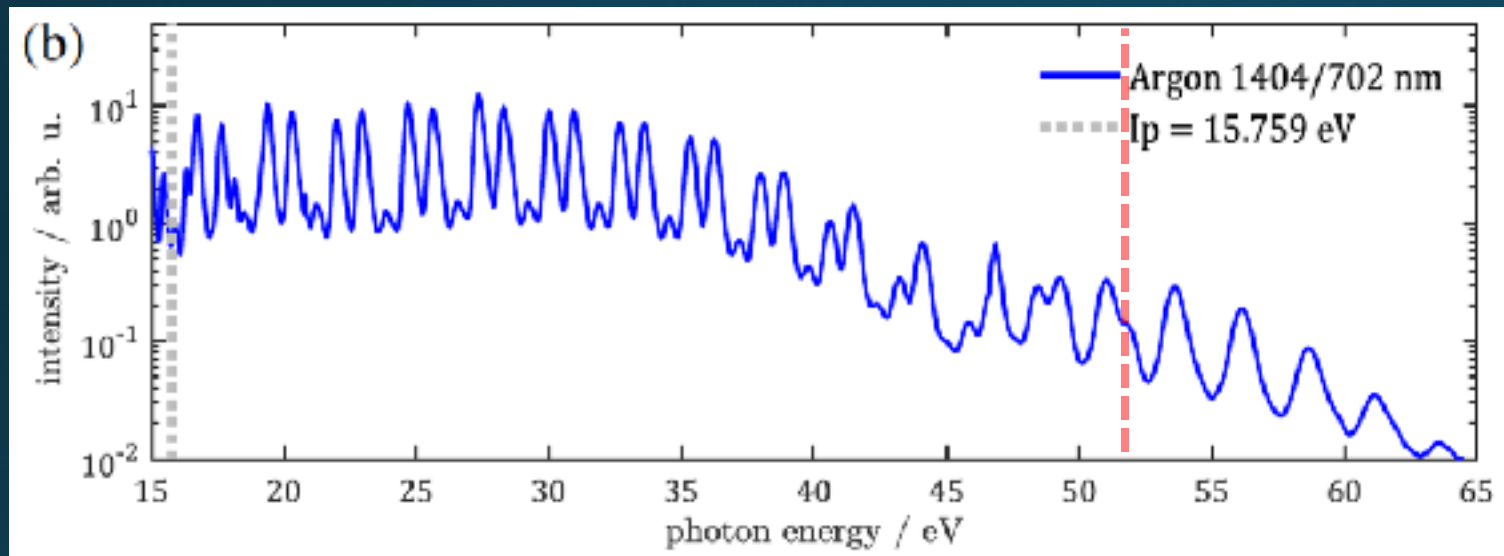
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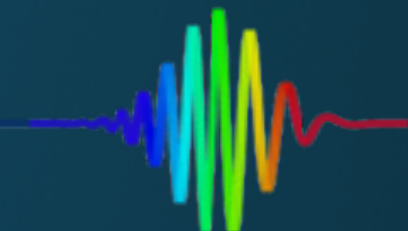


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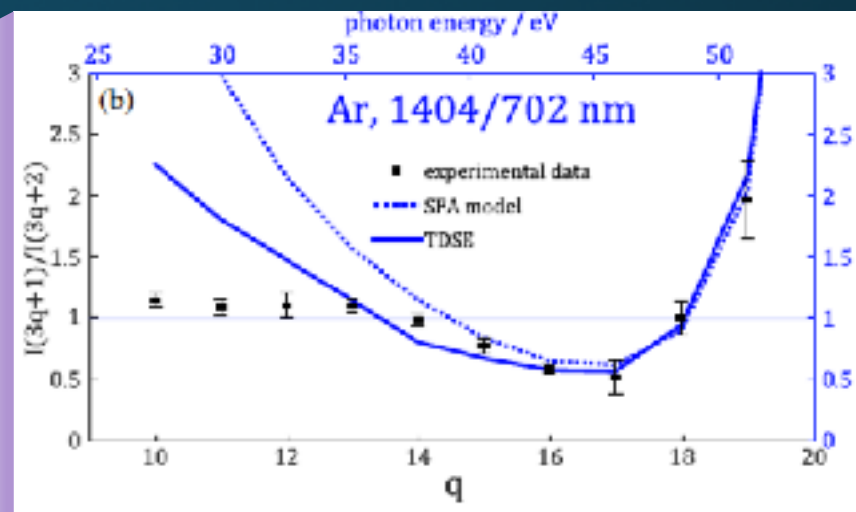
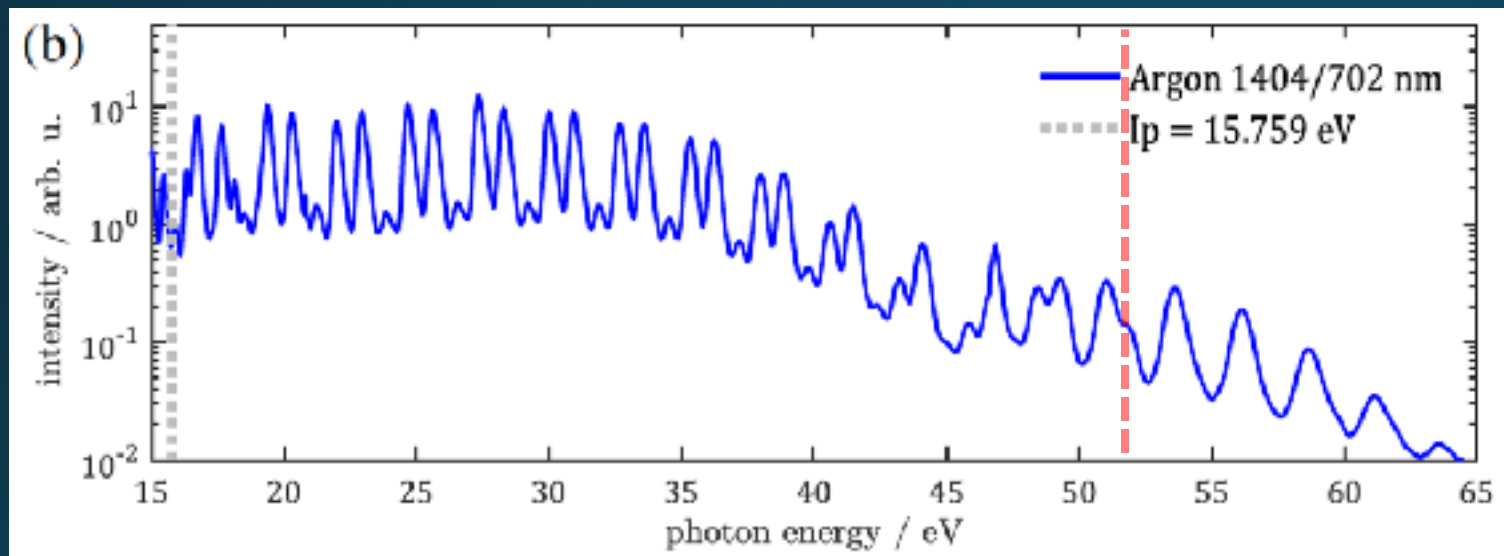
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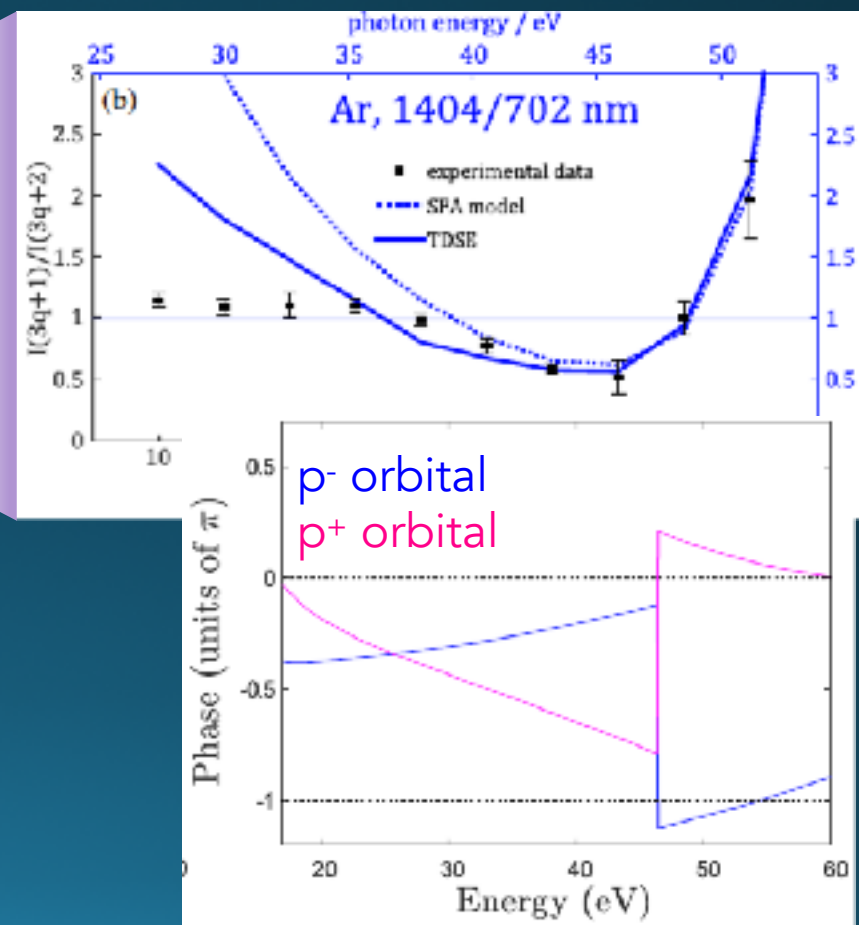
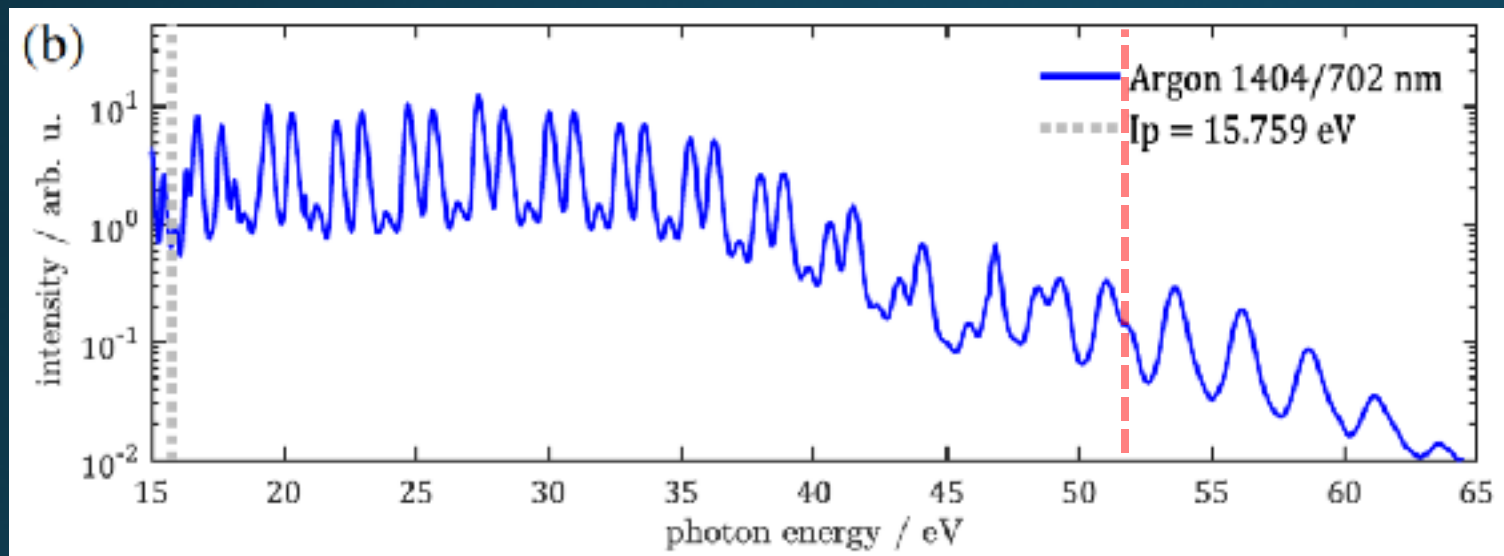


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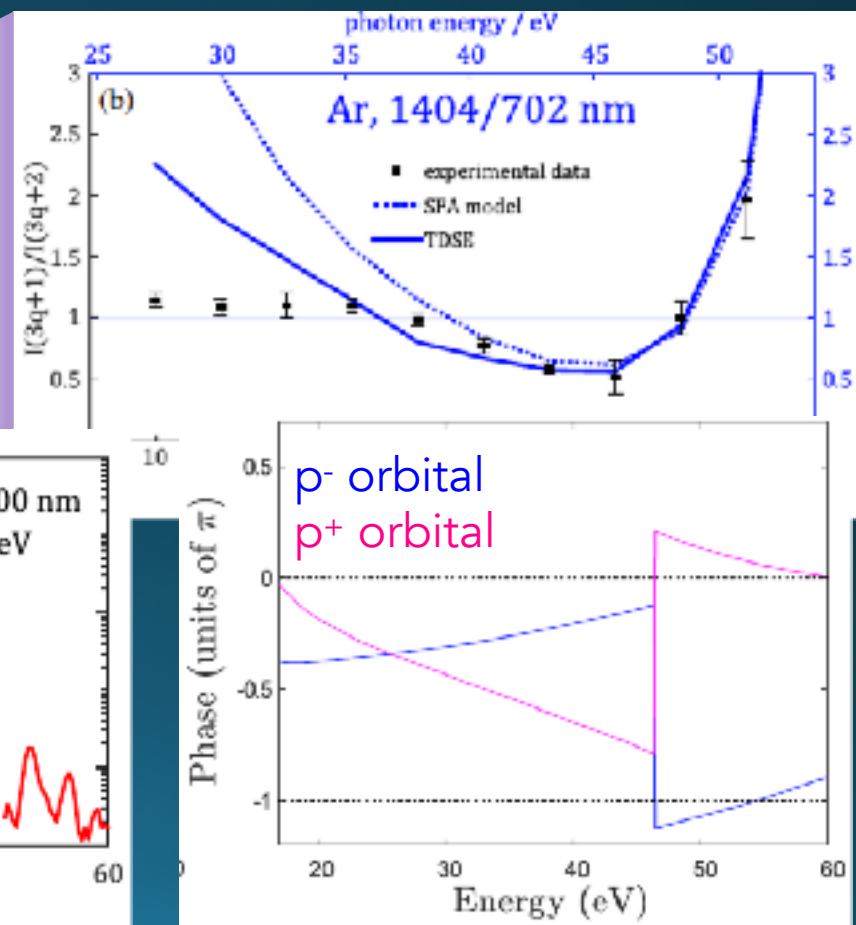
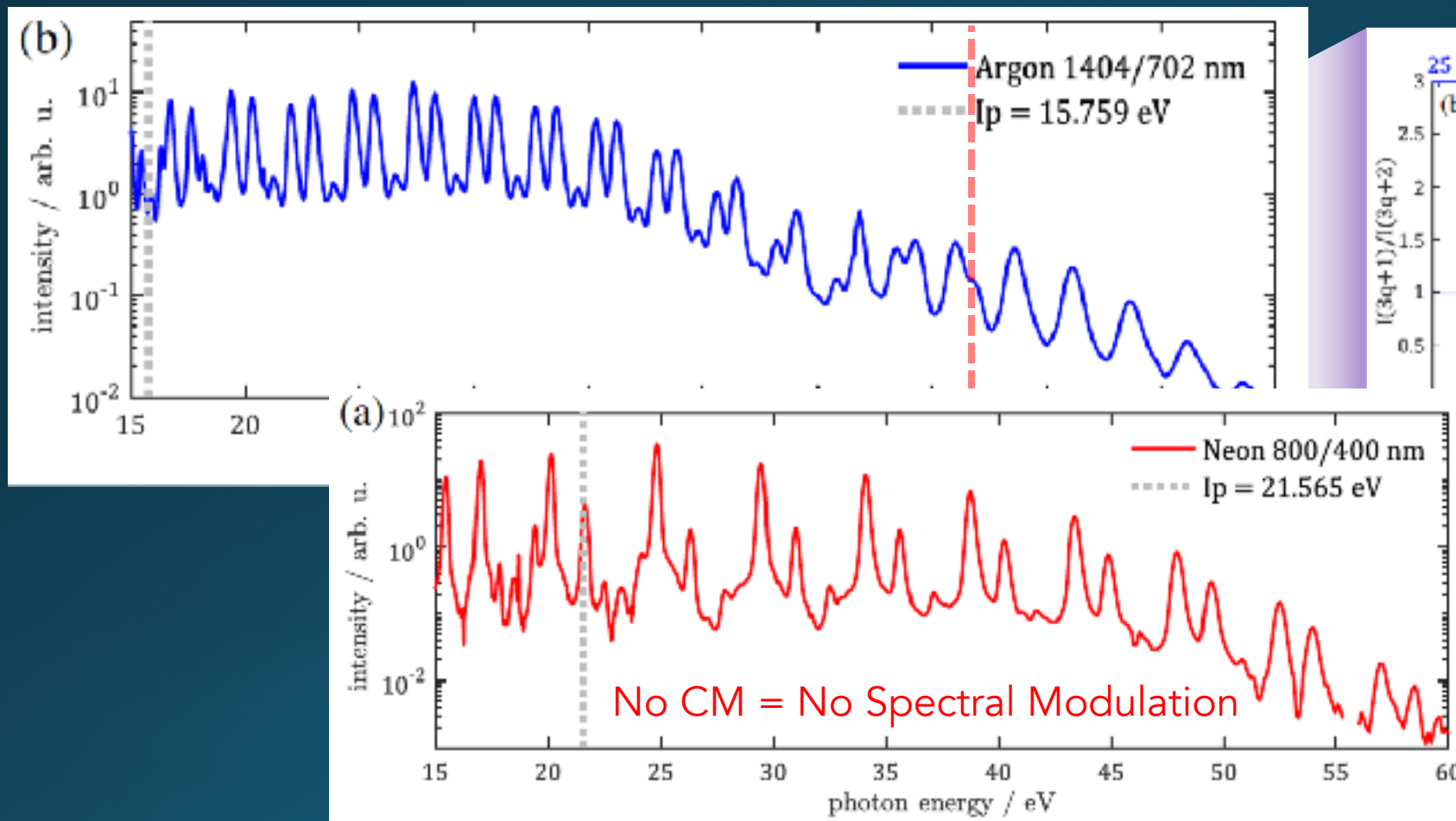
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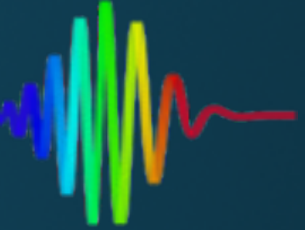
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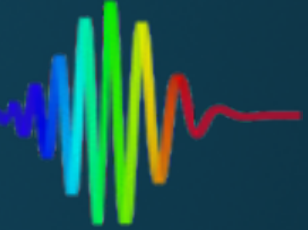


Merging CPHHG and BHHS: Exploiting Electronic Structure as an EUV Waveform Modulator!





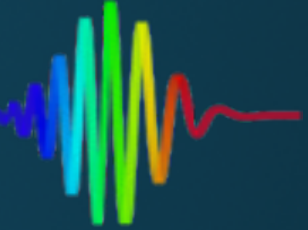
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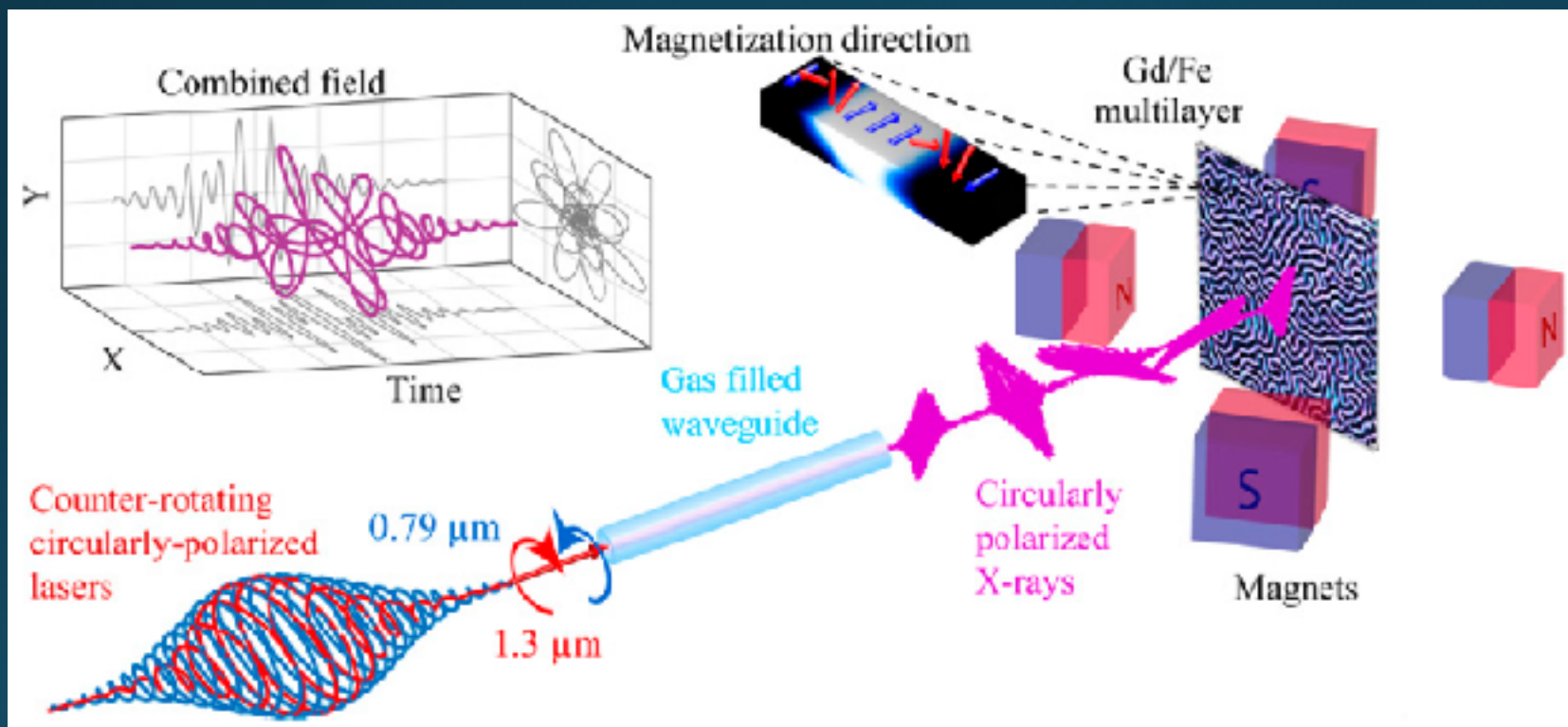


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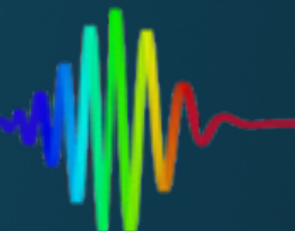
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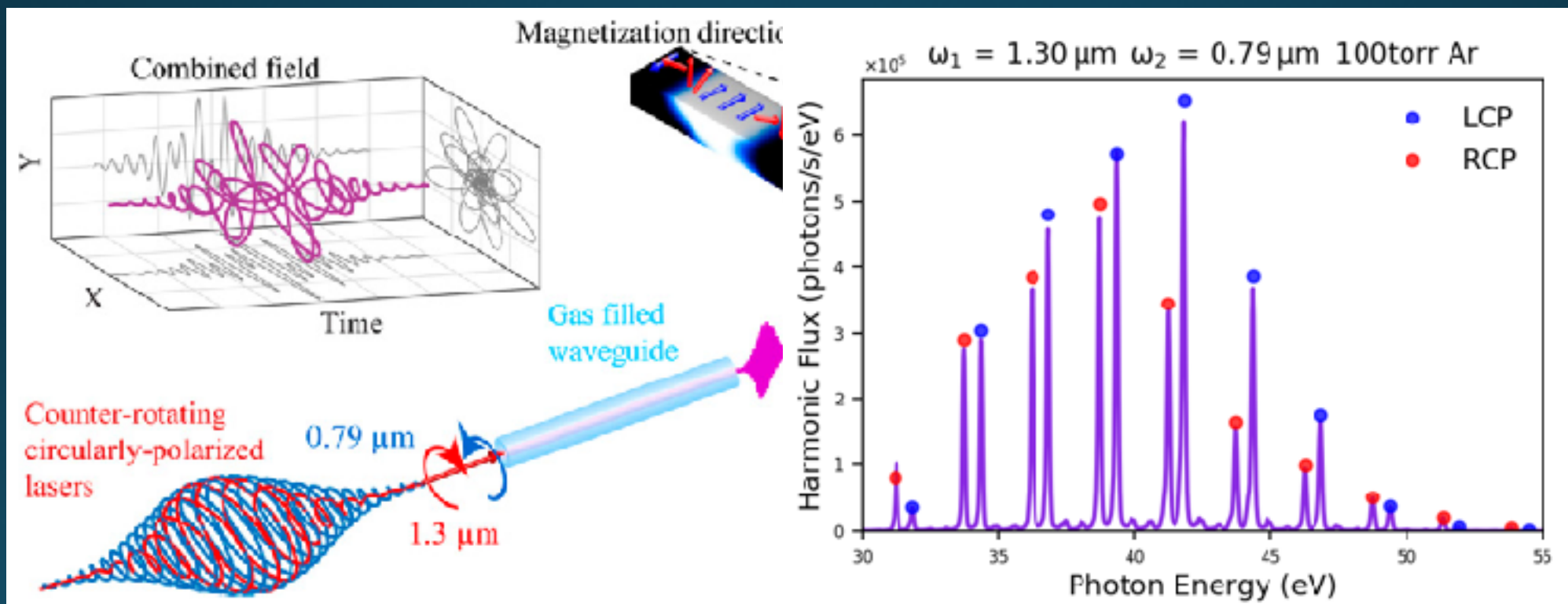
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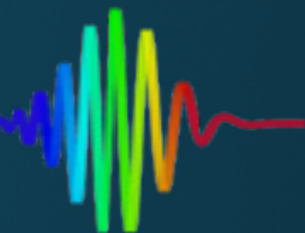
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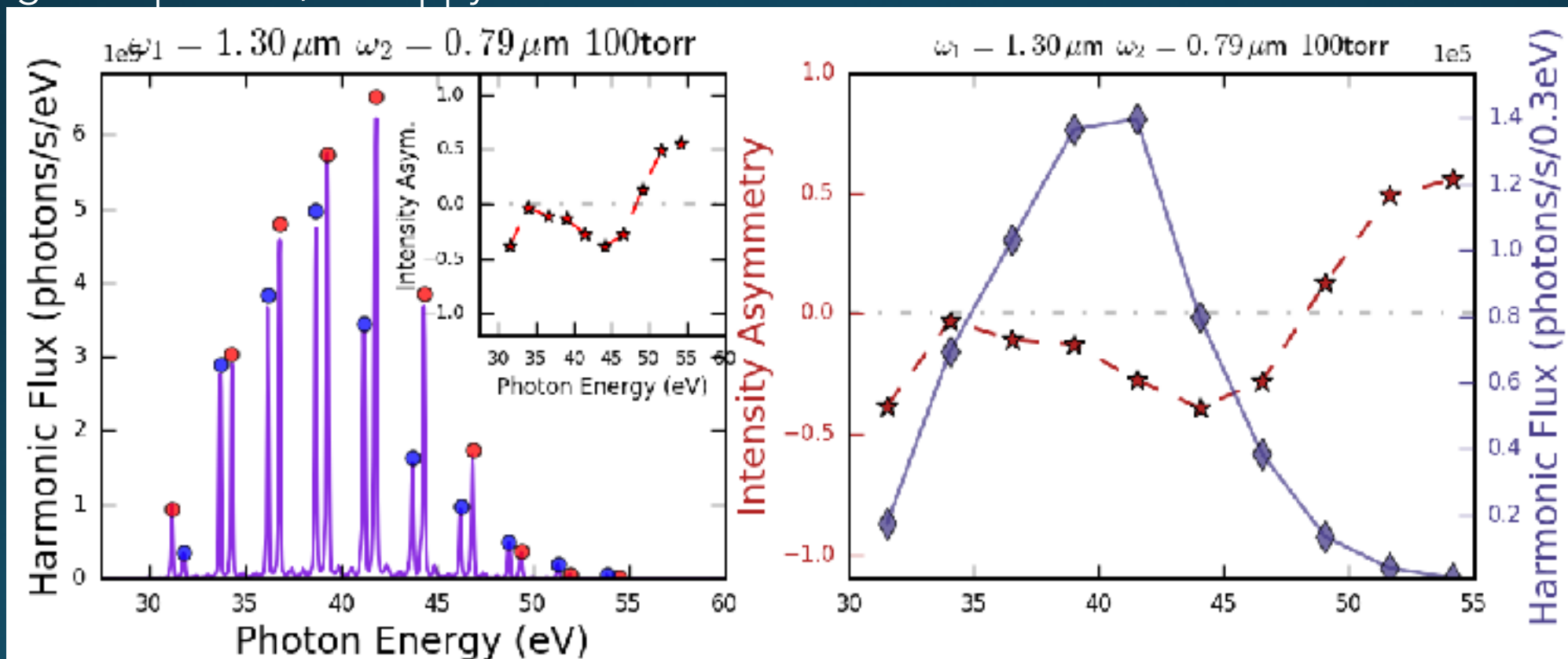
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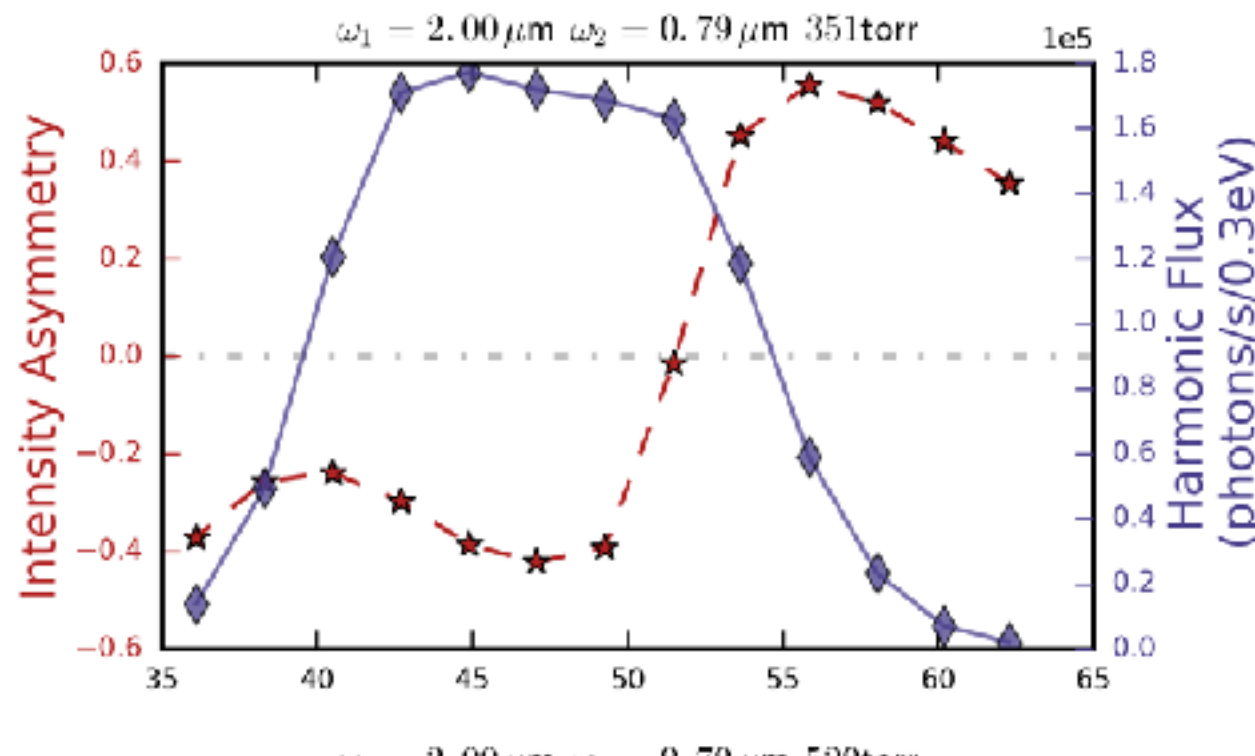
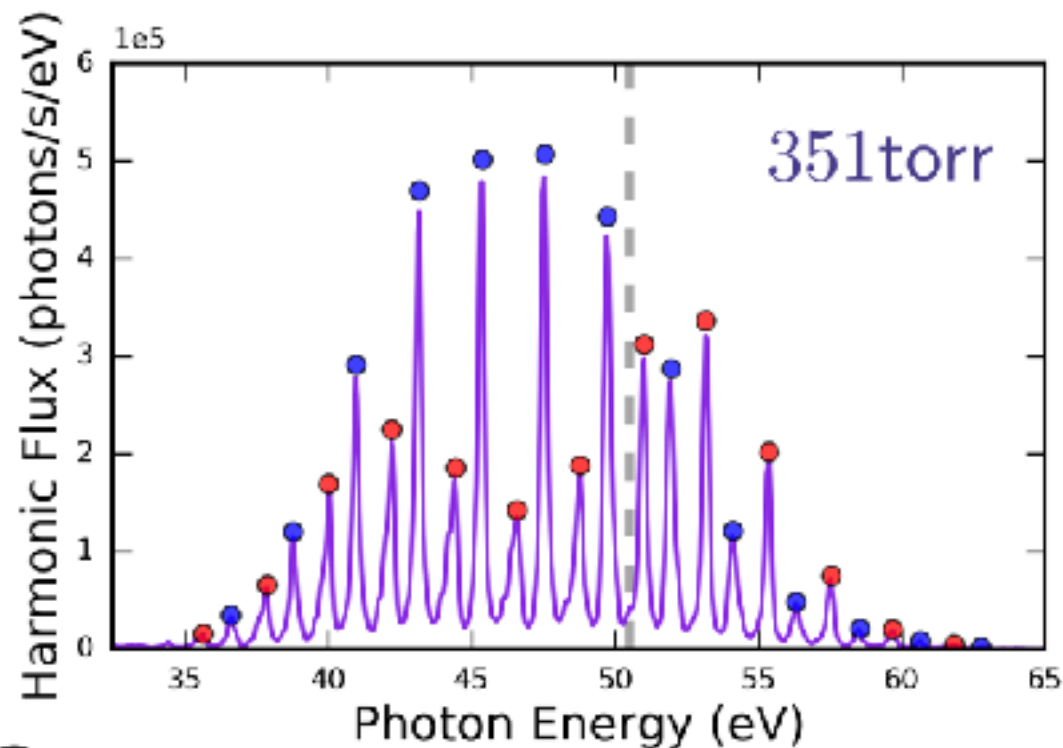
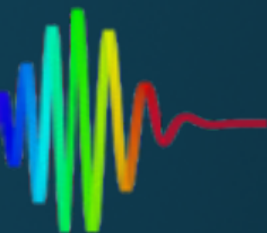
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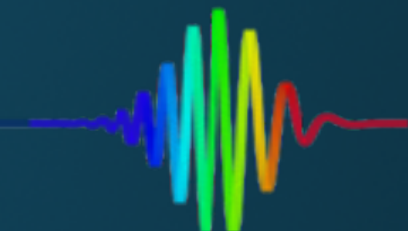
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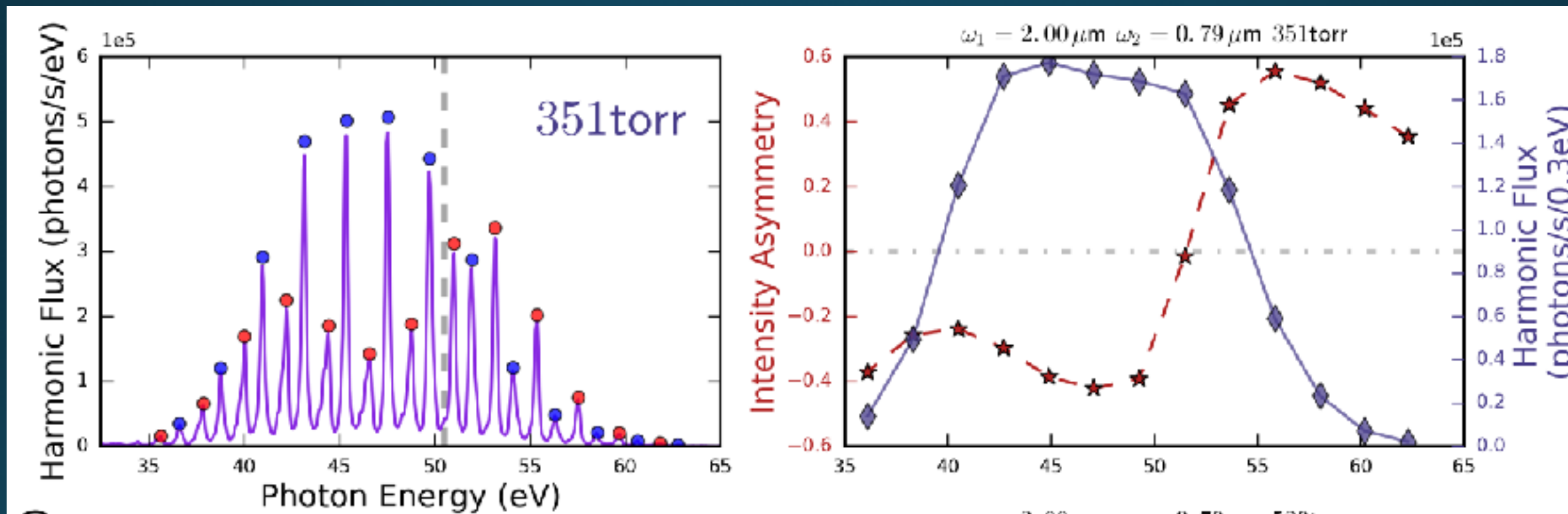
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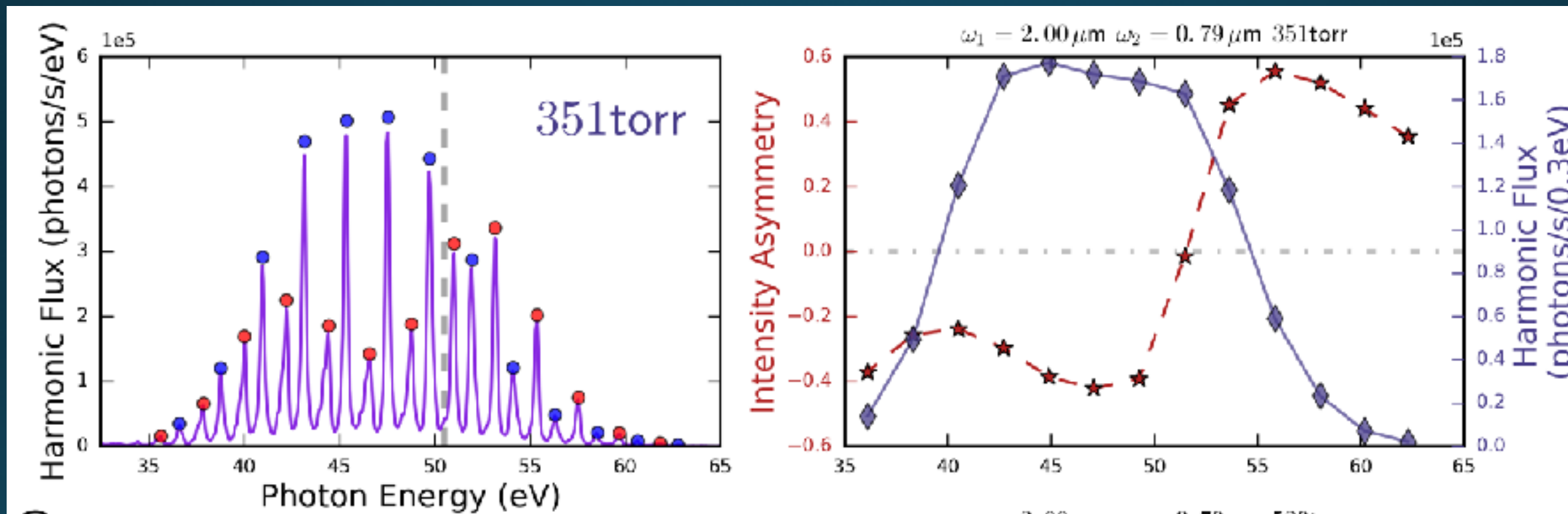


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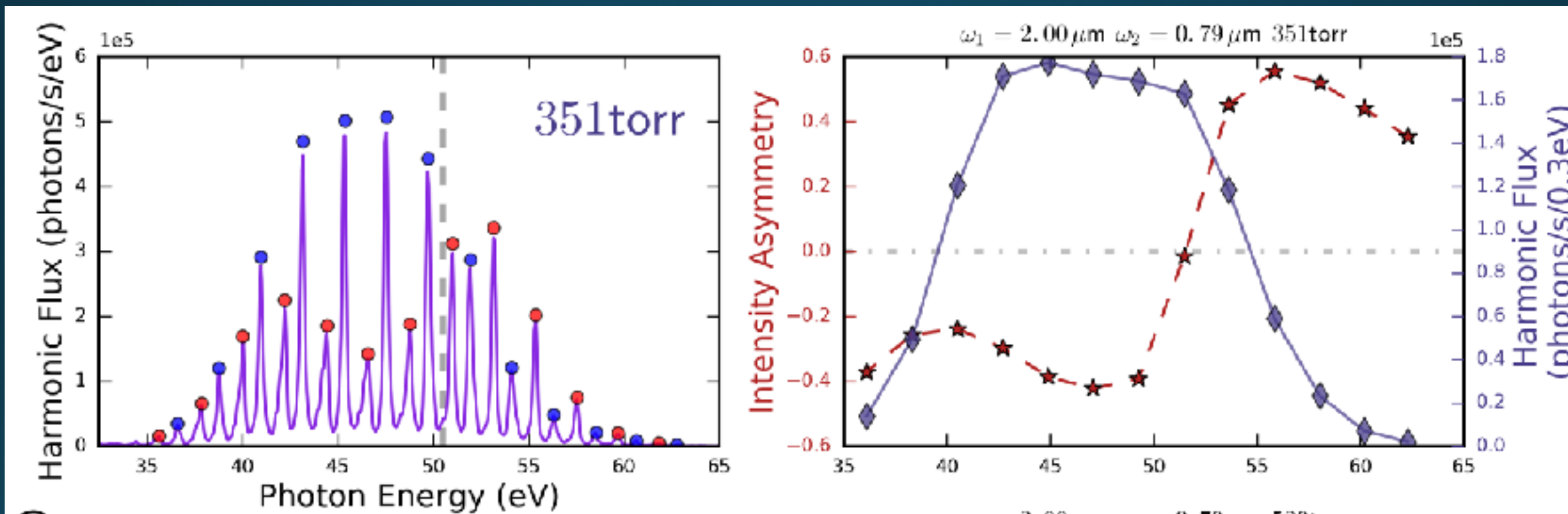
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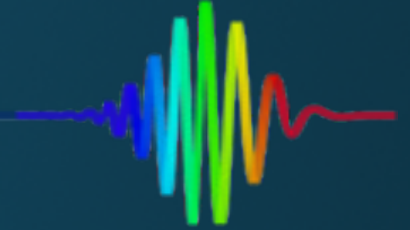
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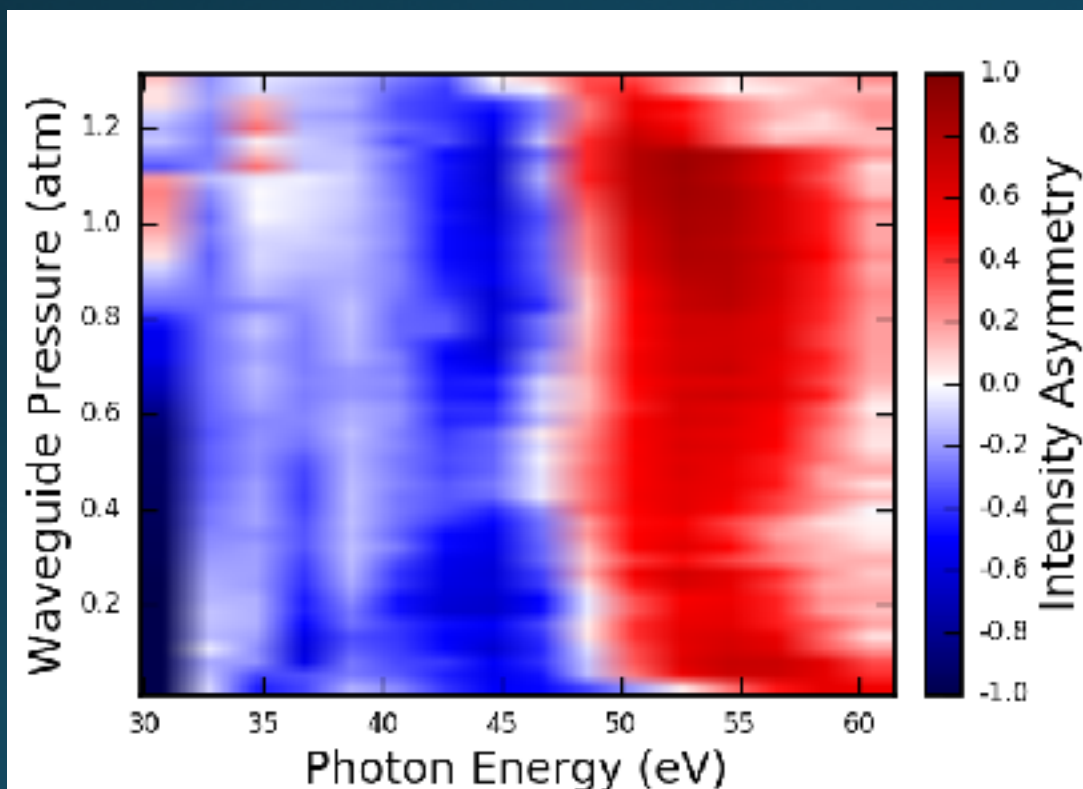
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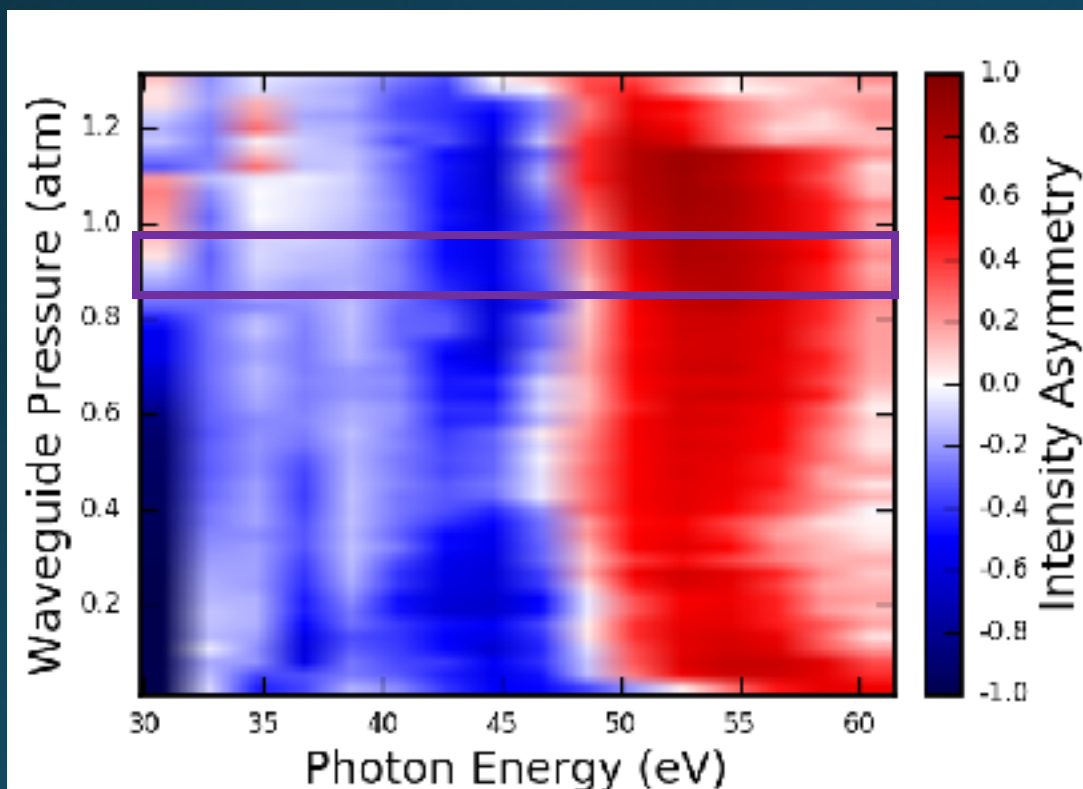


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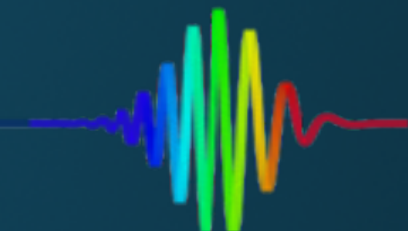


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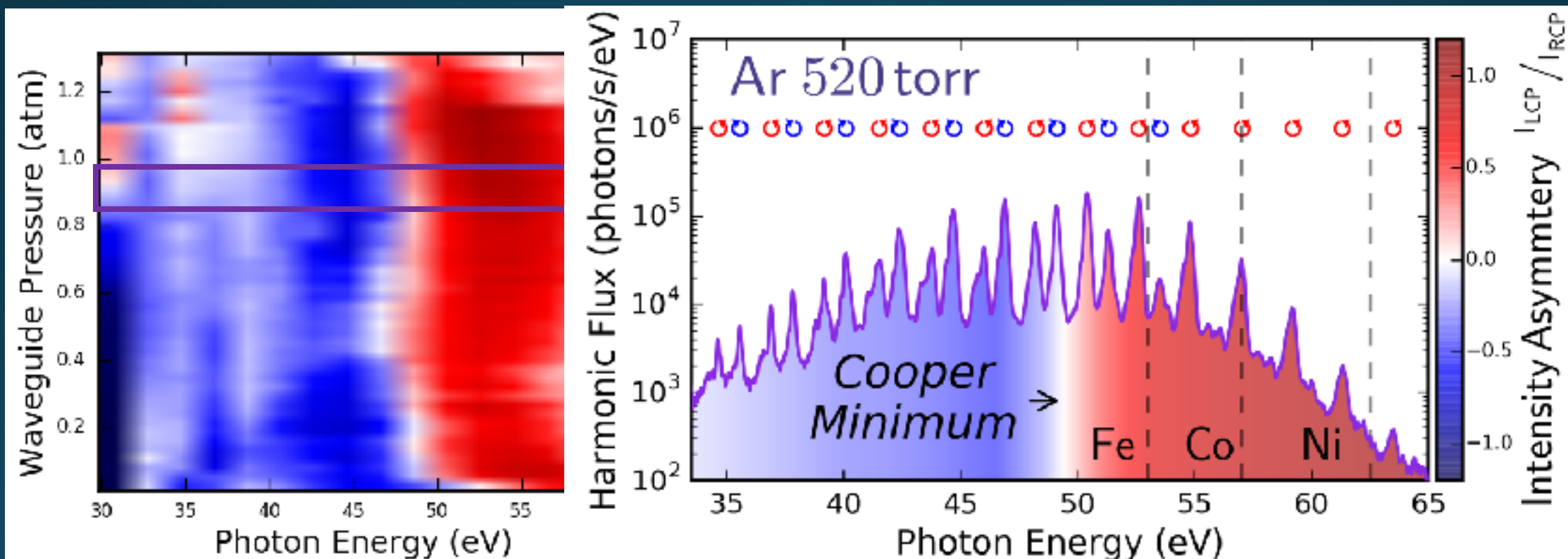


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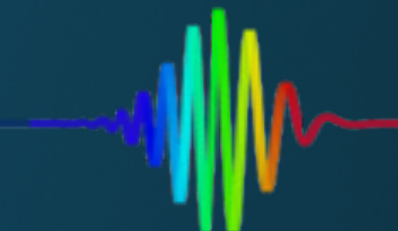


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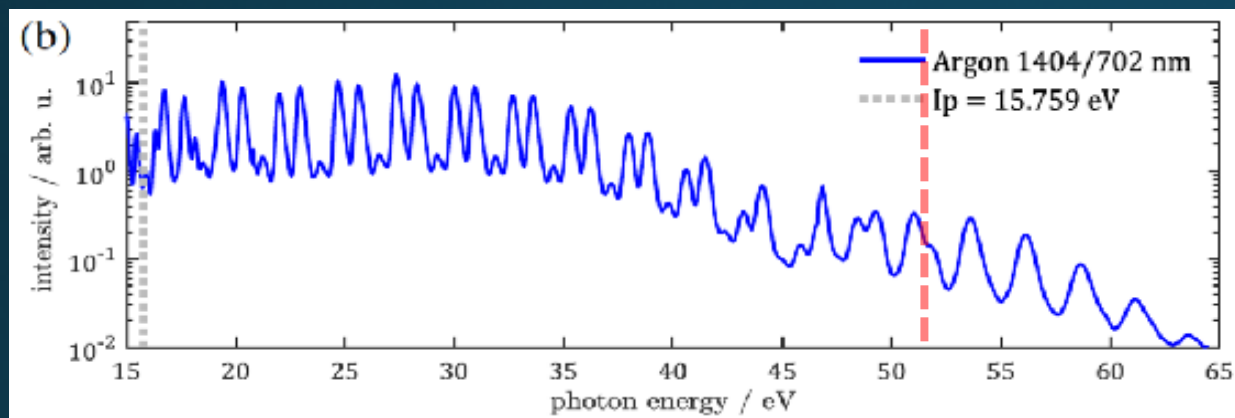
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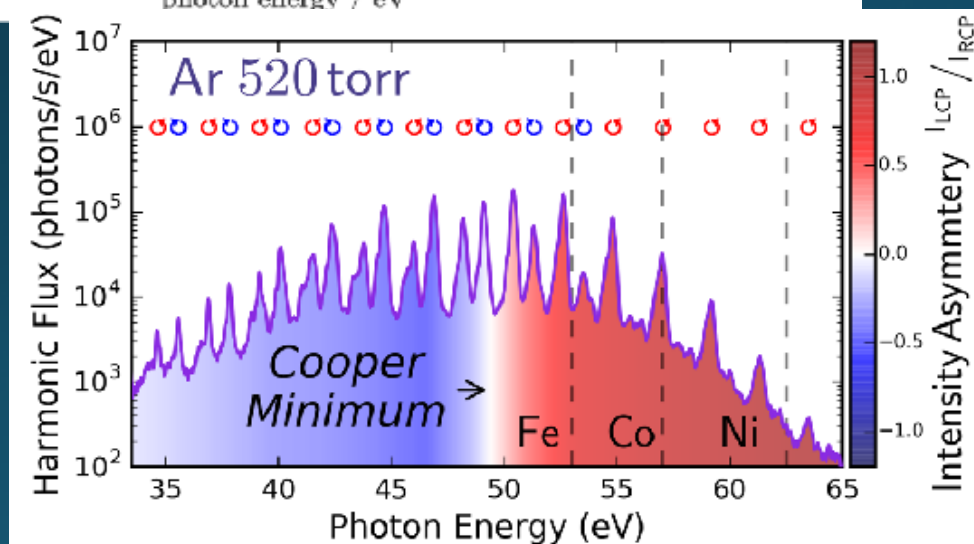
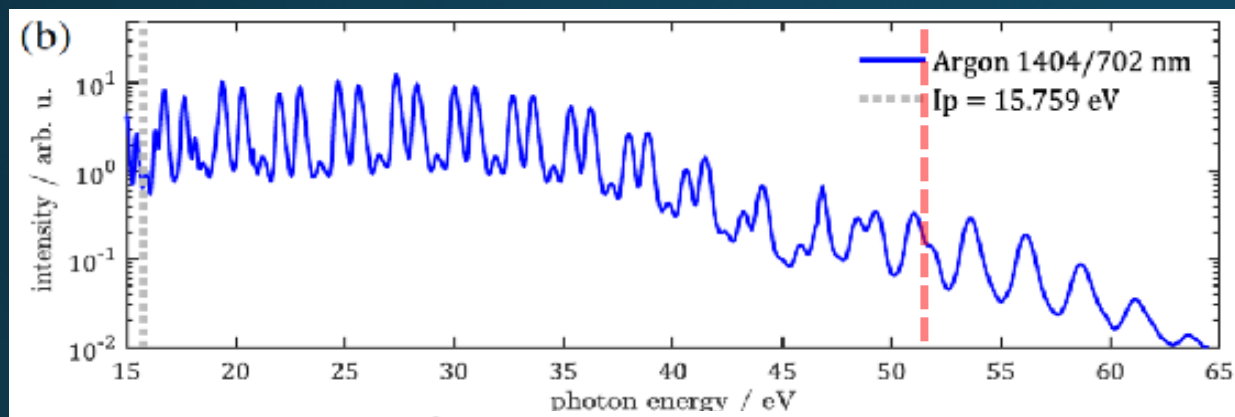


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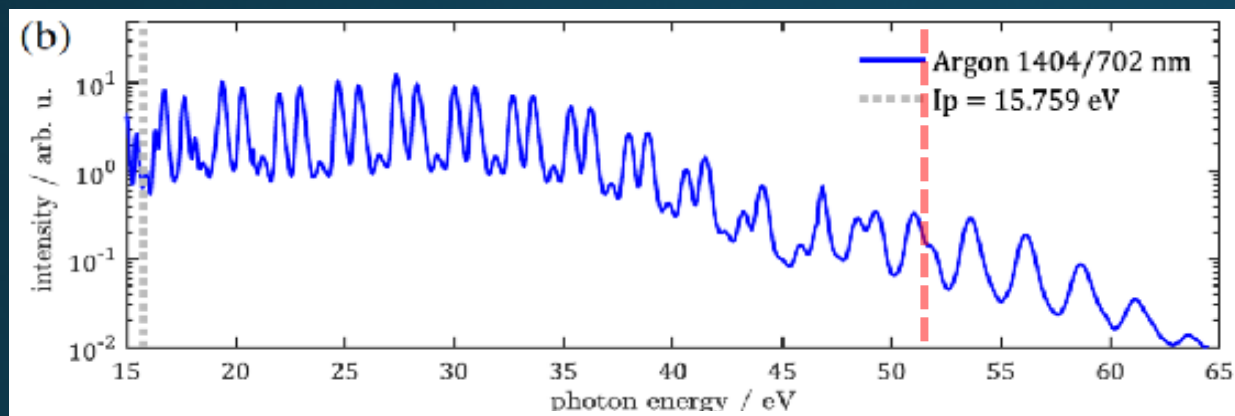
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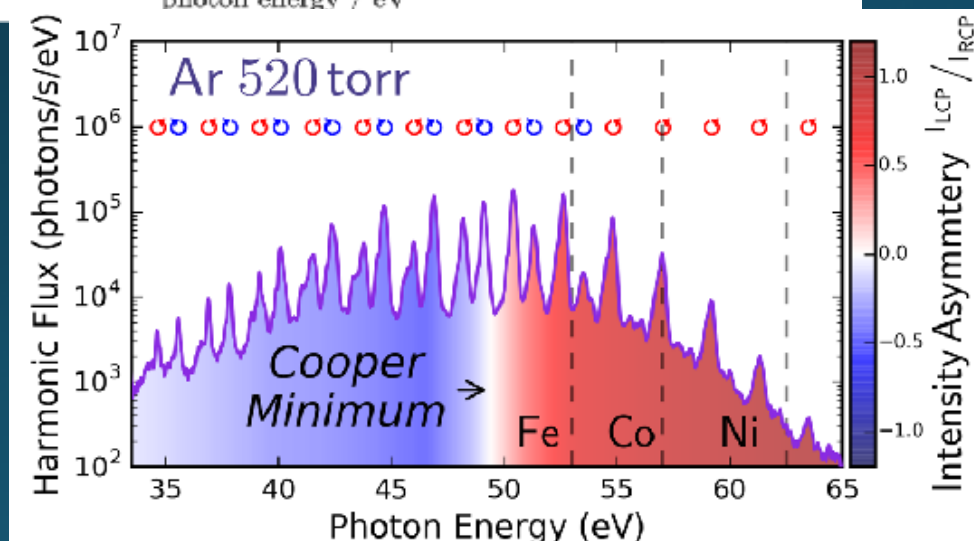


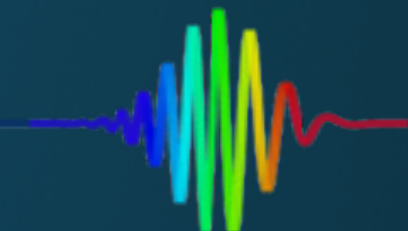


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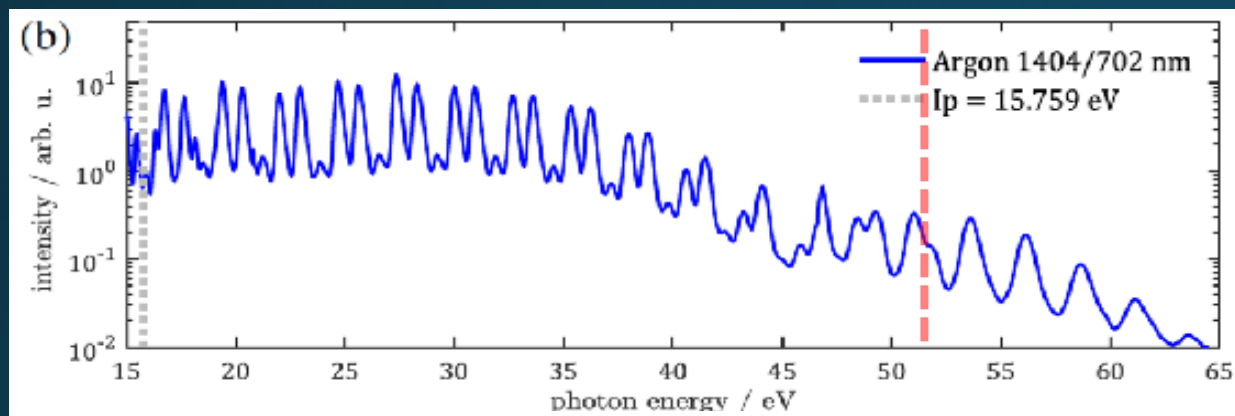


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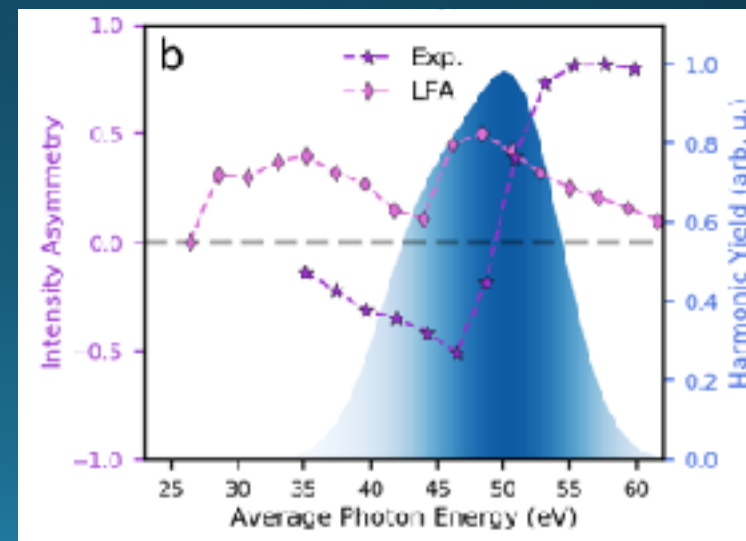
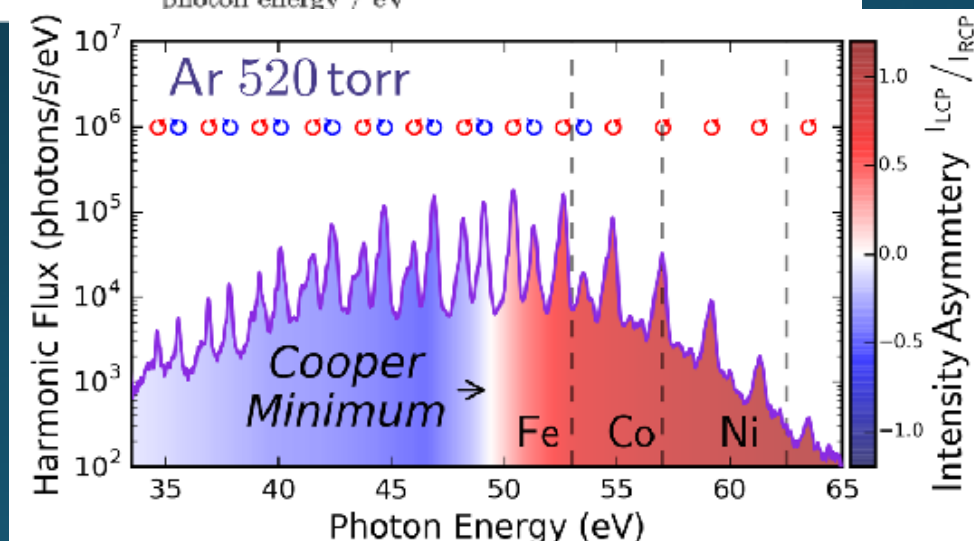




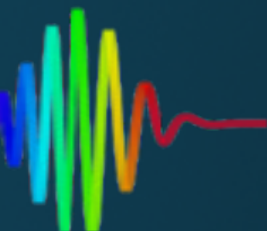
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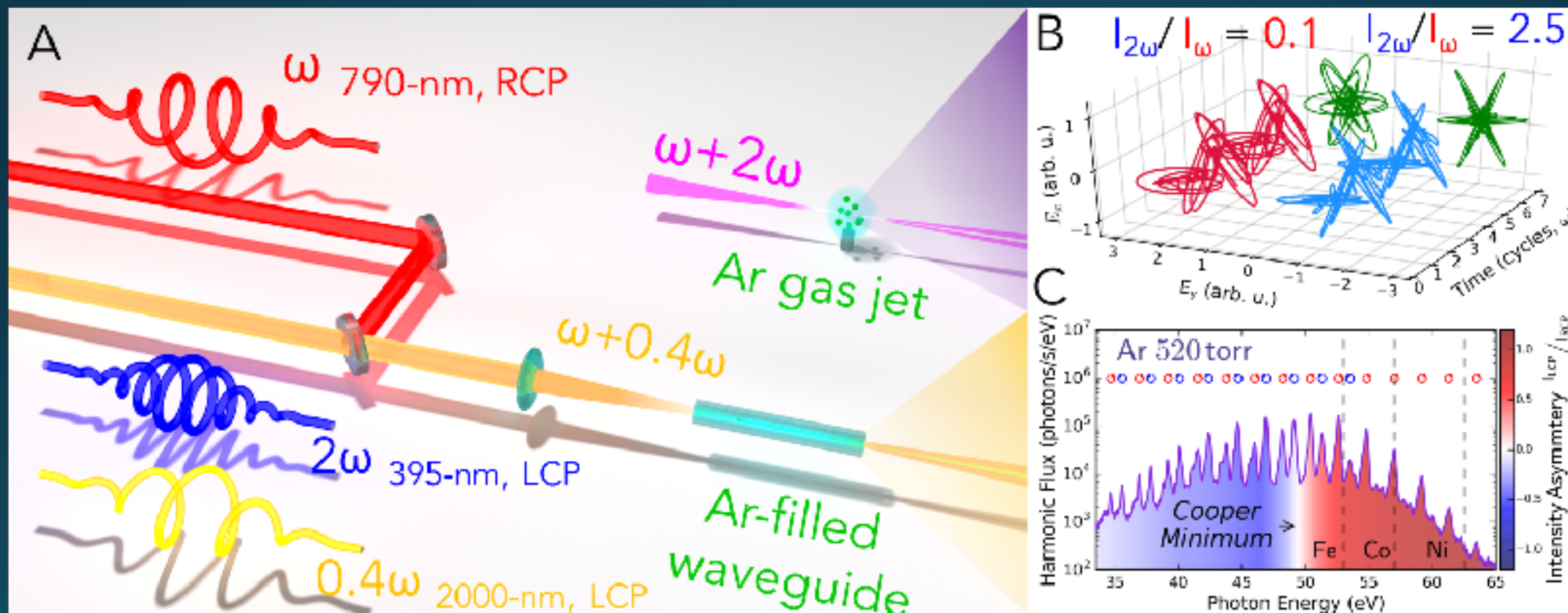
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Bicircular HHG and HHS: A Versatile Light Source for Chiral Spectroscopies and Dynamics!



- Straightforward, versatile control of the attosecond polarization, all while preserving the spectral circularity of the harmonics!



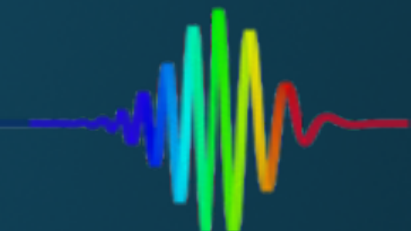


CPHHG from Linearly Polarized HHG Drivers: A New (or Lack of) Twist in Thinking





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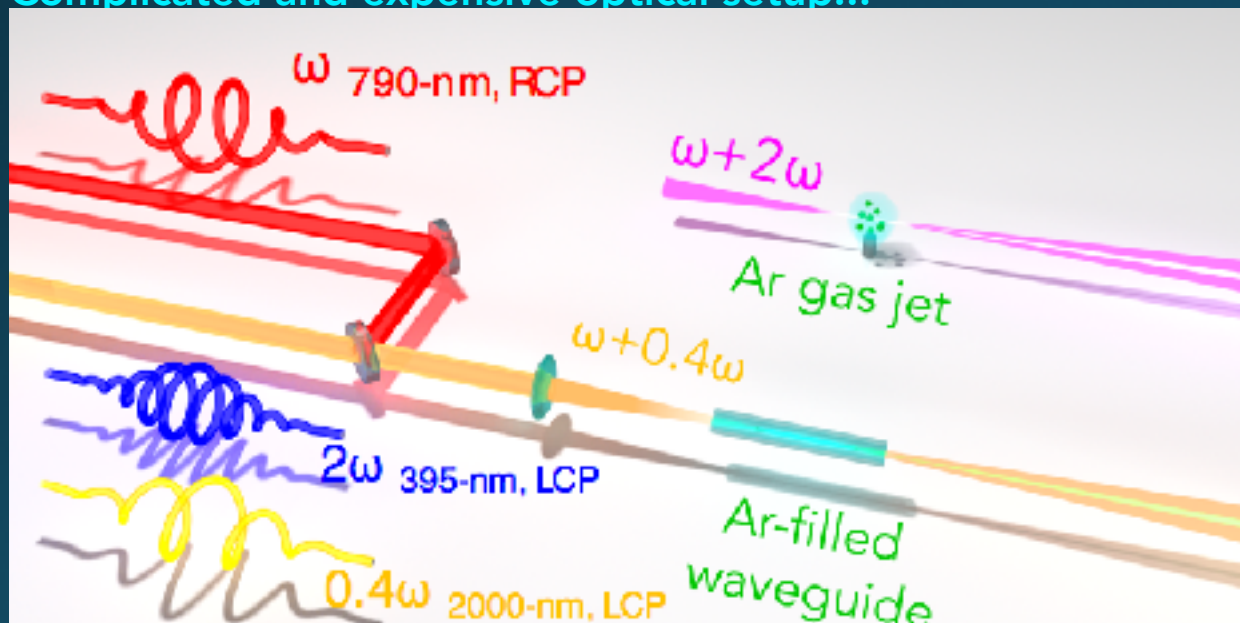


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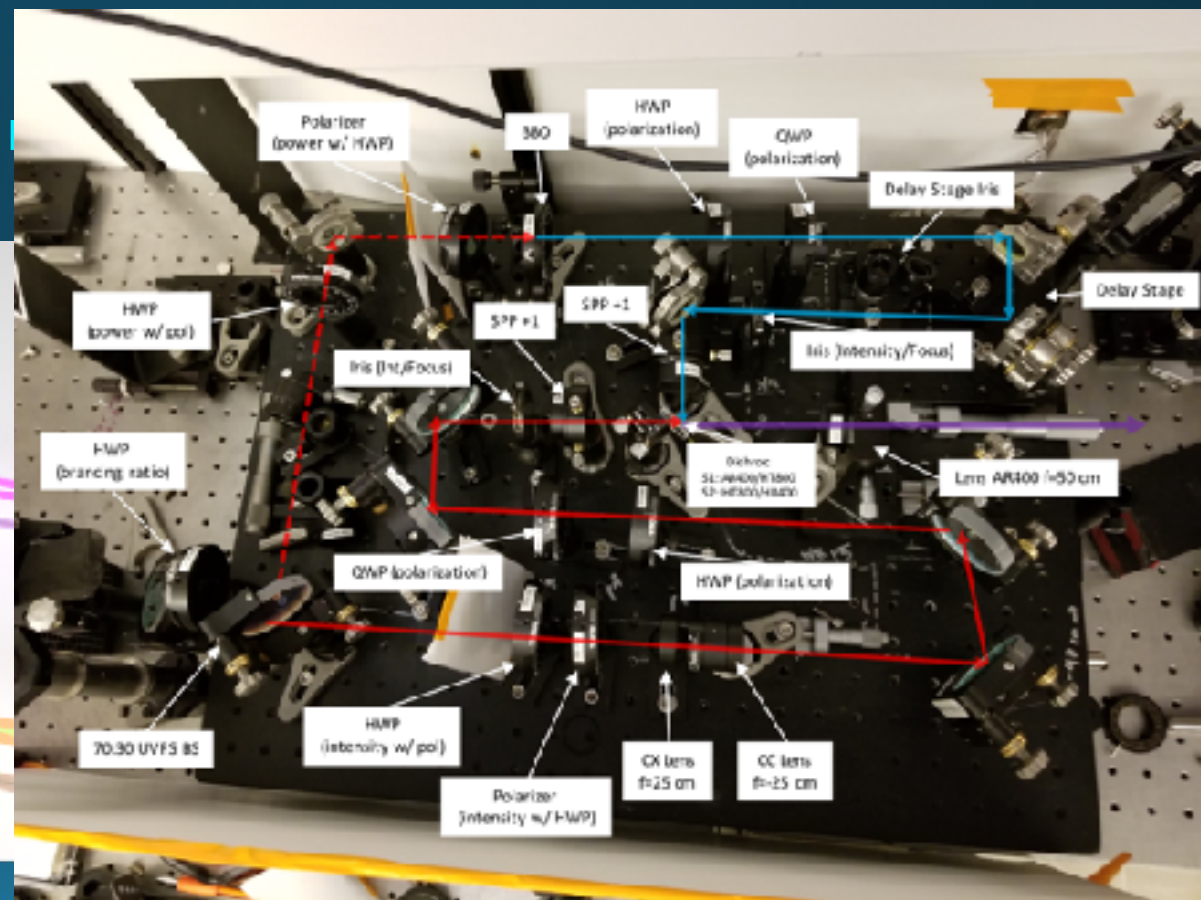
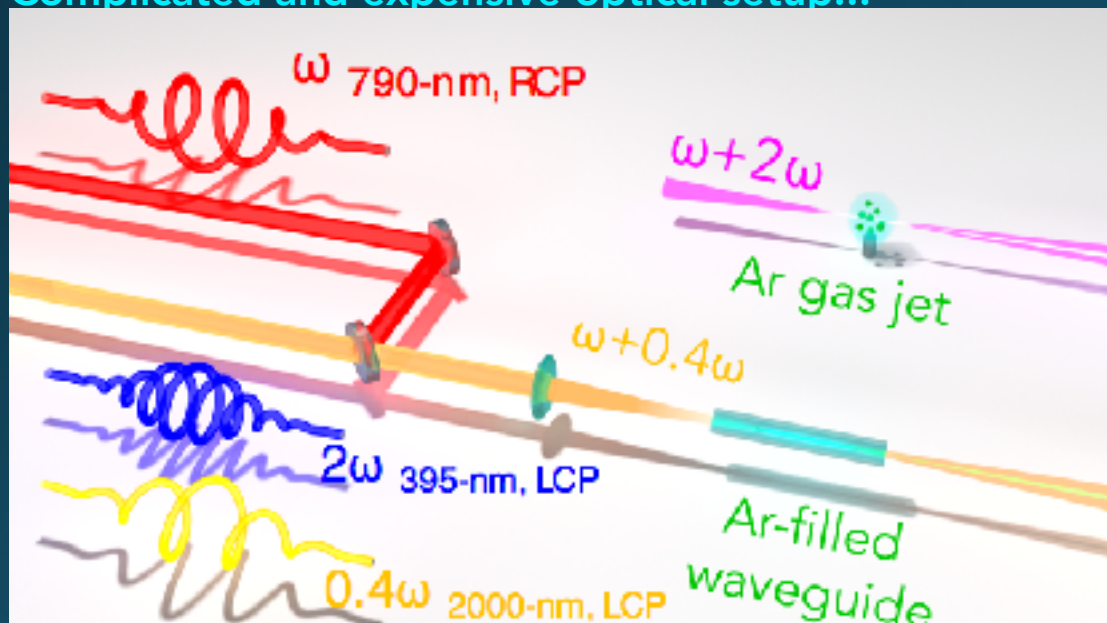
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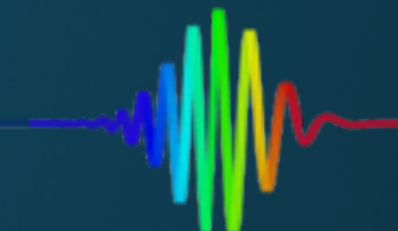


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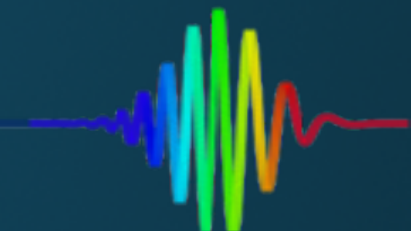


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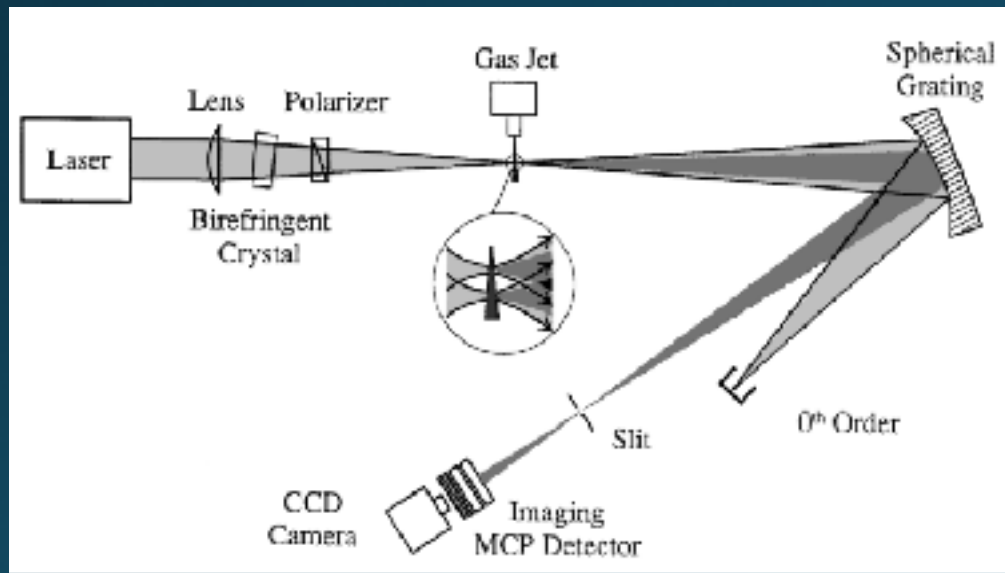


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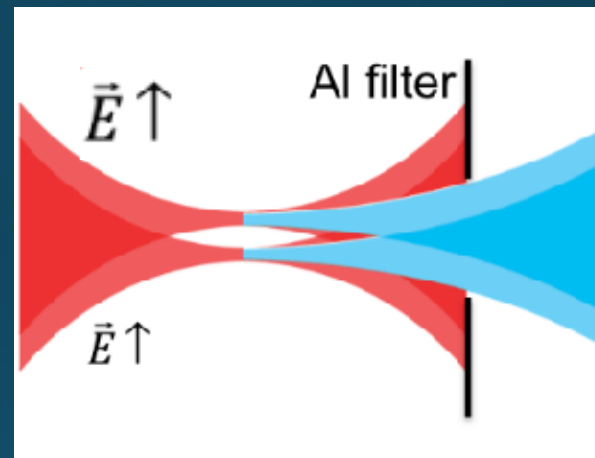
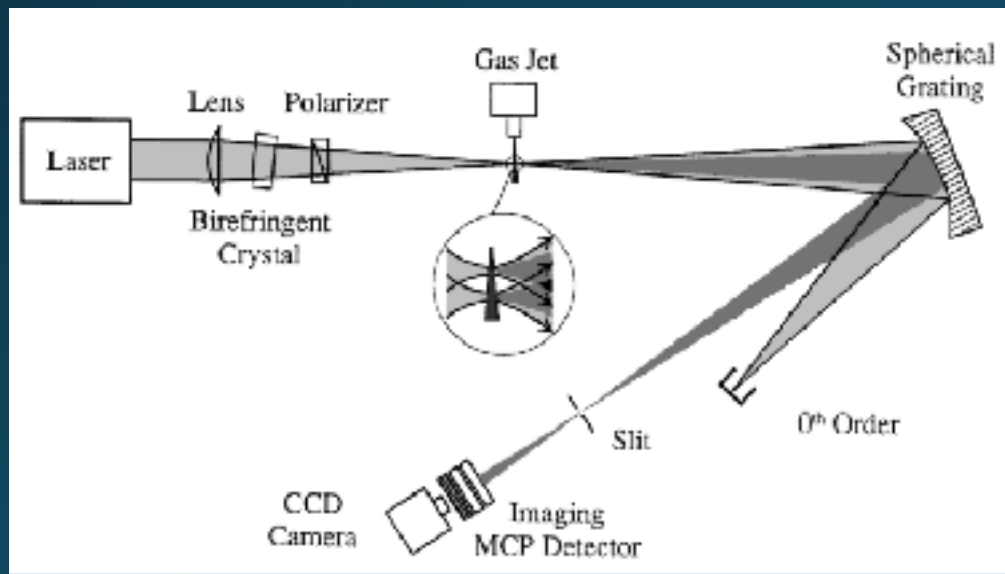
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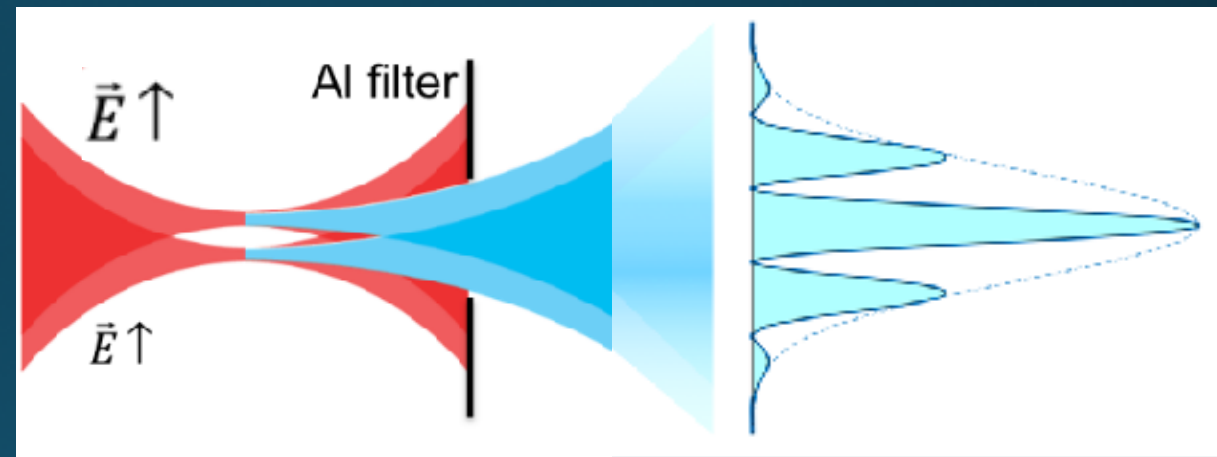
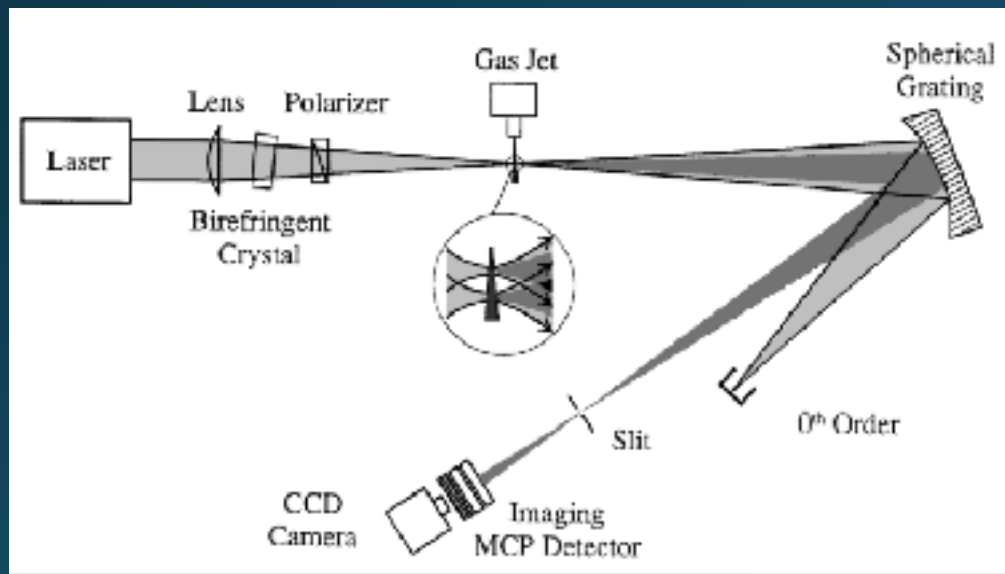


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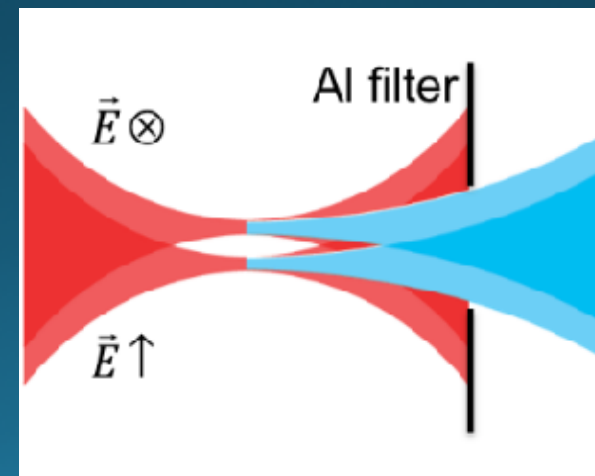
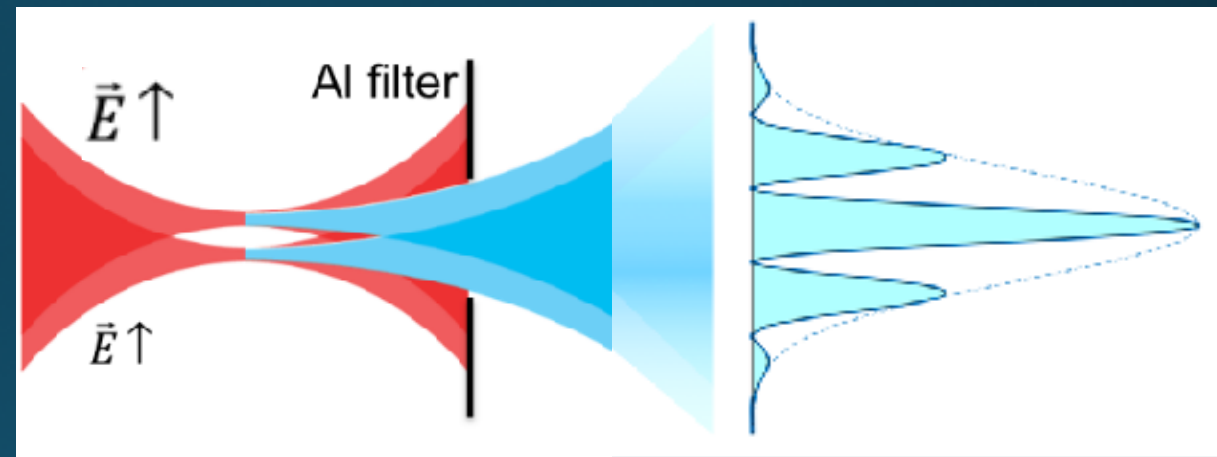
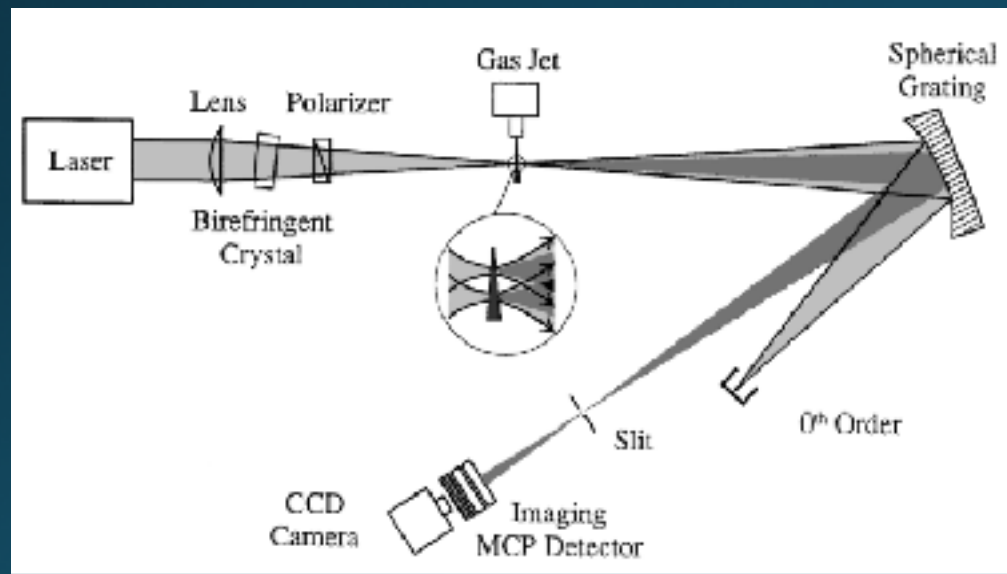
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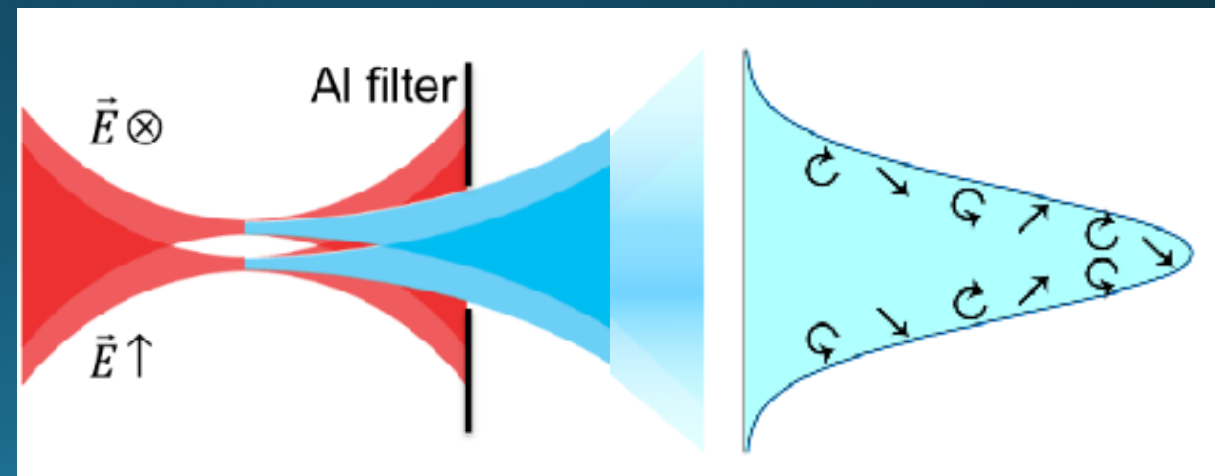
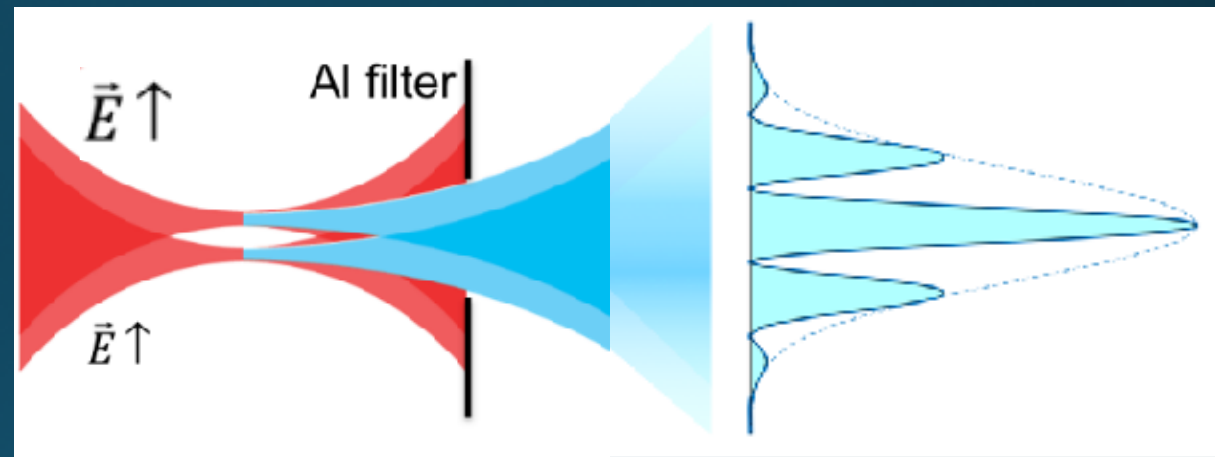
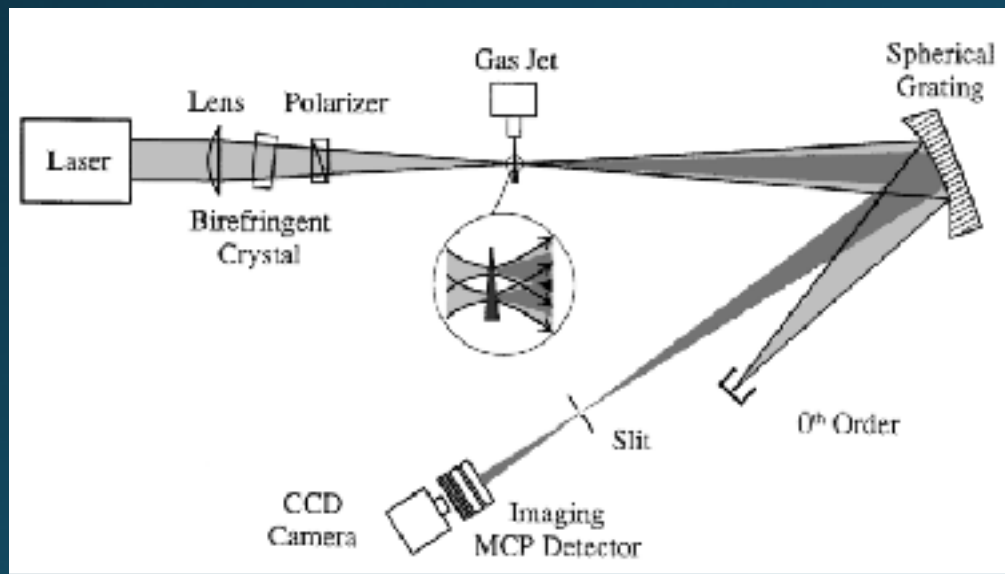
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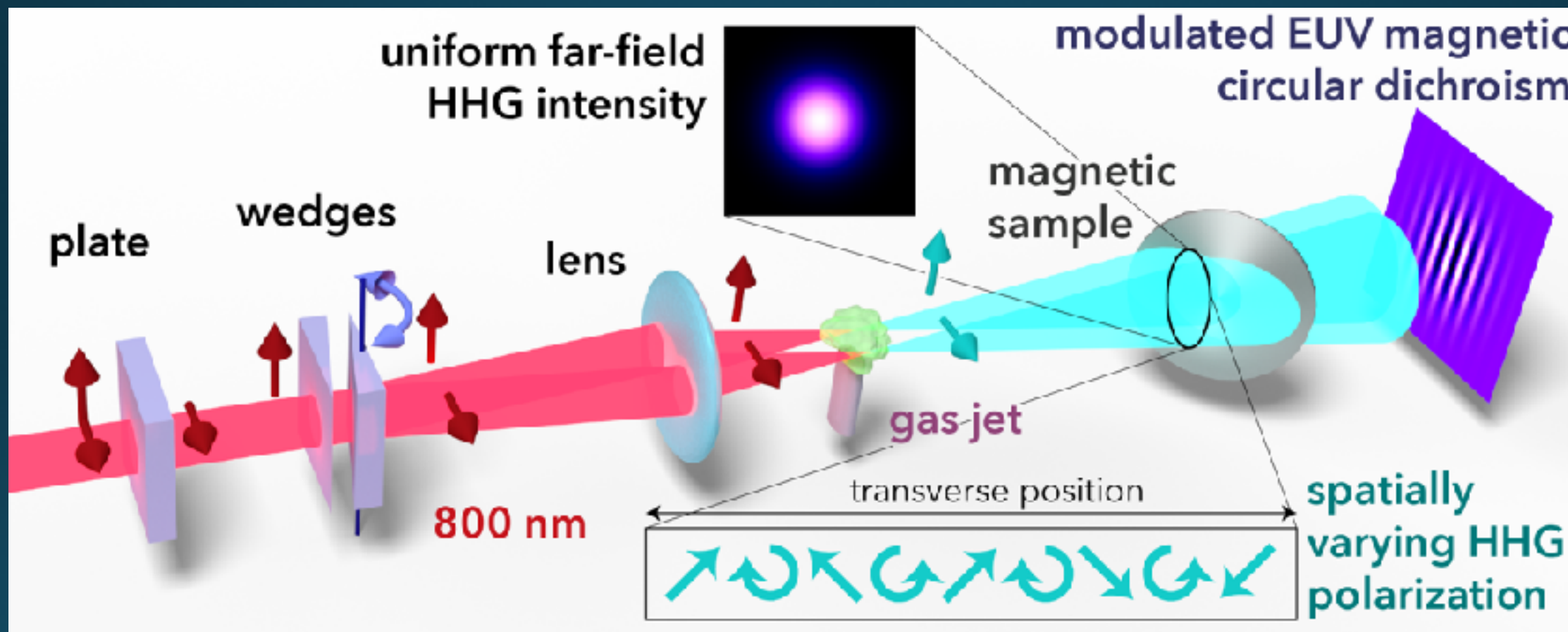
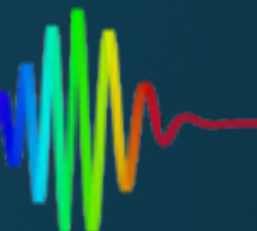


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A Common-Path, Ultra-stable, Interferometer for CPHHG and
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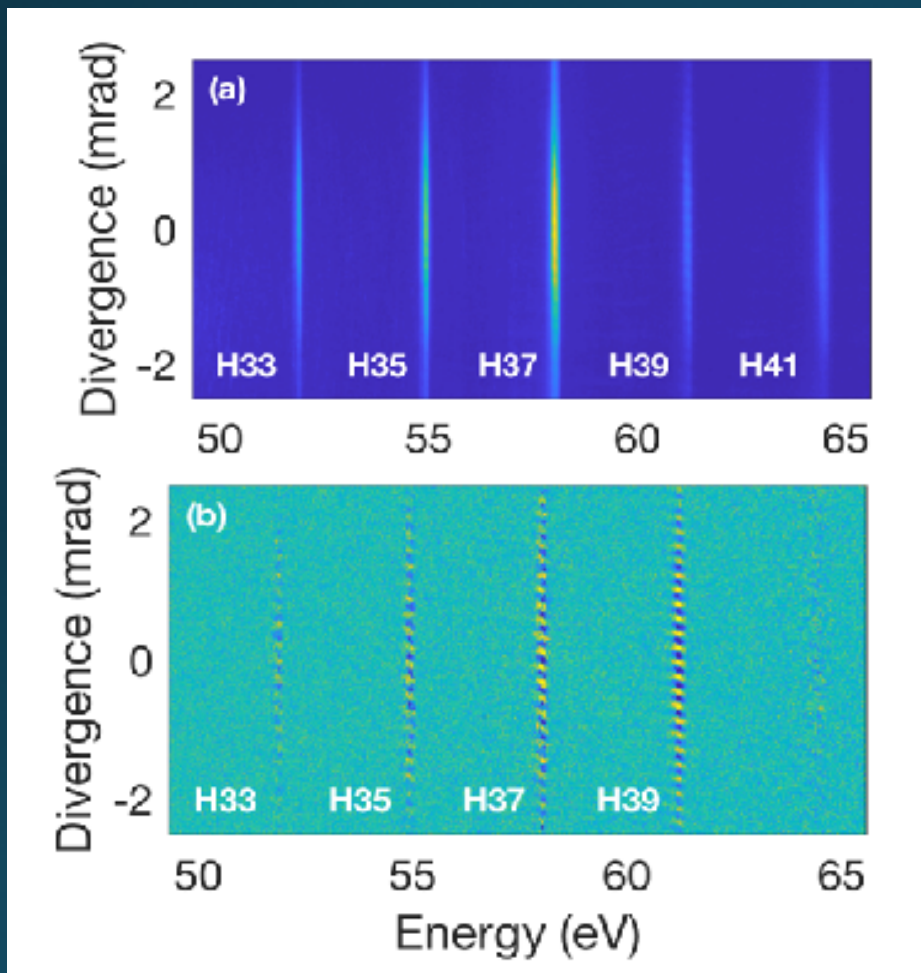


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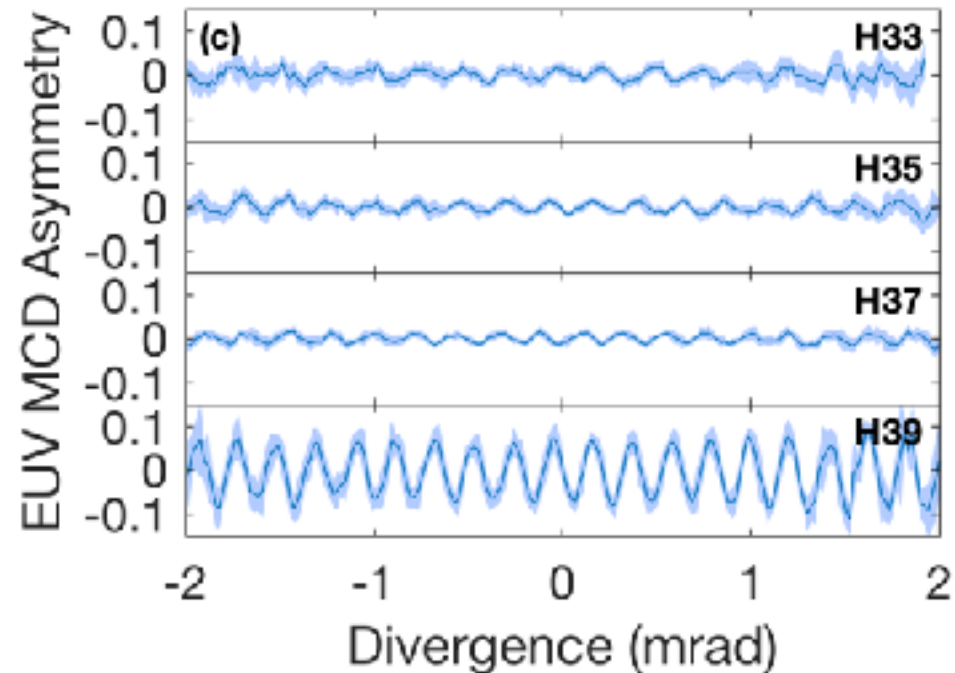
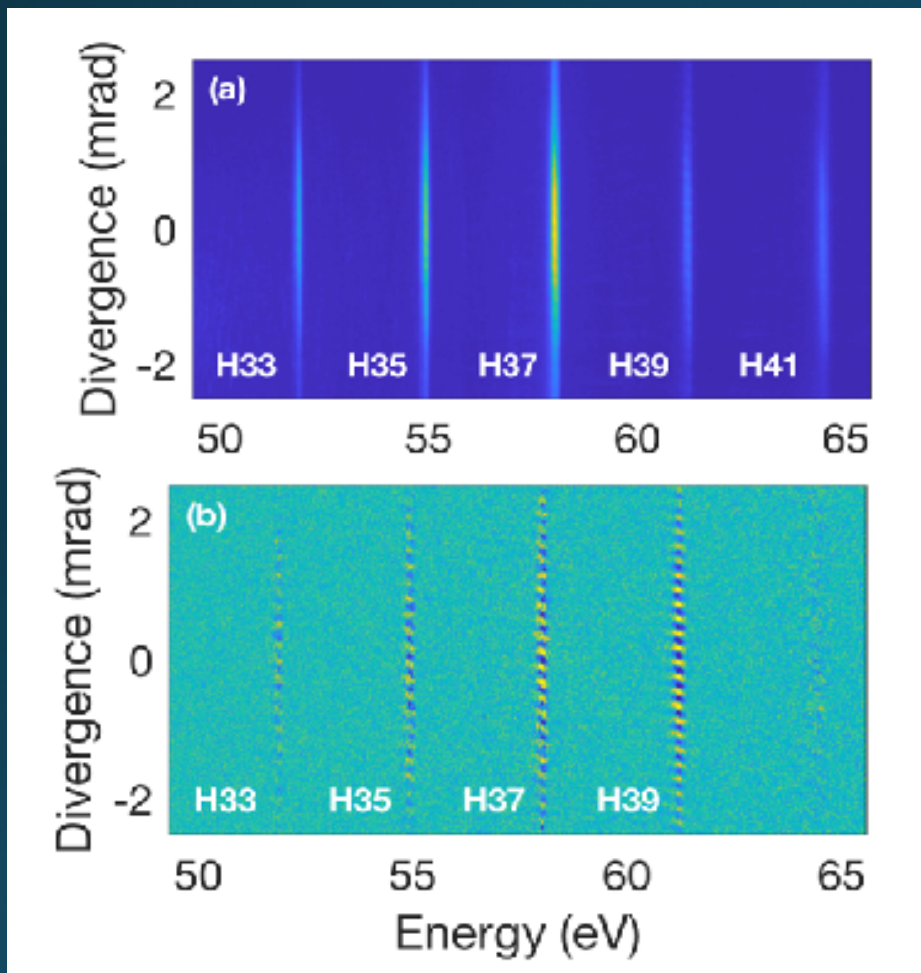
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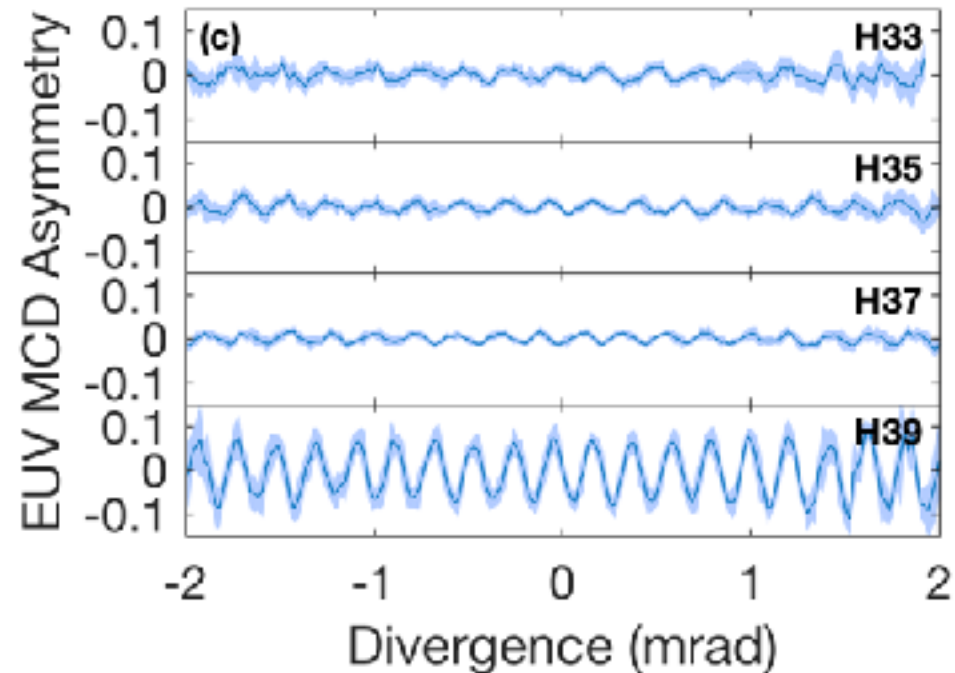
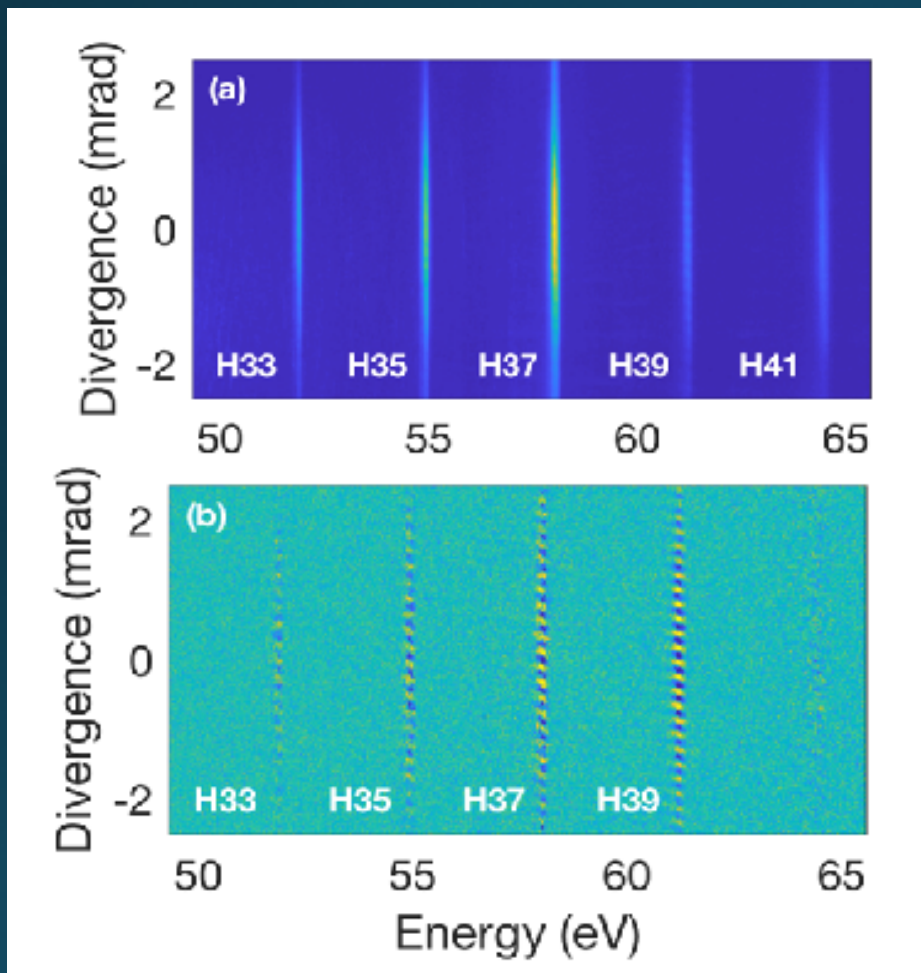


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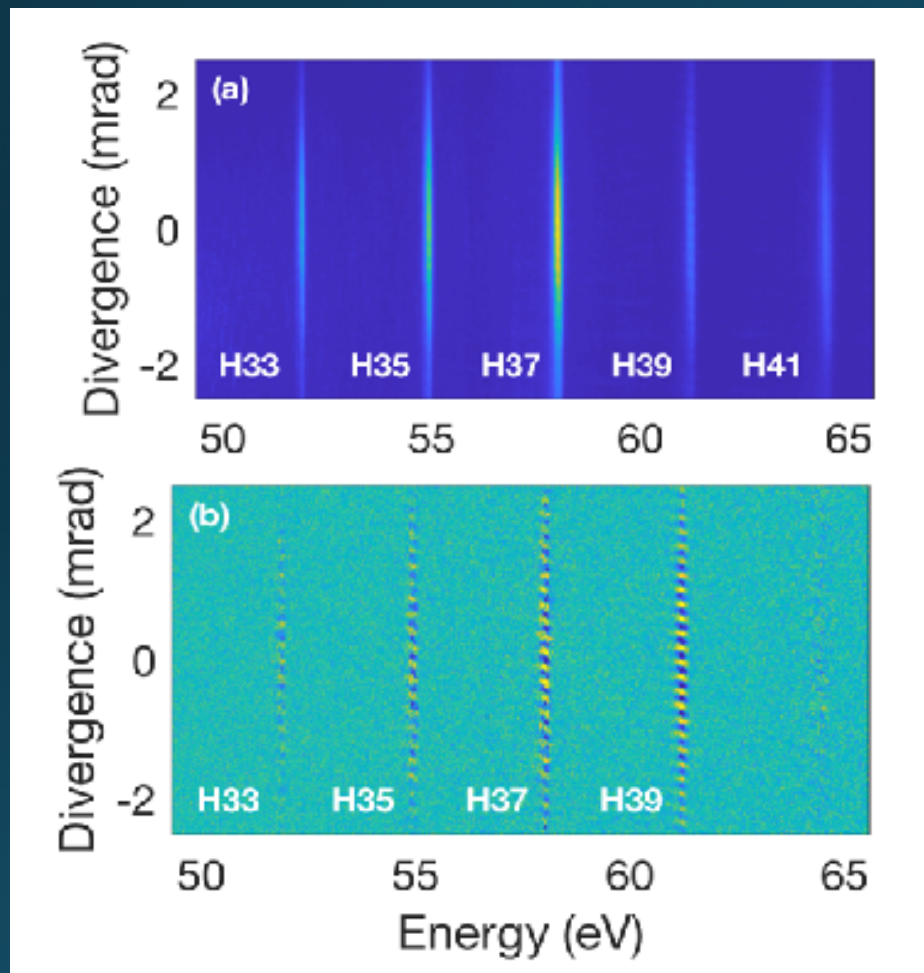
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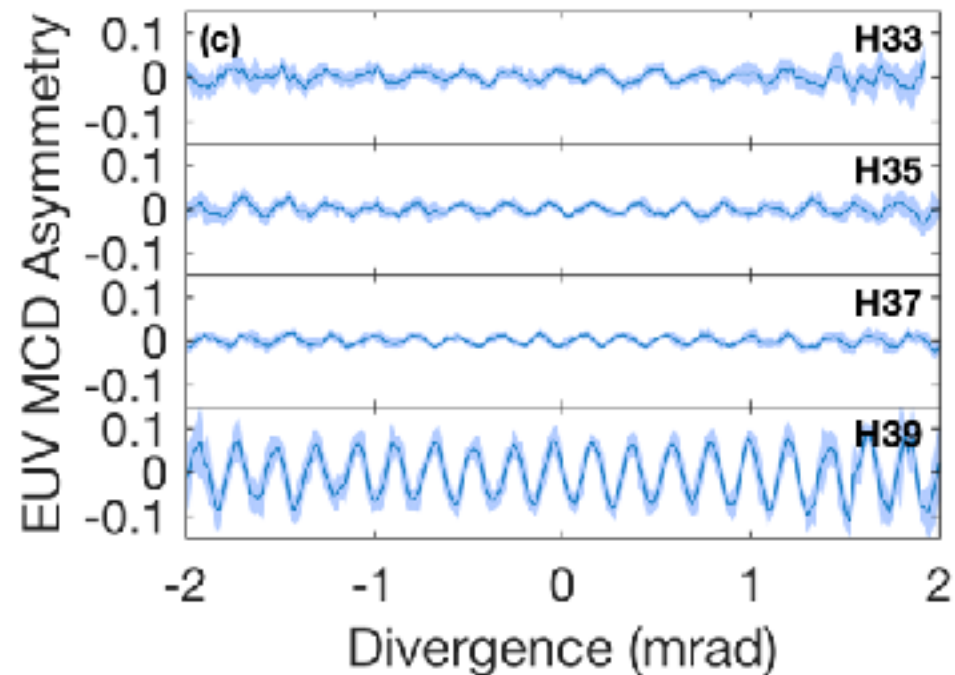


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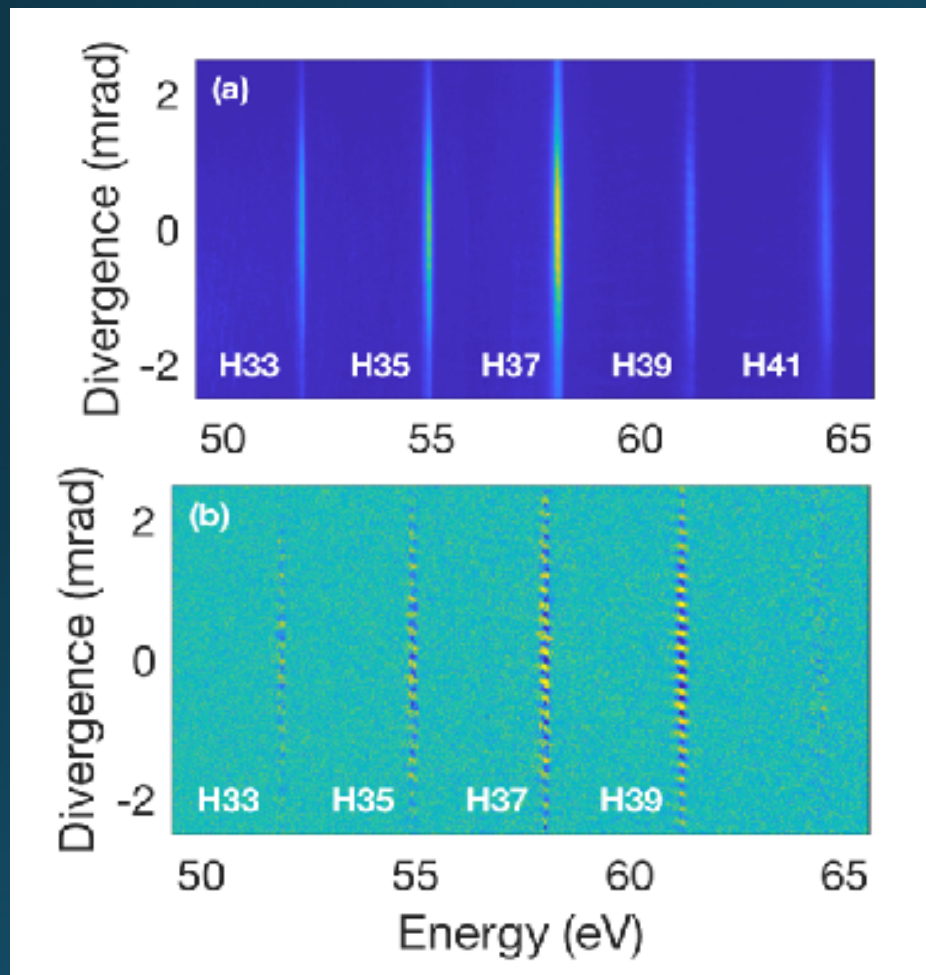
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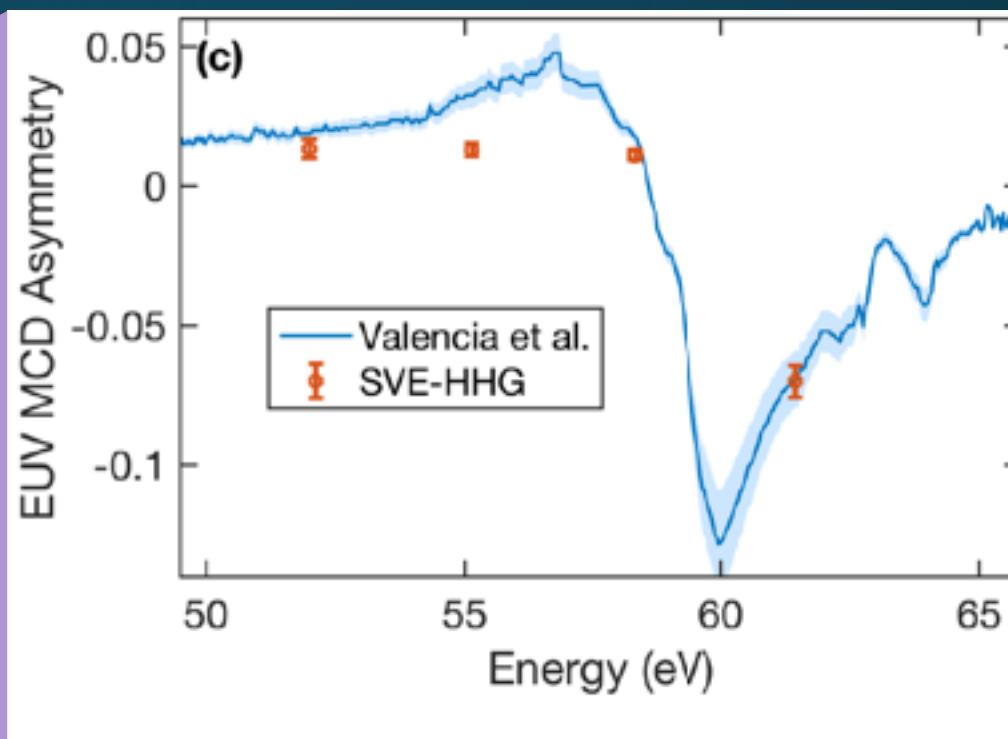


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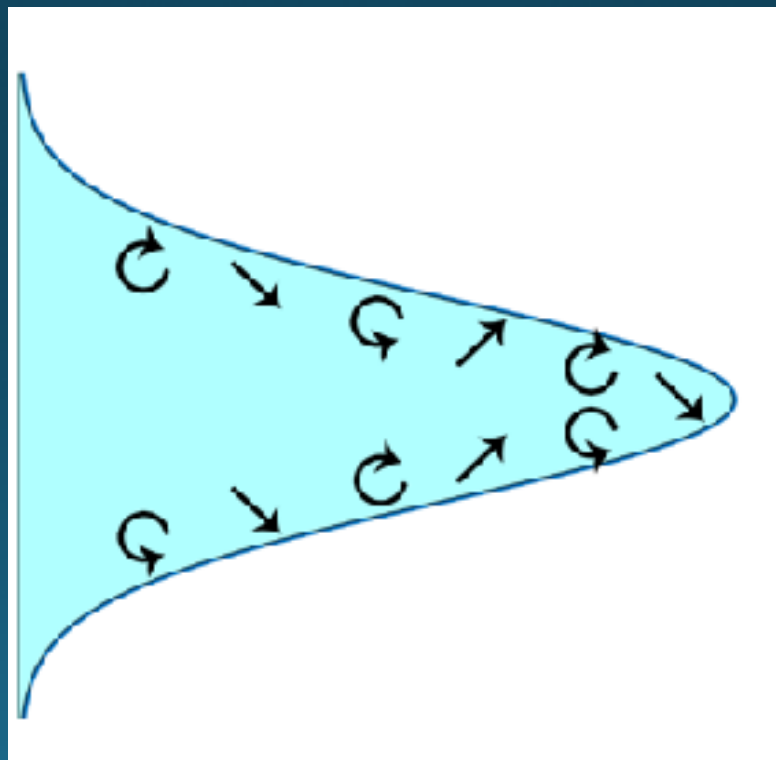


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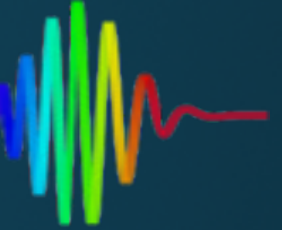
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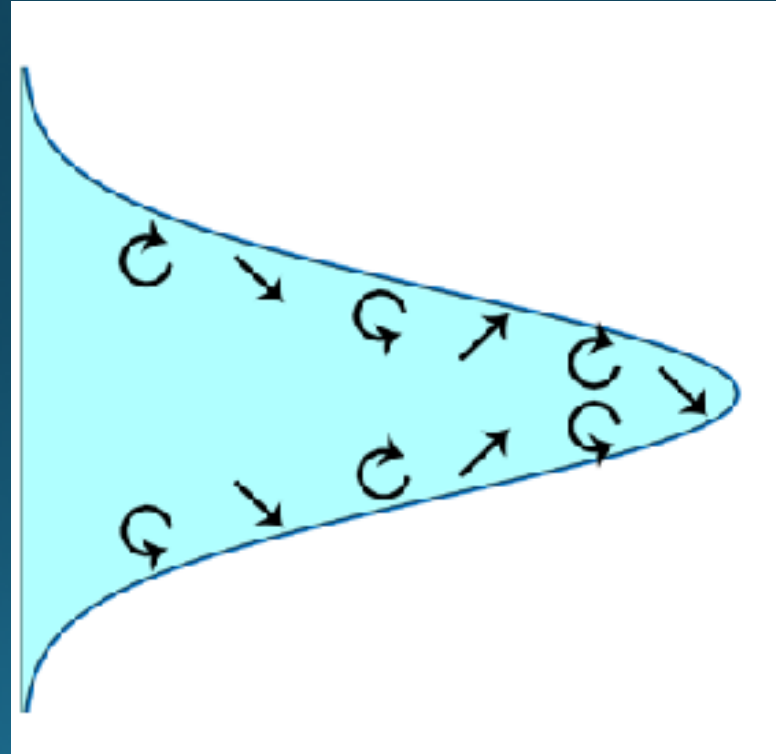
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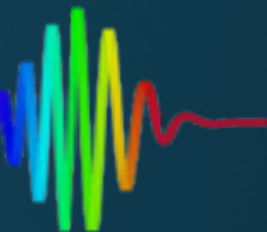
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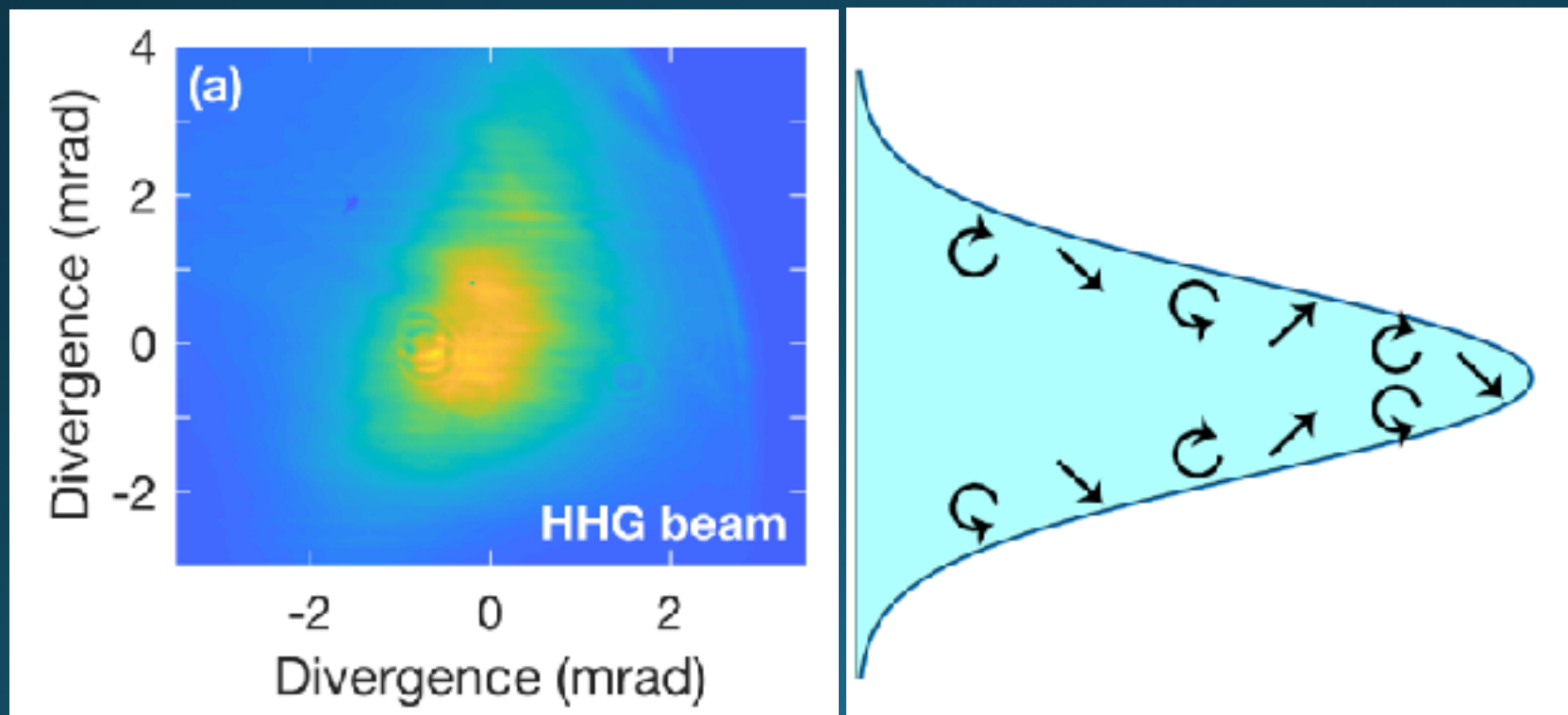
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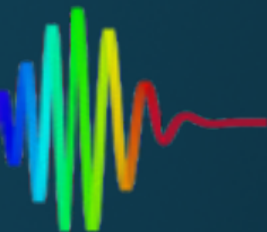
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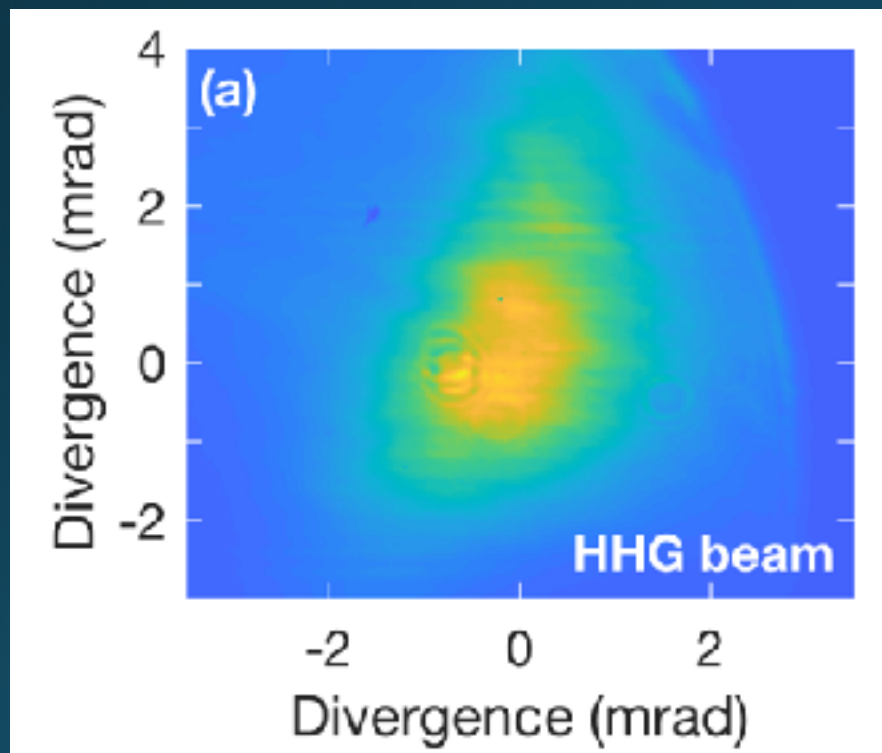
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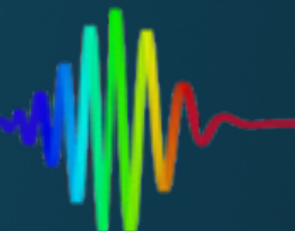
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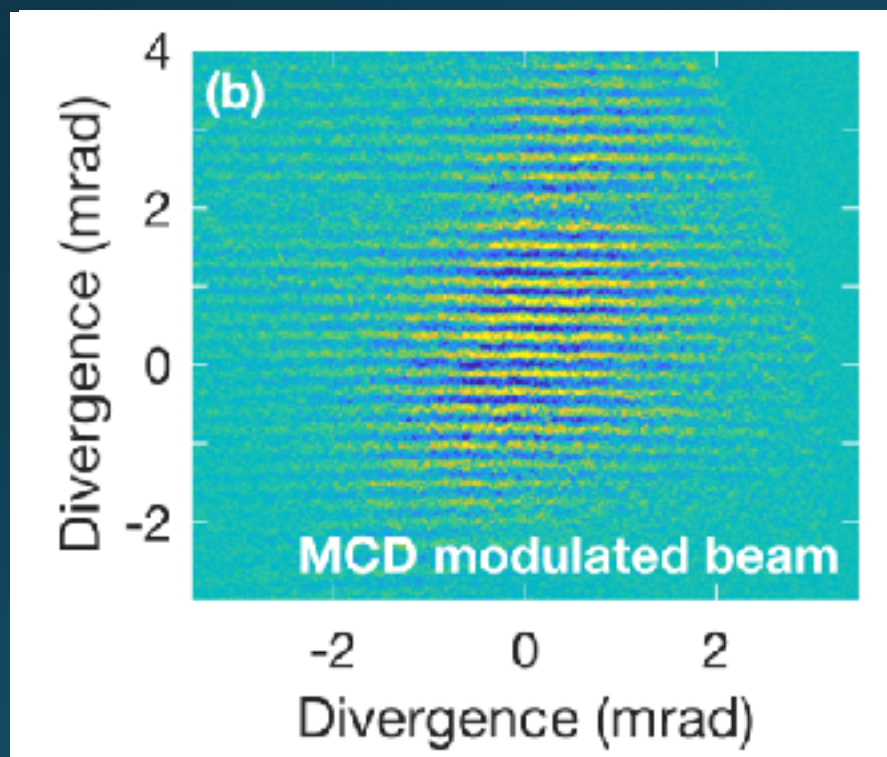
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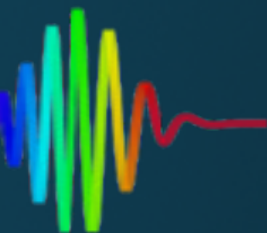
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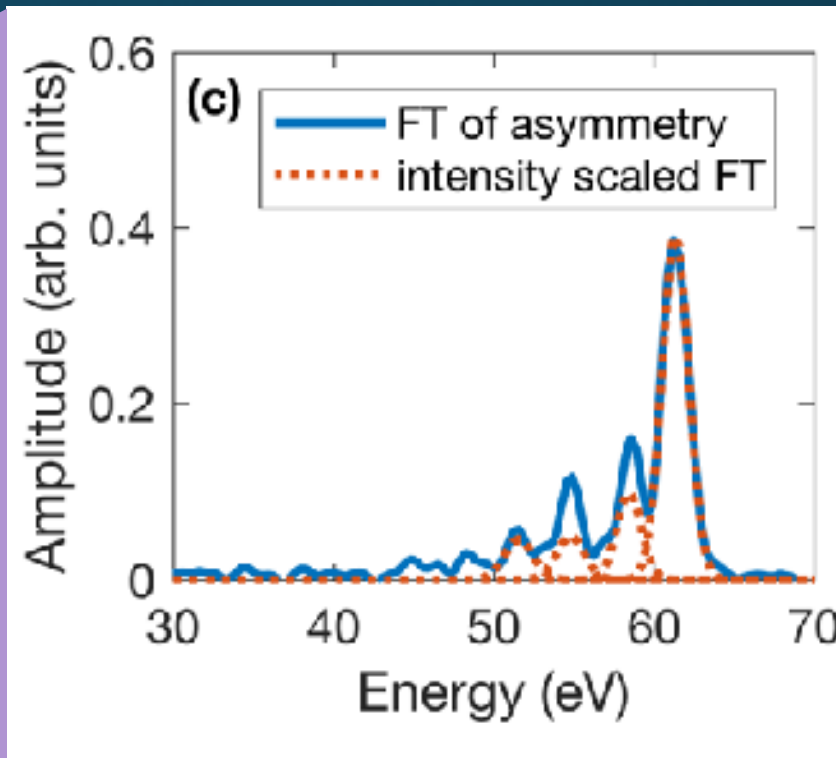
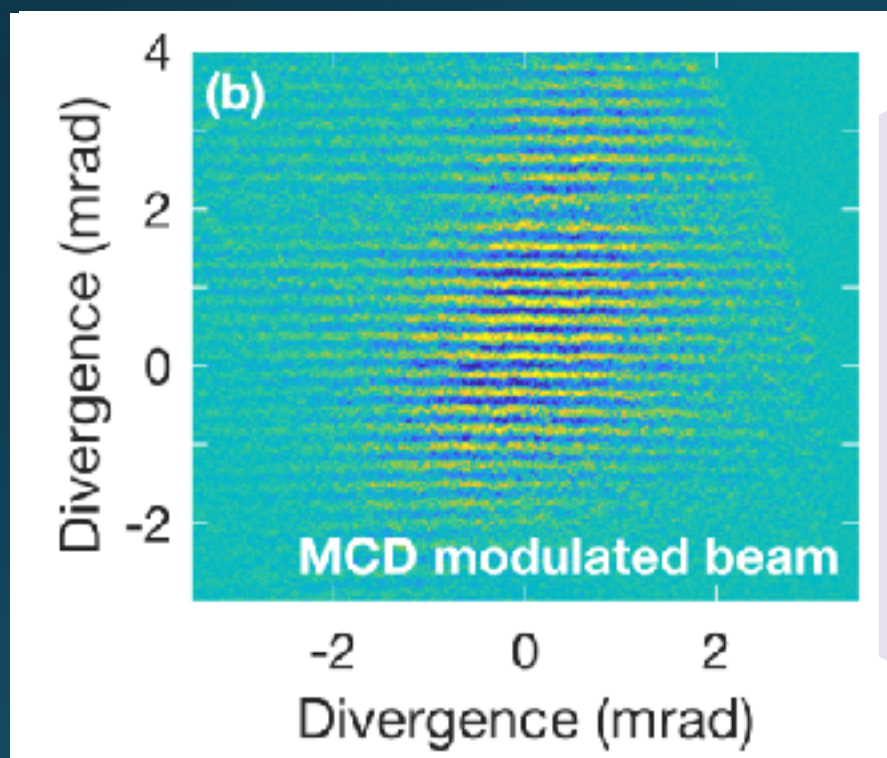
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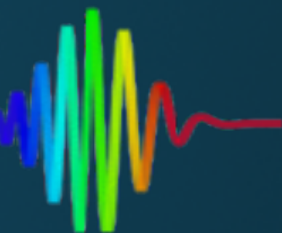
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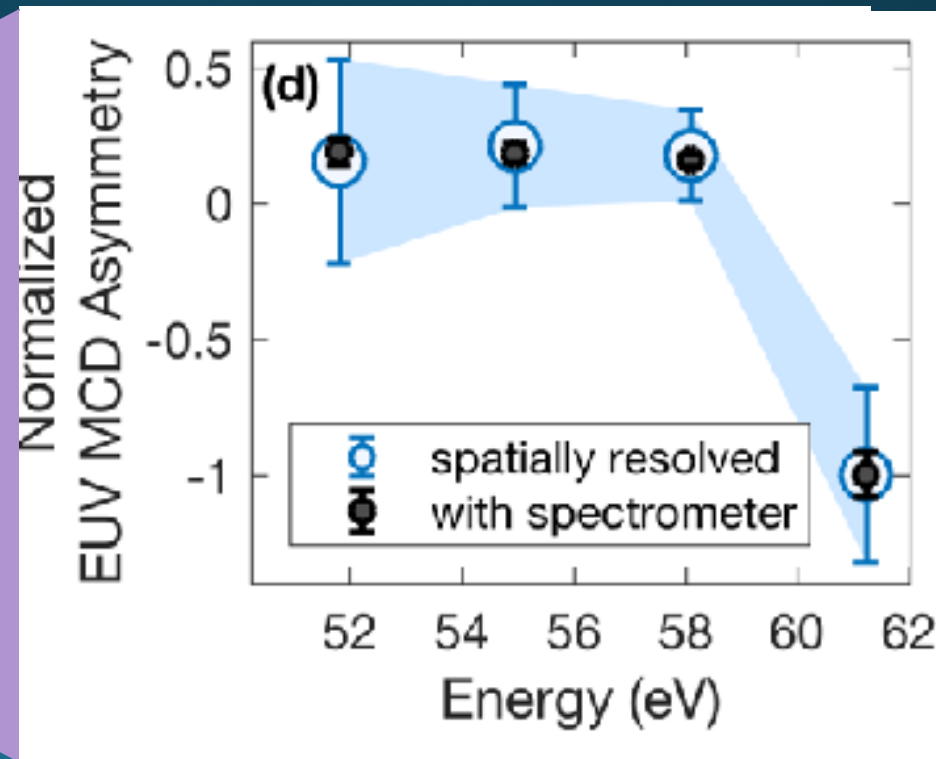
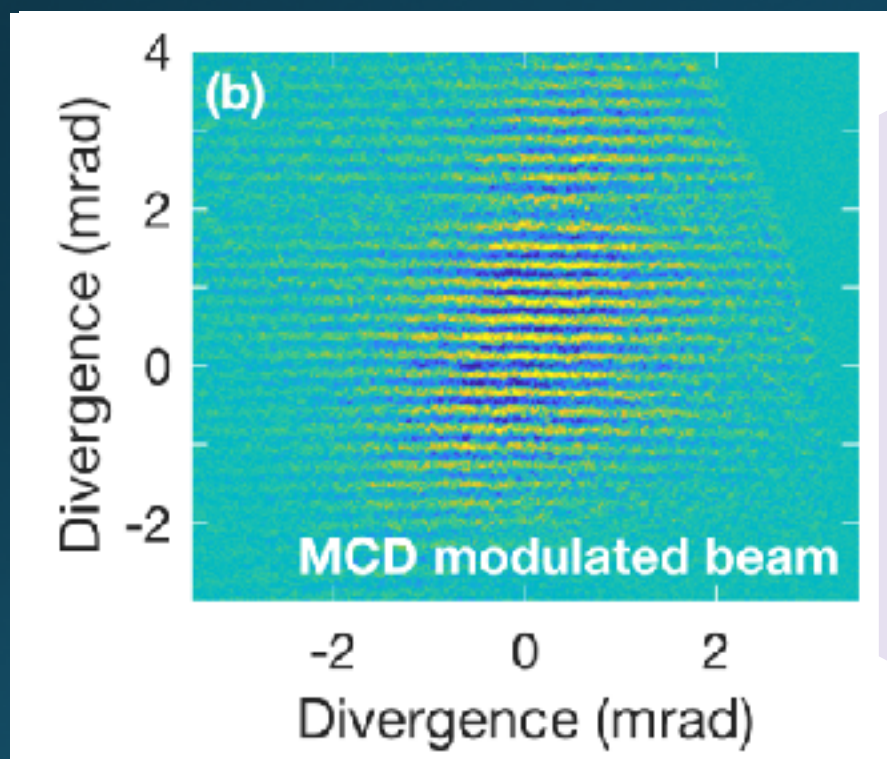
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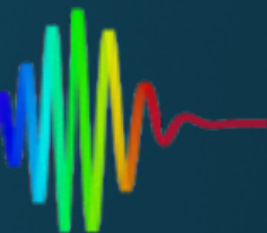
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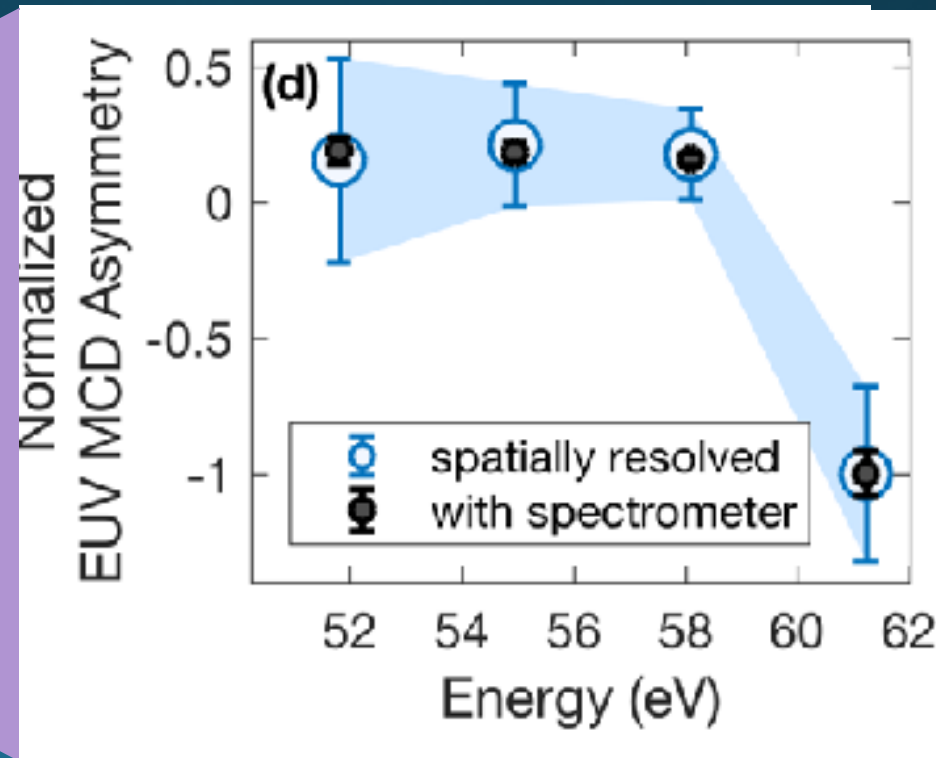
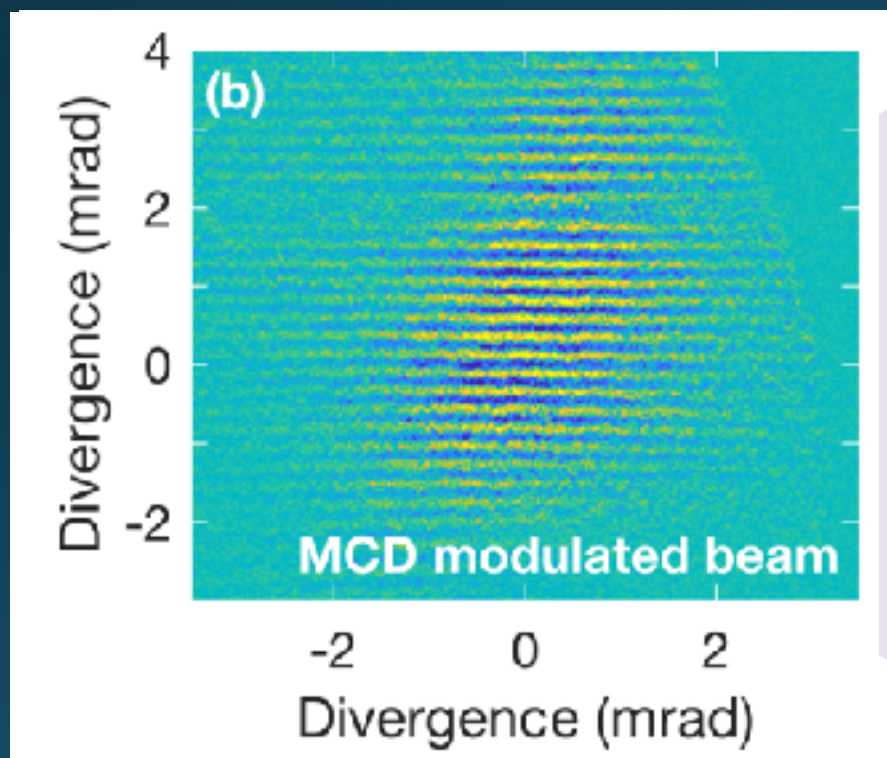


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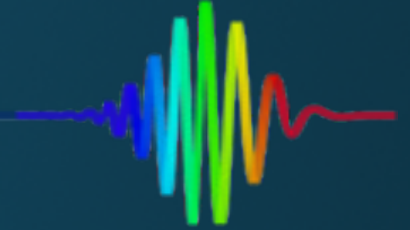
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Nearly Identical MCD Spectra from Different Samples Measured Months Apart





Temporal Evolution of Two-Focal Spot CPHHG: Supercontinuum and Isolated, Circular Attosecond Pulses!





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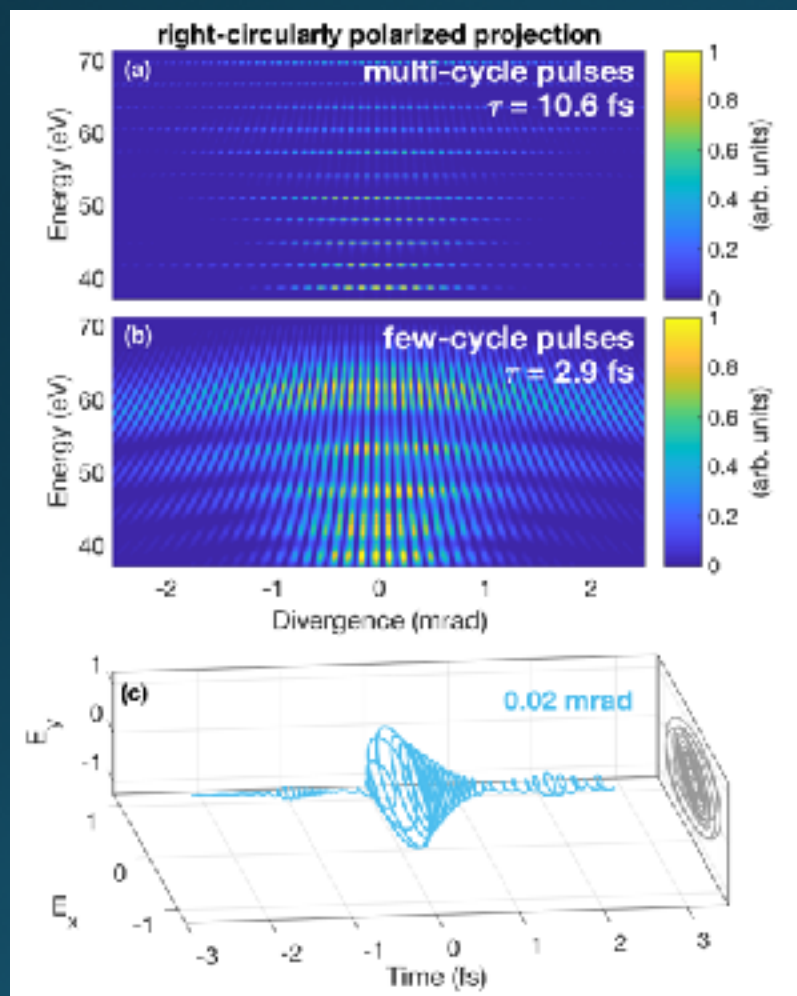


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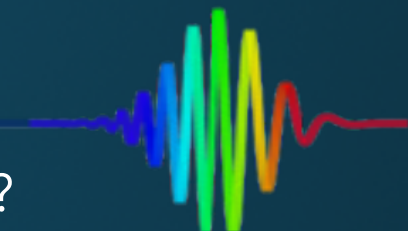
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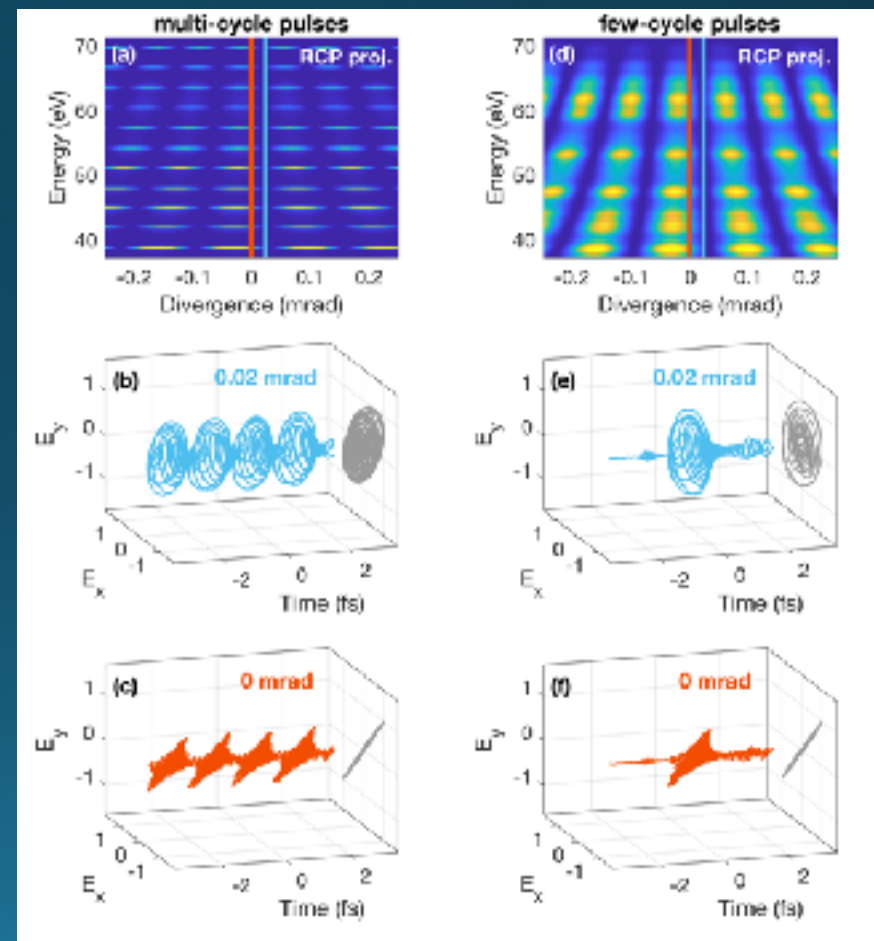
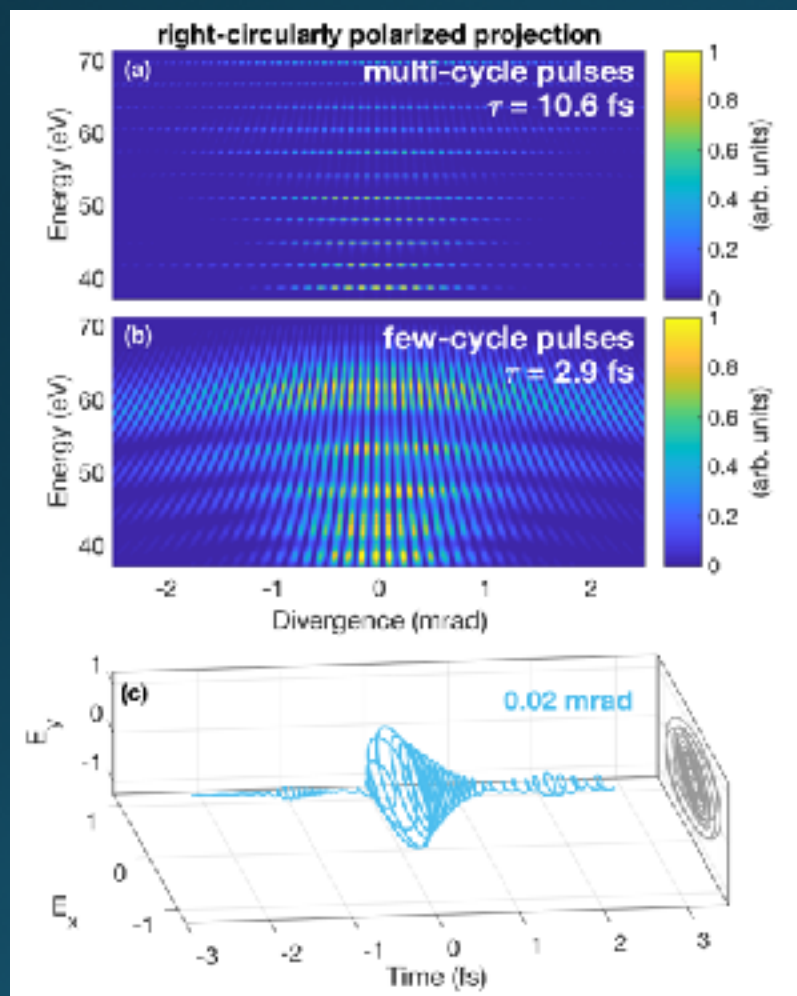
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Production and Control of Elliptically Polarized Waveforms: Versatile Light Sources for Advanced Spectroscopies





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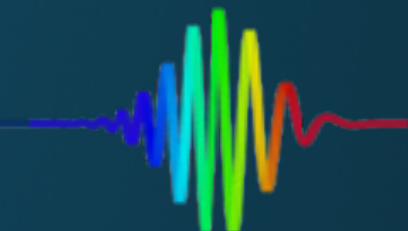


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- "On-demand" ellipticity of APTs with simple intensity mixing
- Fully compatible with existing phase-matching schemes
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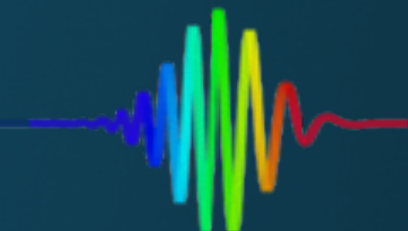
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- Bright, single-helicity CPHHG spectra
- Single-helicity CPHHG spectrum = fully circular pulse trains!
- CM-induced effects in HHG are robust to generating conditions

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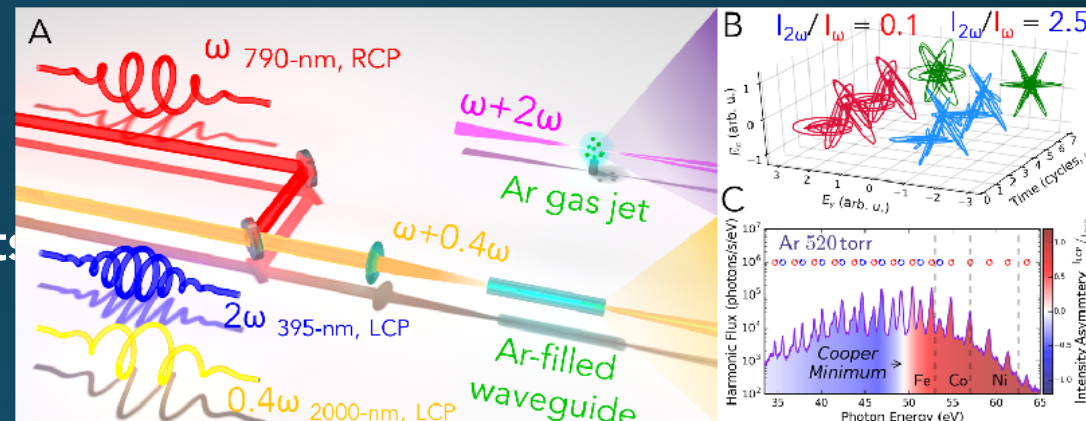


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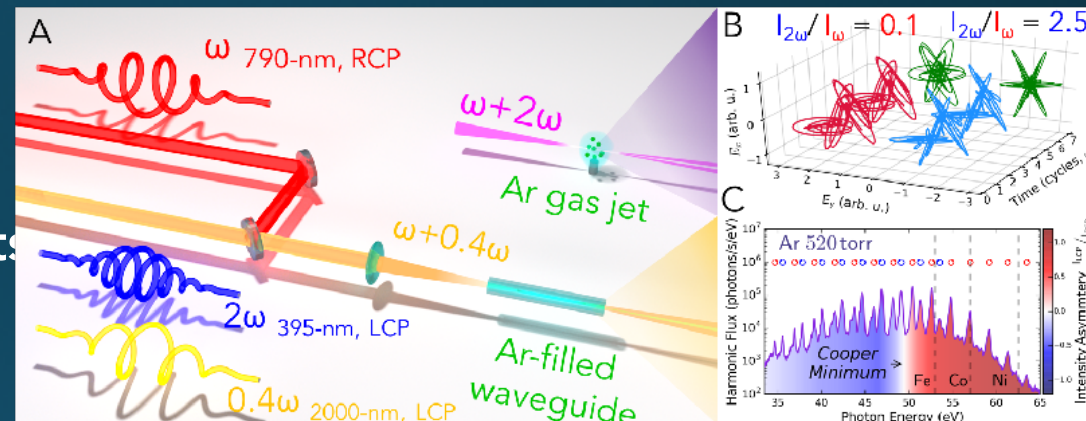


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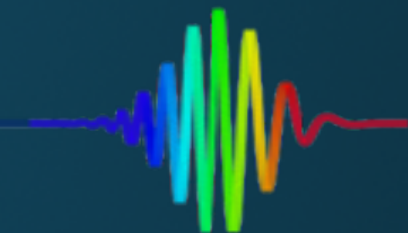
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➤ Phase-locked pulse pairs for spatio-spectral chiral spectroscopies

- Microscopic physics is identical to linear, one-color HHG
- Single arm geometry results in ultrahigh stability
- Combined with timing information, can allow for hyperspectral imaging of chiral systems

Production and Control of Elliptically Polarized Waveforms: Versatile Light Sources for Advanced Spectroscopies

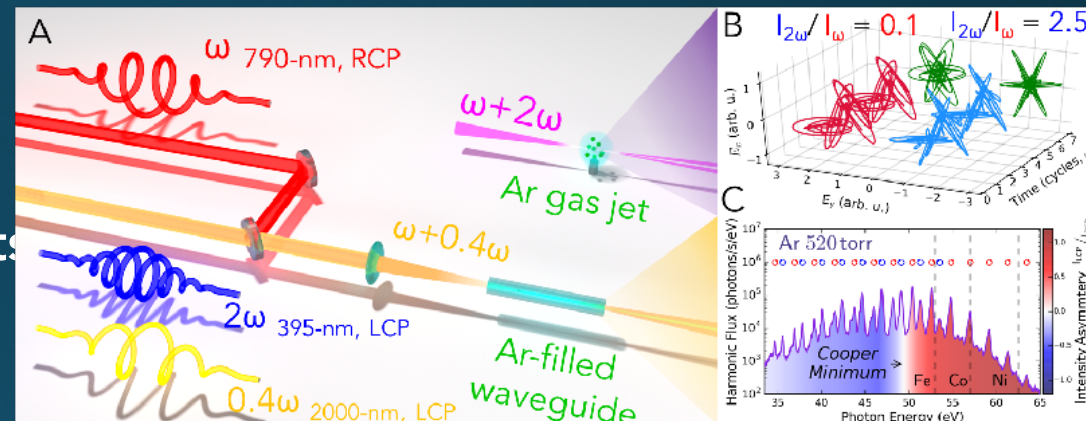


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- Fully compatible with time-gating techniques

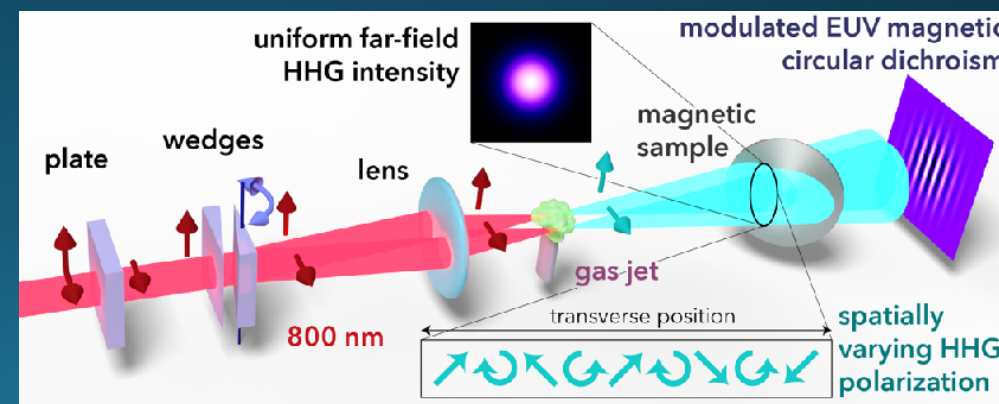
➤ Single-helicity CPHHG spectra via CM-Induced Effects

- Bright, single-helicity CPHHG spectra
- Single-helicity CPHHG spectrum = fully circular pulse trains!
- CM-induced effects in HHG are robust to generating conditions



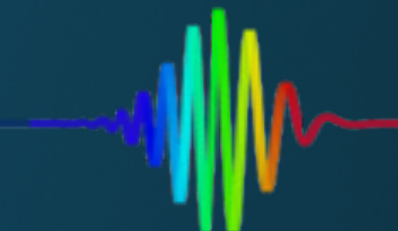
➤ Phase-locked pulse pairs for spatio-spectral chiral spectroscopies

- Microscopic physics is identical to linear, one-color HHG
- Single arm geometry results in ultrahigh stability
- Combined with timing information, can allow for hyperspectral imaging of chiral systems





Thanks for Listening to a Very, Very, Very
Loud and Energetic **Researcher** 😊



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