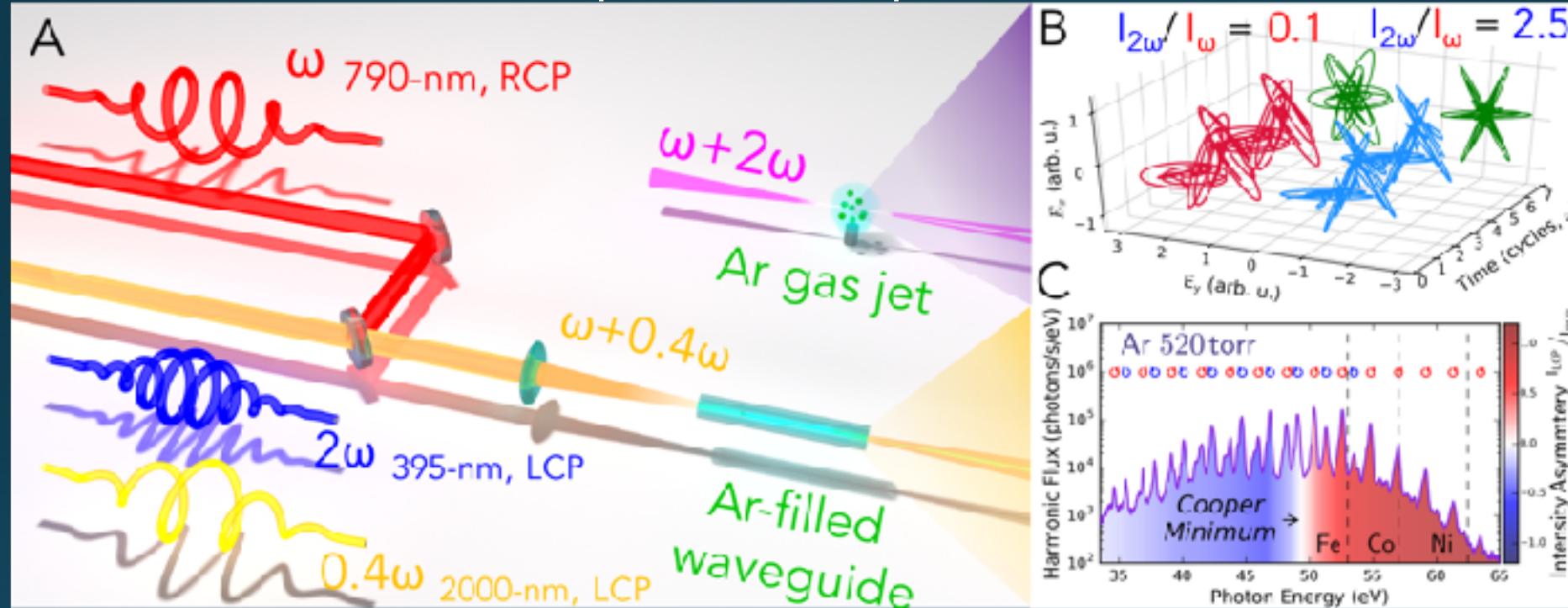


Helicity Gets a New Twist: Straightforward Production of Polarization Sculpted High-Harmonic Attosecond Waveforms for Chiral Spectroscopies and Imaging

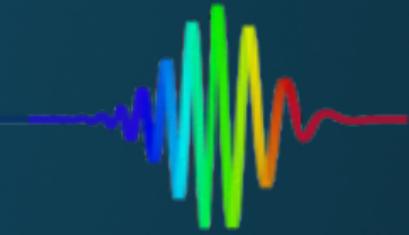


Kevin M. Dorney

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

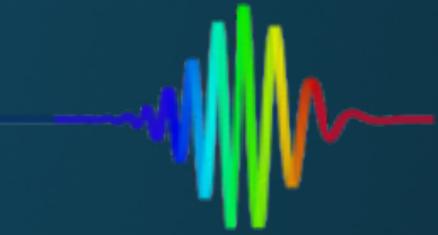


From Snowy Mountains to Warm... Beautiful...
Snow-covered Beaches?





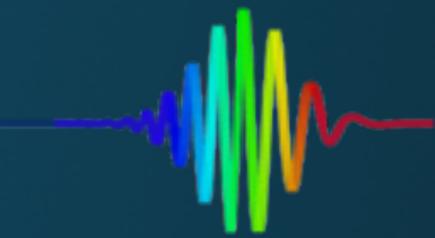
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Boulder, CO (10-02-2018)



From Snowy Mountains to Warm... Beautiful...
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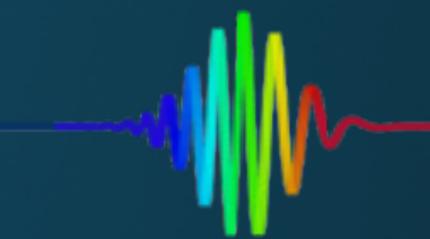


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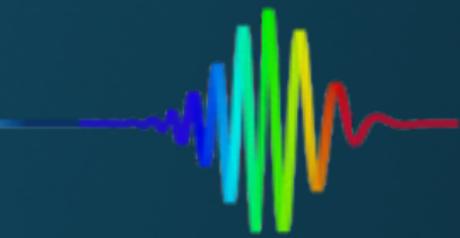


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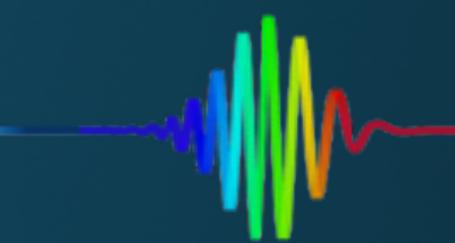
Boulder, CO (10-02-2018)



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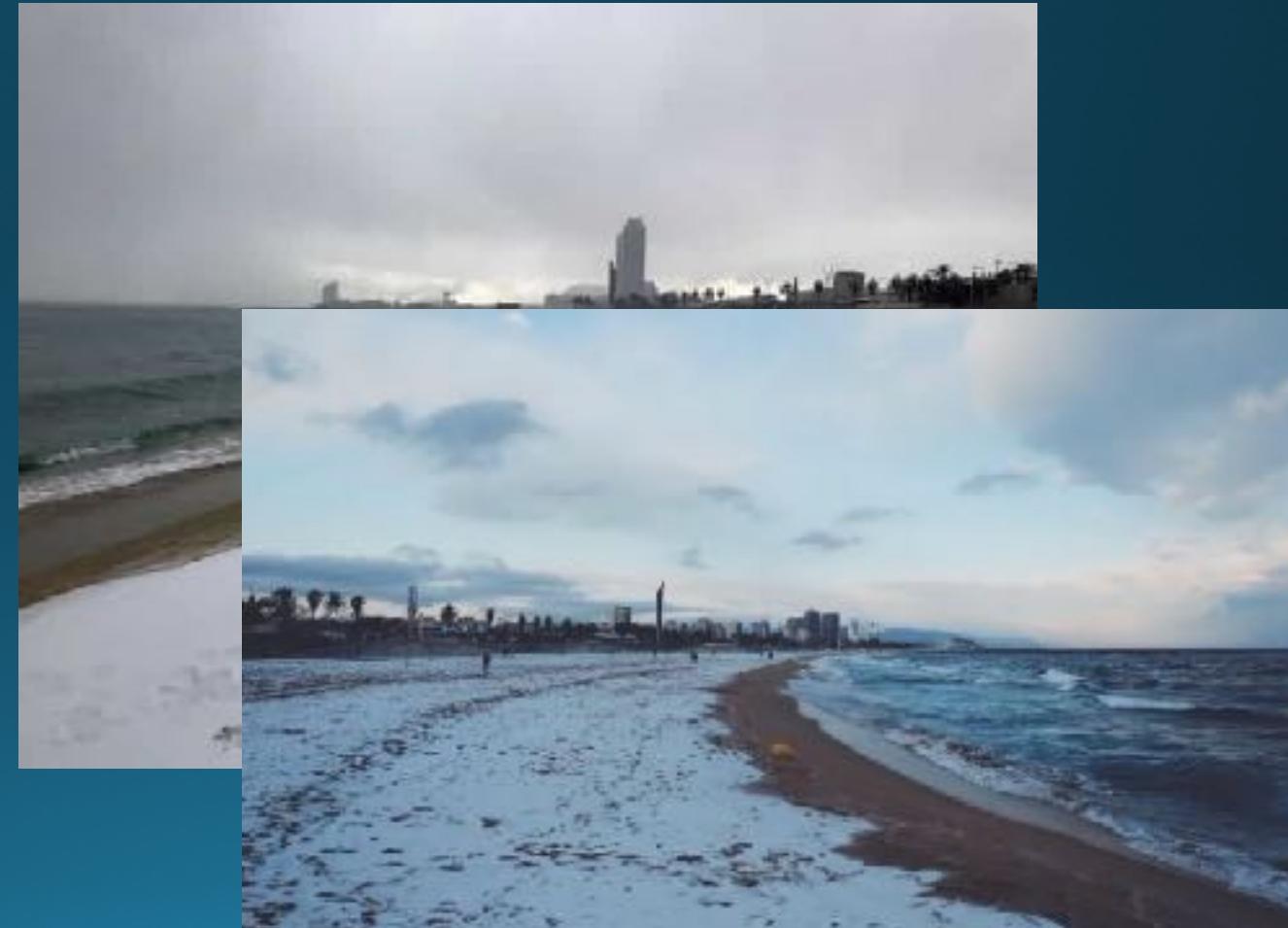
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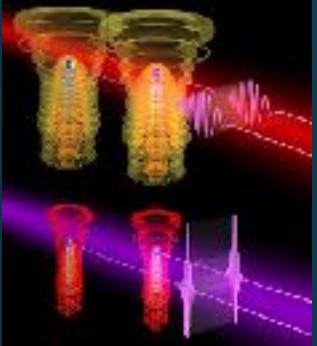
Barcelona, ESP (10-02-2018)



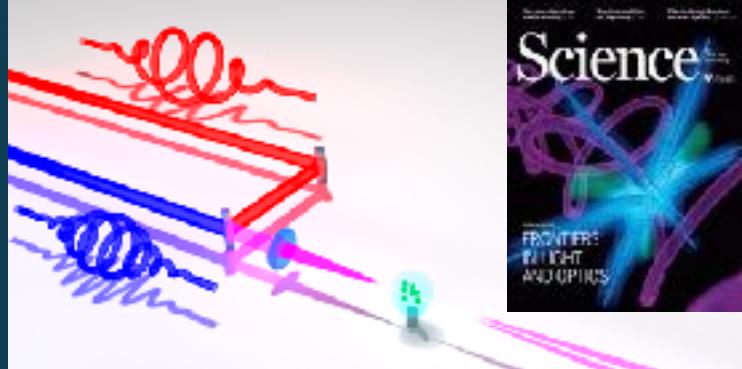
Light and Materials Science in the KM Group: AMO Dynamics at Extreme Spatial and Temporal Scales



Attosecond Extreme Nonlinear Optics



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



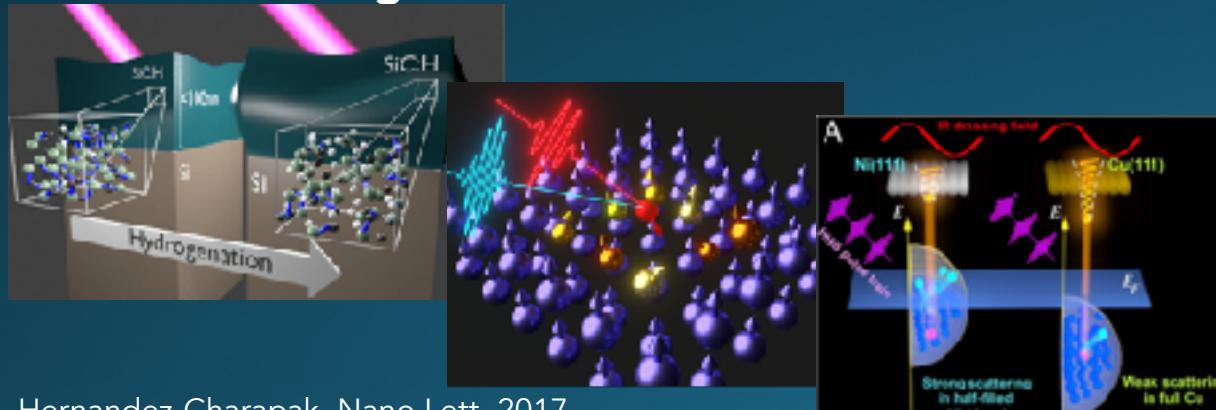
Coherent X-ray Imaging



Gardner, 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

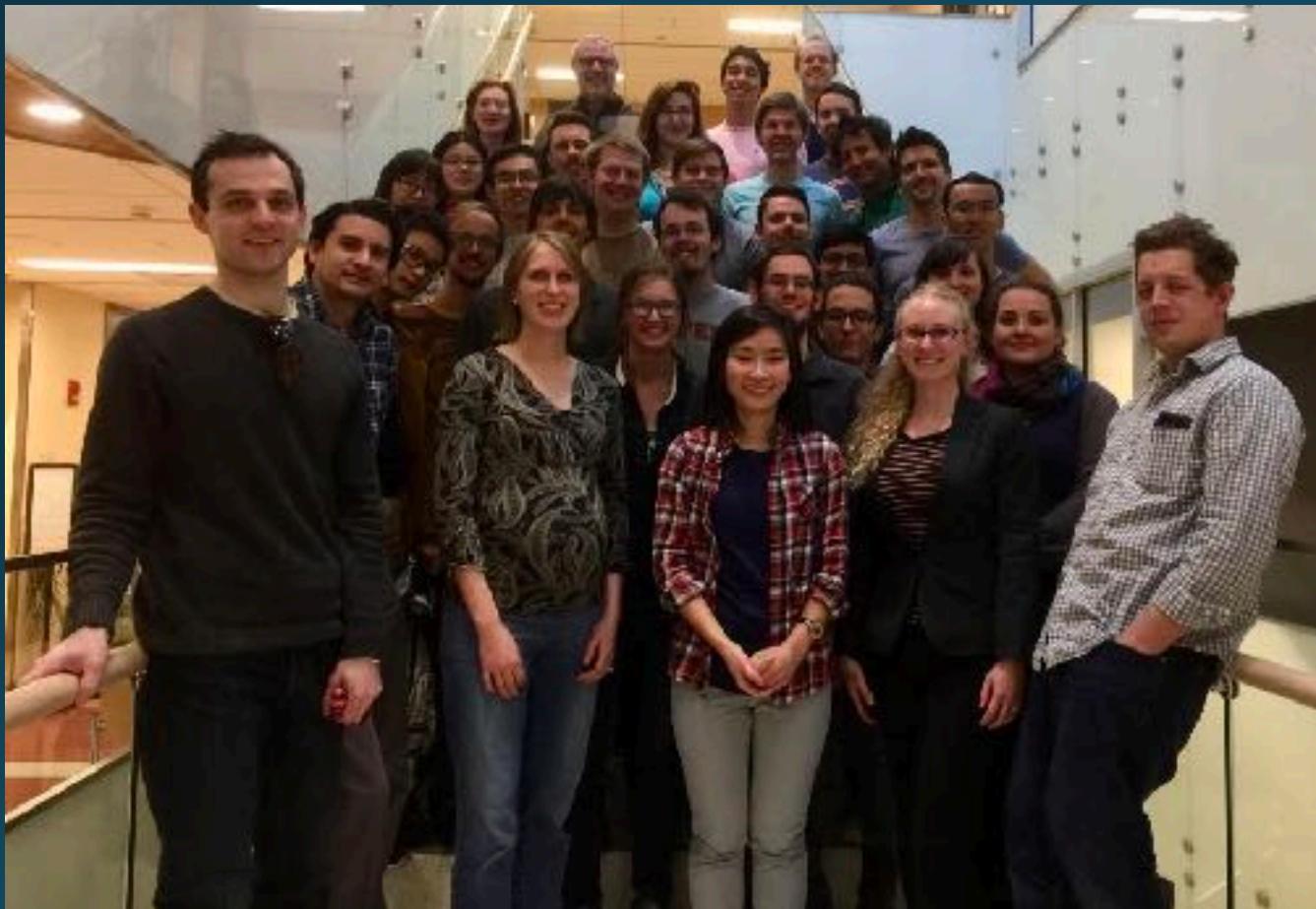


Hoogeboom-Pot, PNAS 112, 2015
Ellis, JACS, 11 2015

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



KM Group Spring 2017



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Collaborators



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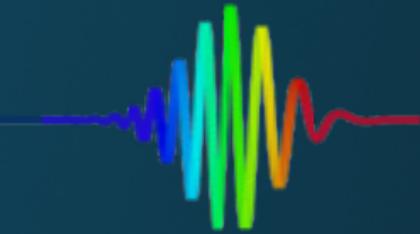


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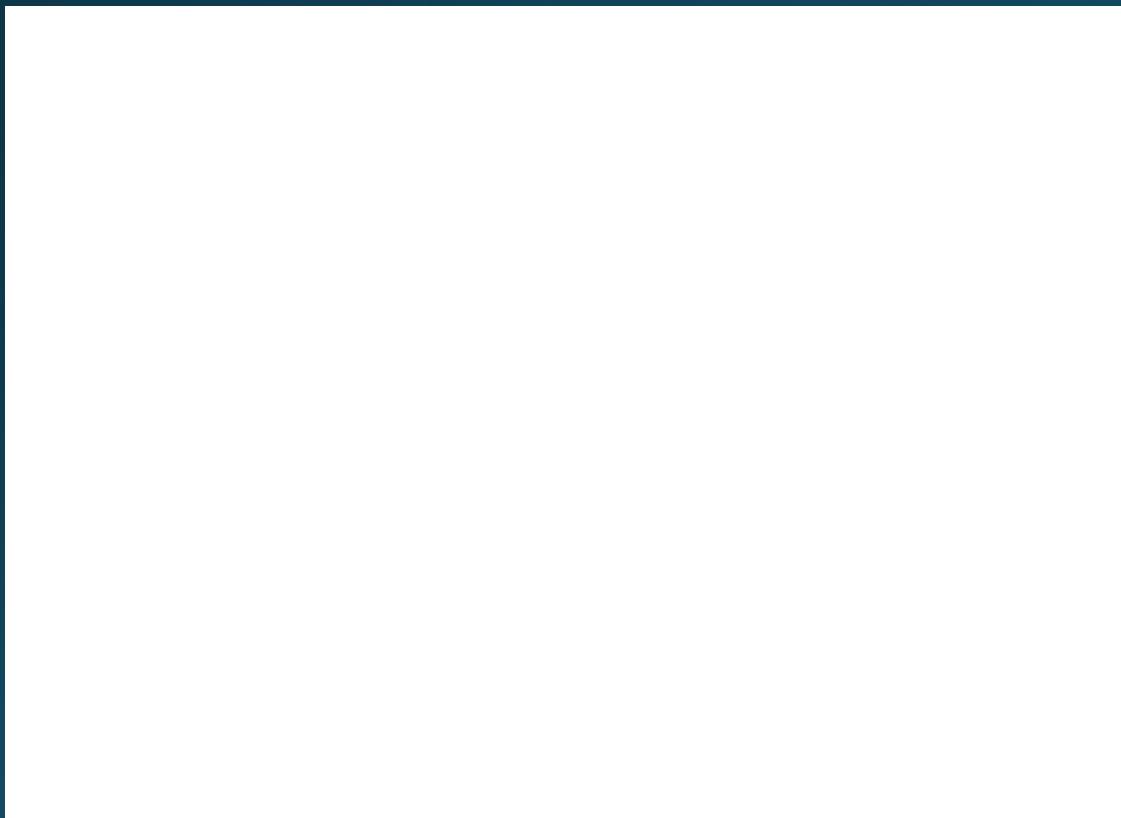
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The White Whale of the Physical Sciences

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

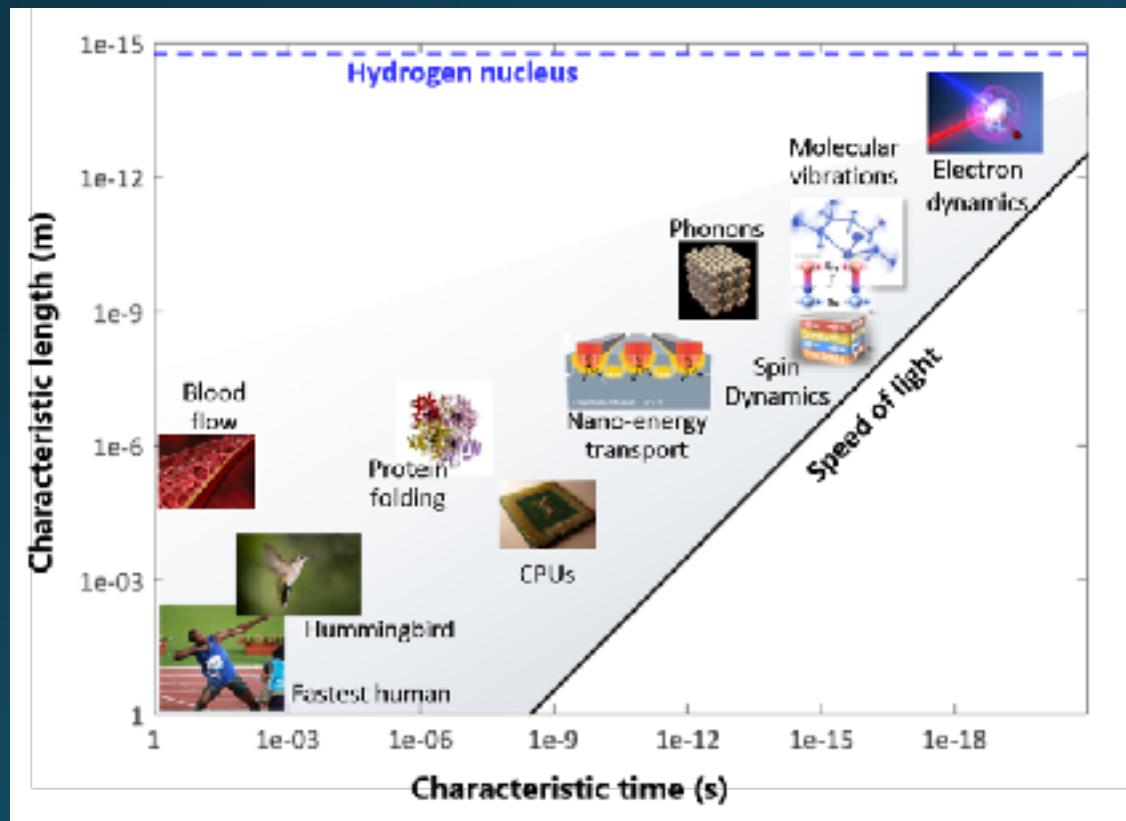


Courtesy: Nico Hernandez Chupak, KM Group



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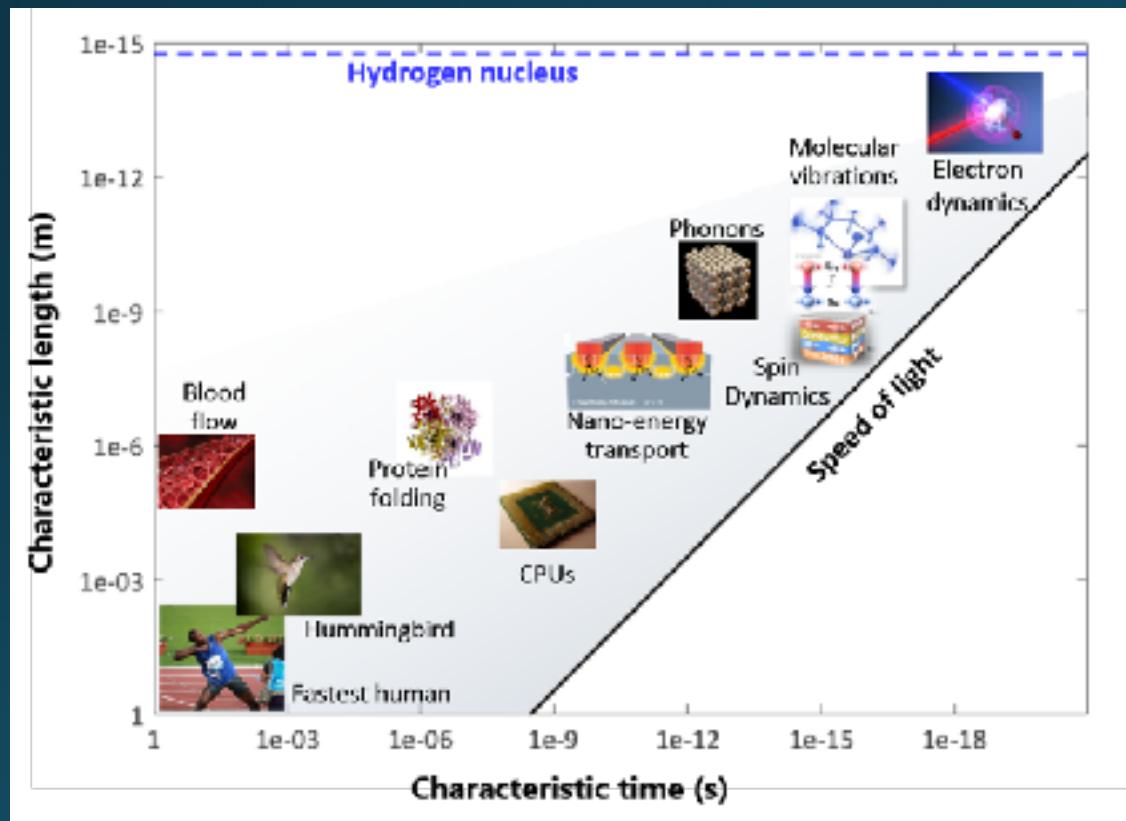


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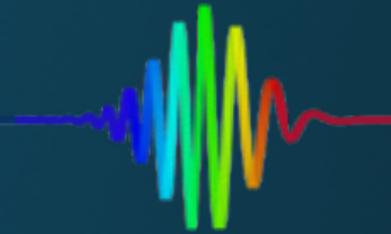


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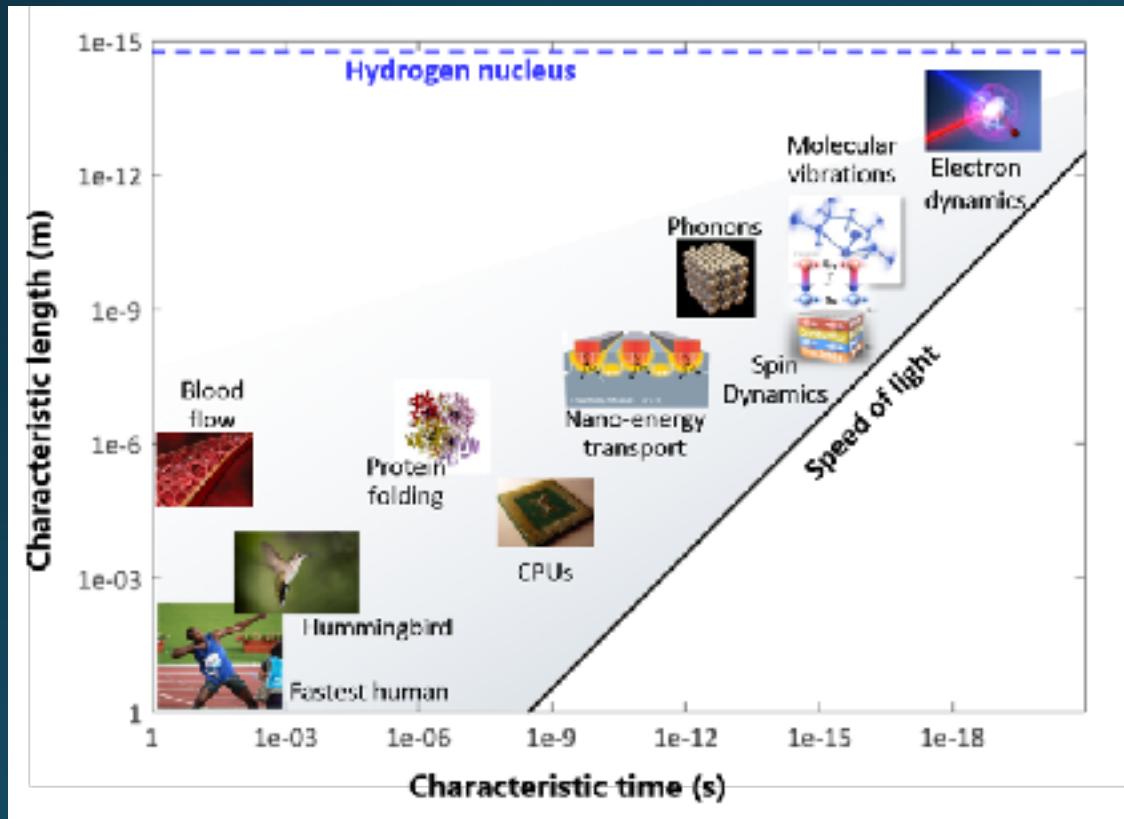


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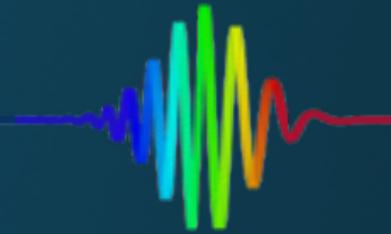


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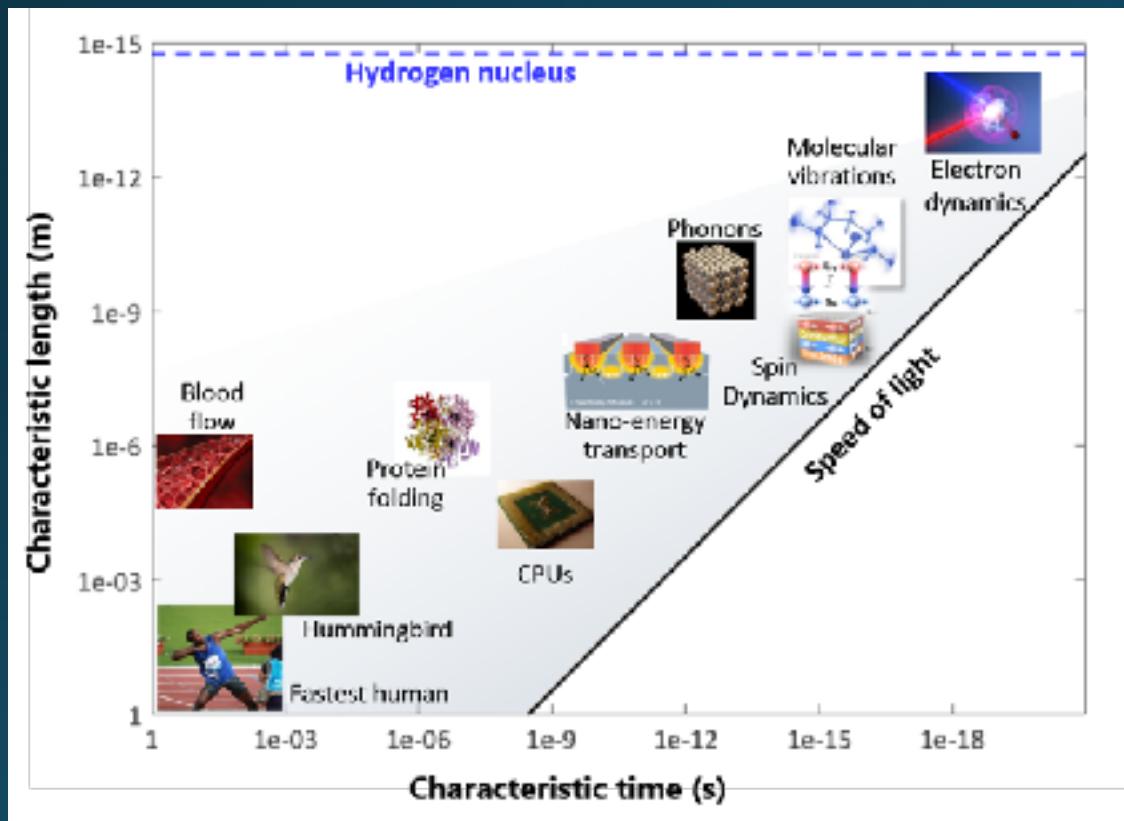


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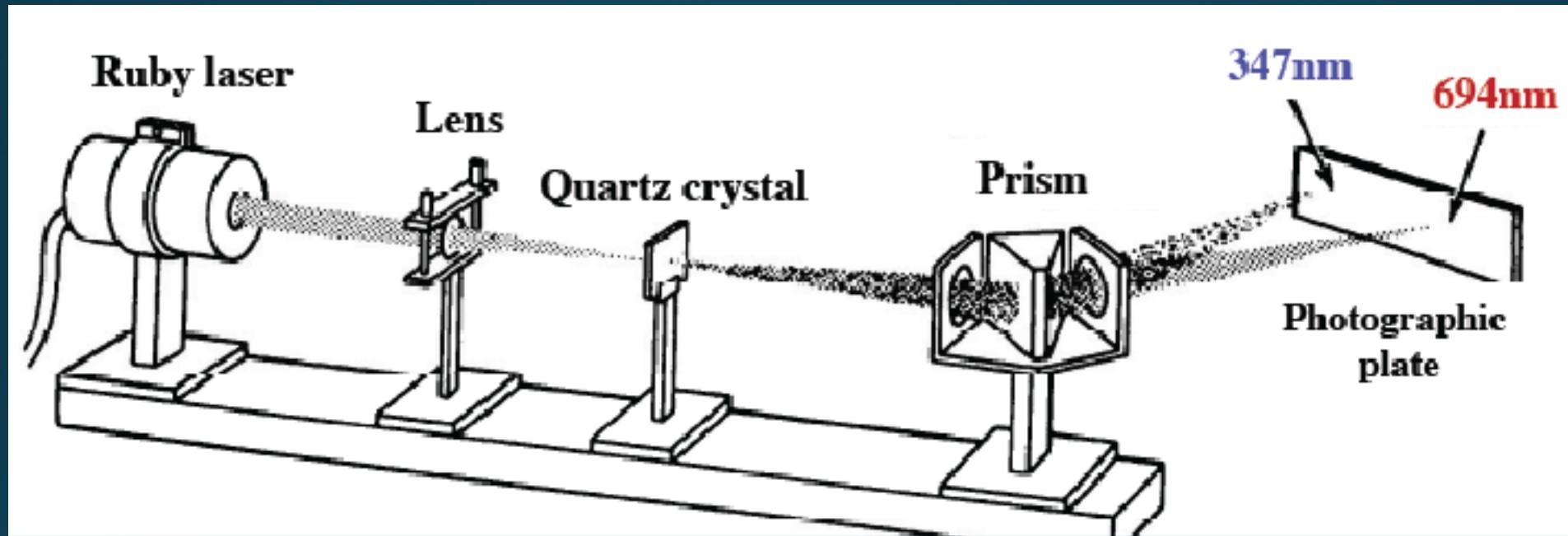
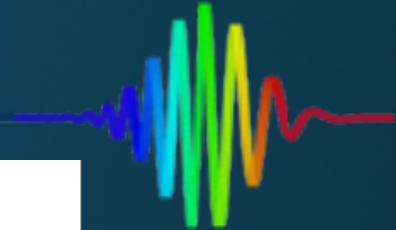
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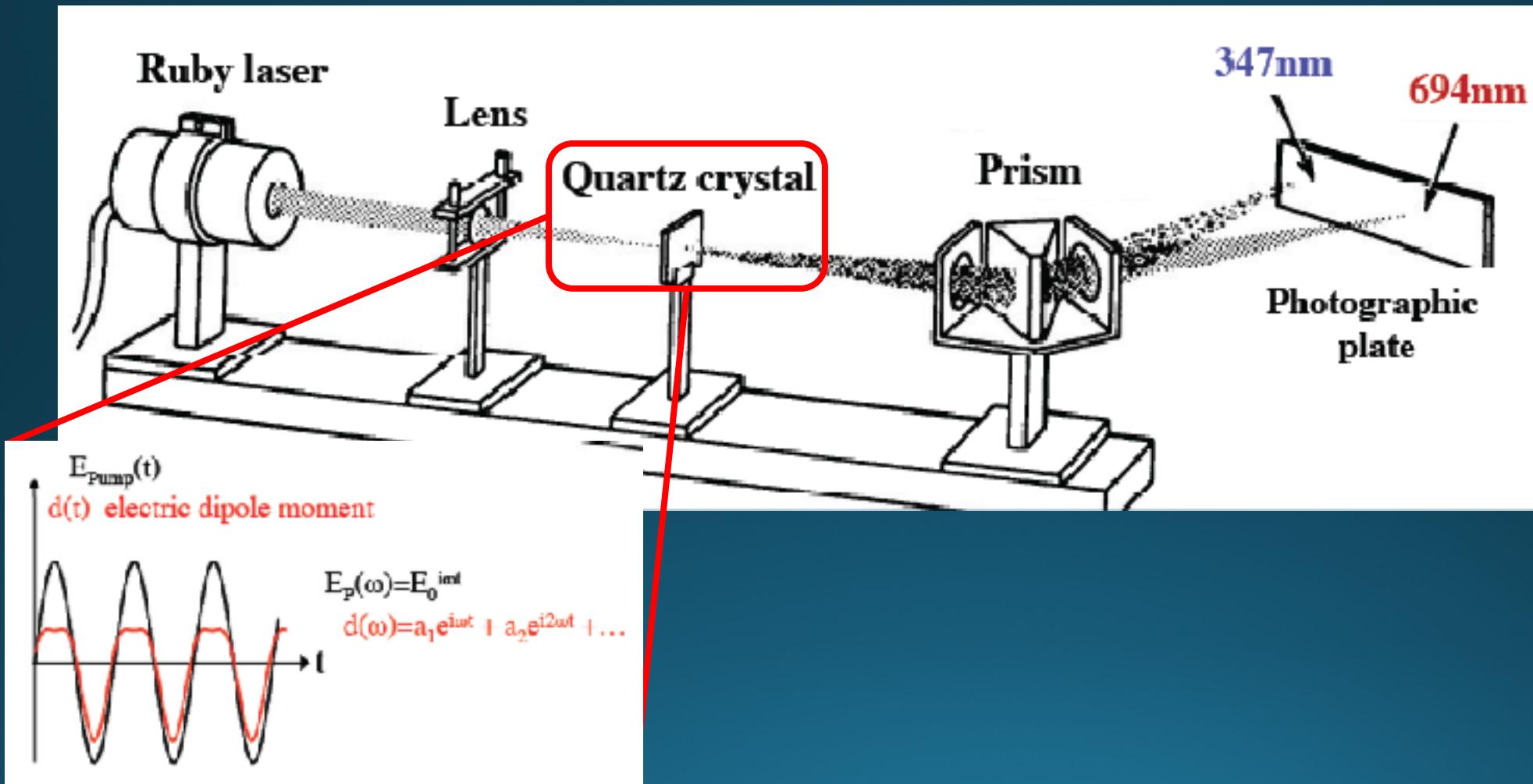
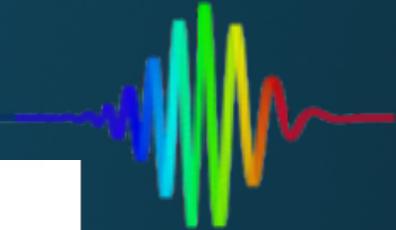


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HHG in the Beginning: A Barely Visible "Smudge" Revolutionizes Optical Science

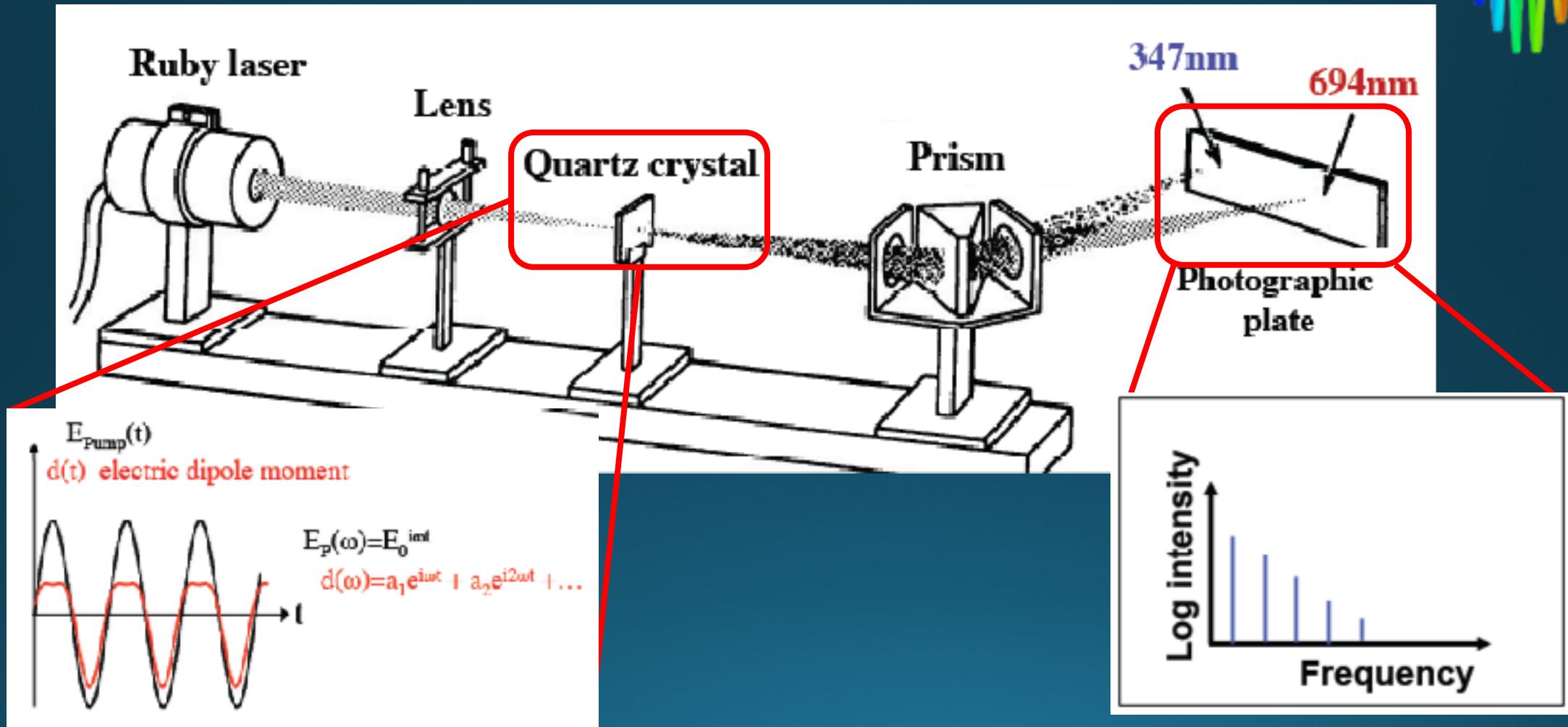
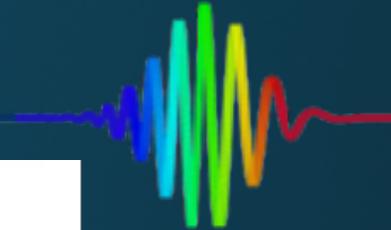


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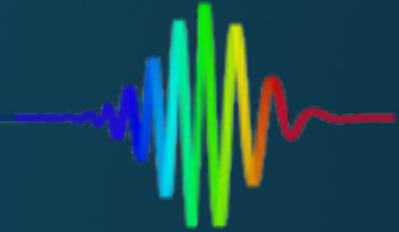
Franken et al. PRL, 7, 1961

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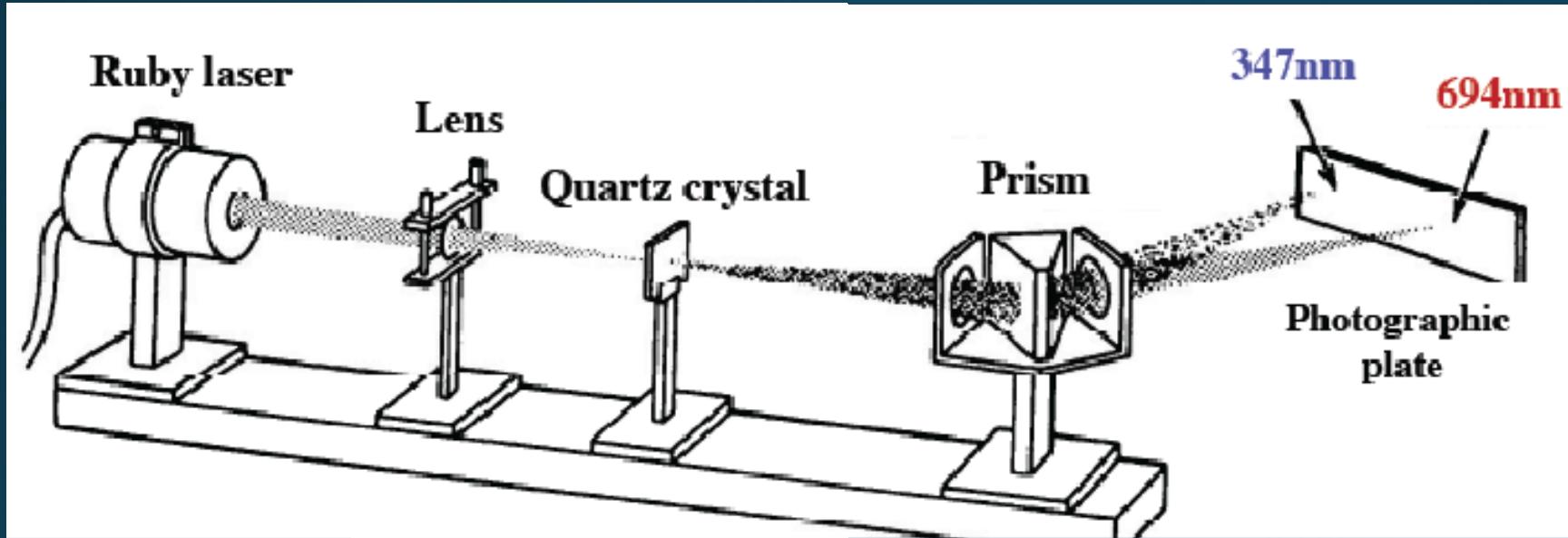


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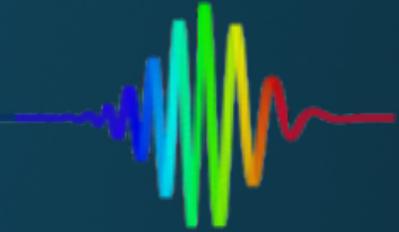
Evolution of HHG: Perturbative Optics to Extreme Nonlinear Optical Science



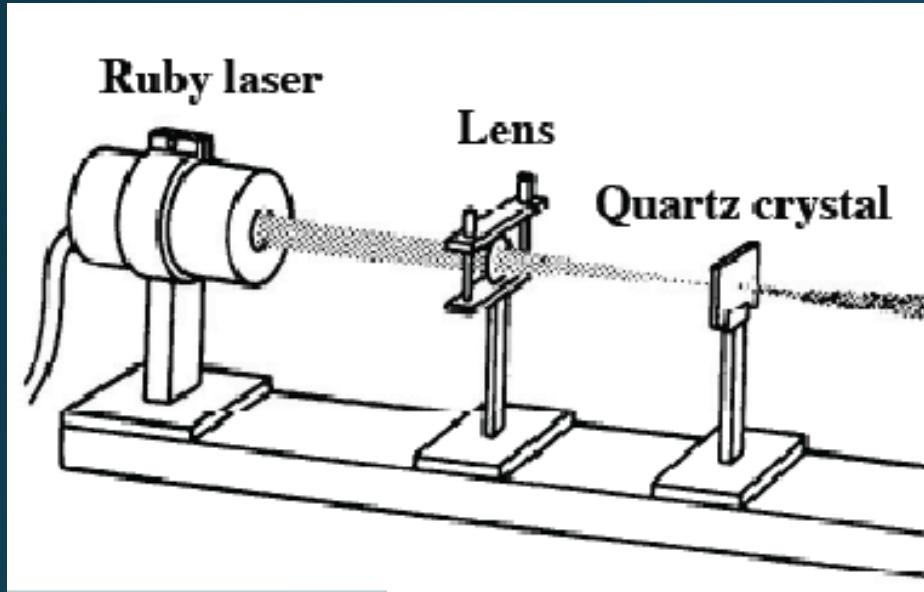
- From perturbative optics to extreme nonlinear optical science.



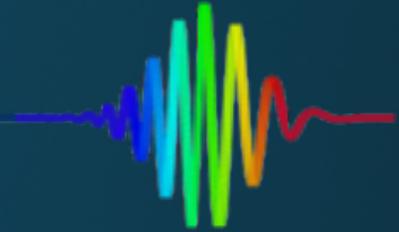
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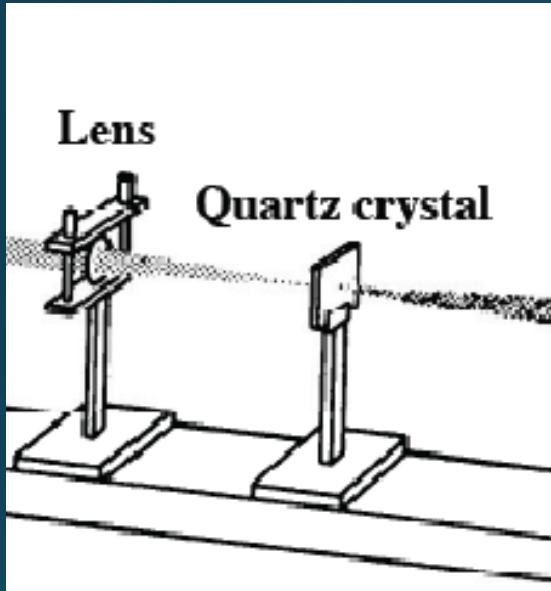
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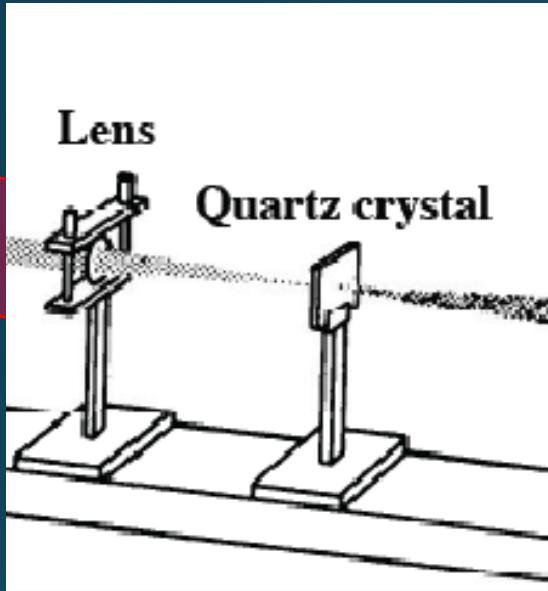
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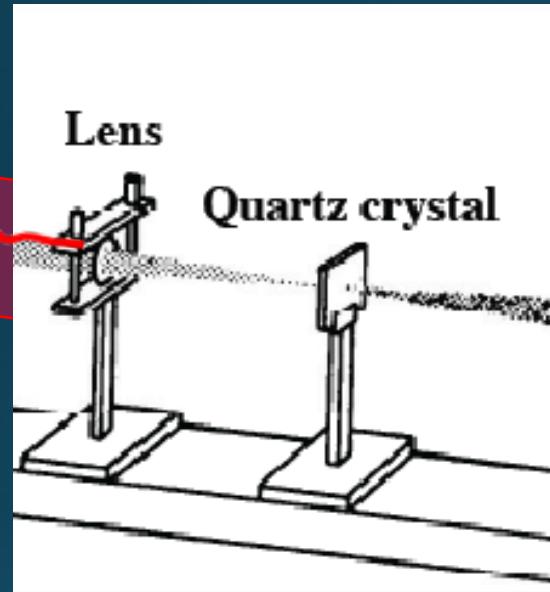
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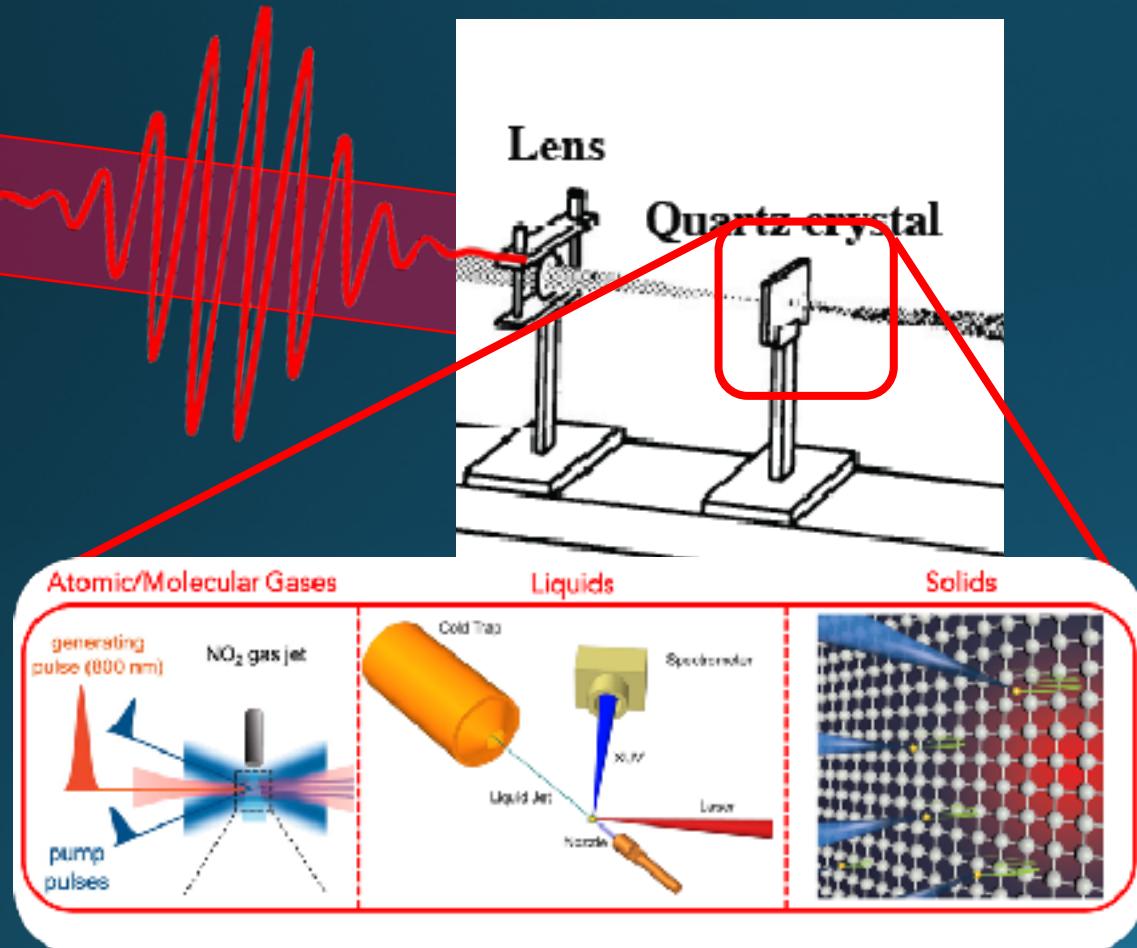
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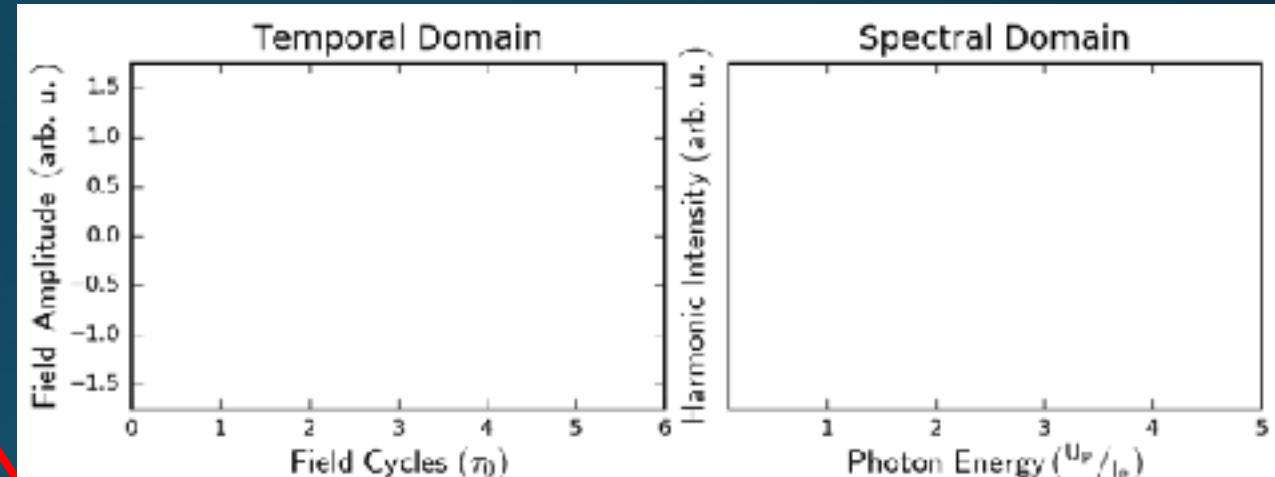
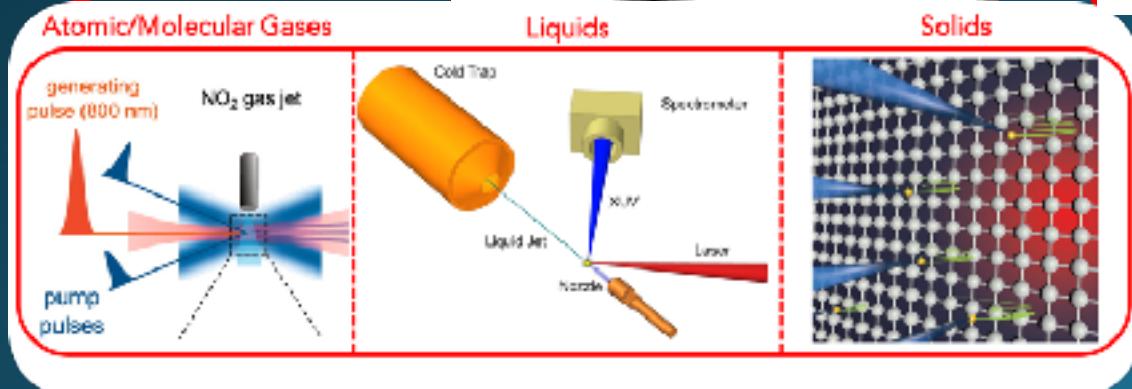
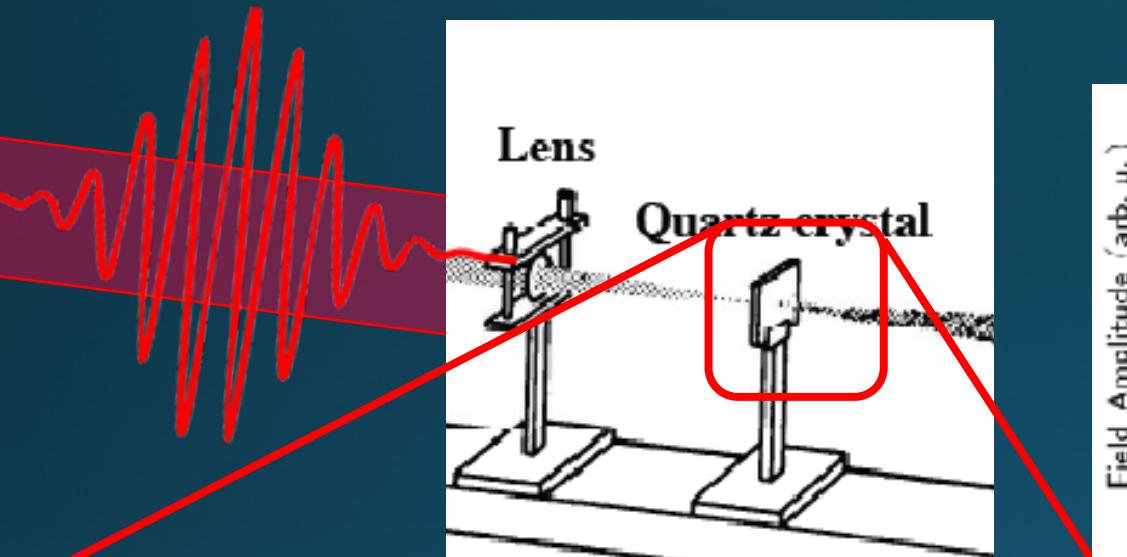
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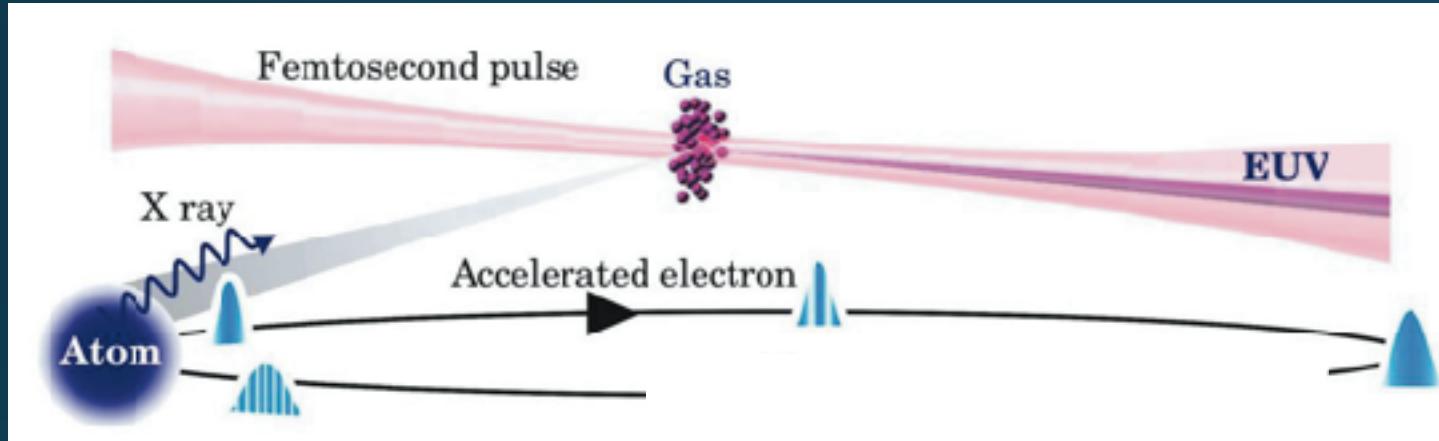
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Microscopic Mechanism of HHG: Epitome of Classical Correspondence Principle



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



Kuchiev, JETP, 45. 404 (1987)

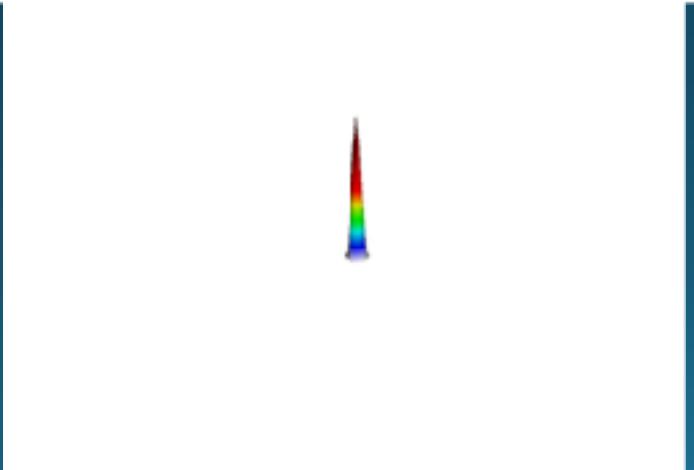
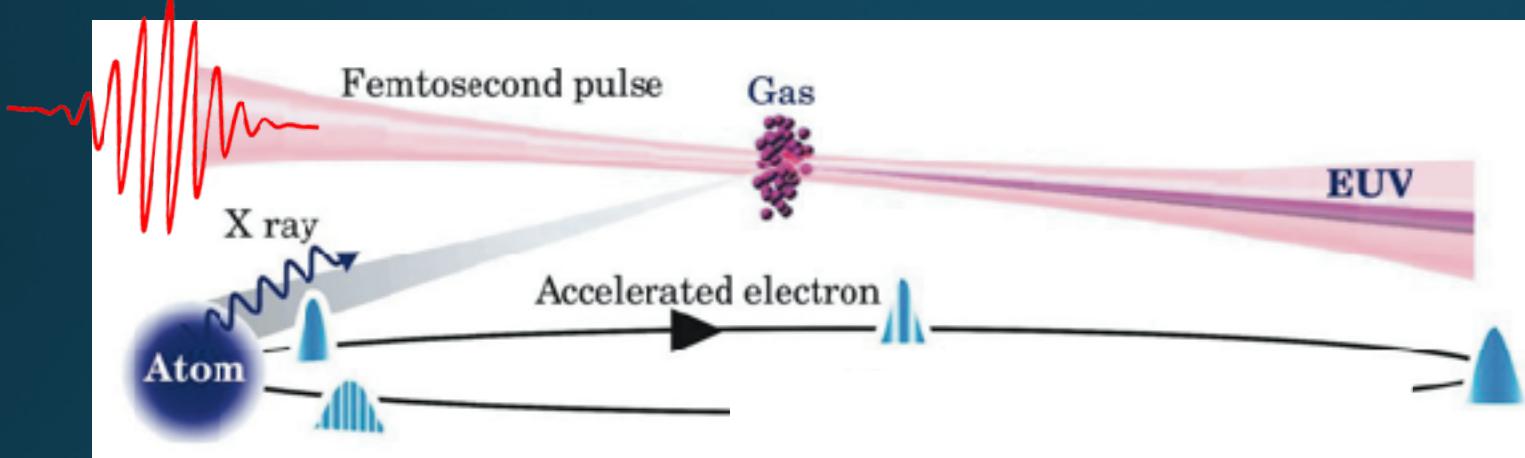
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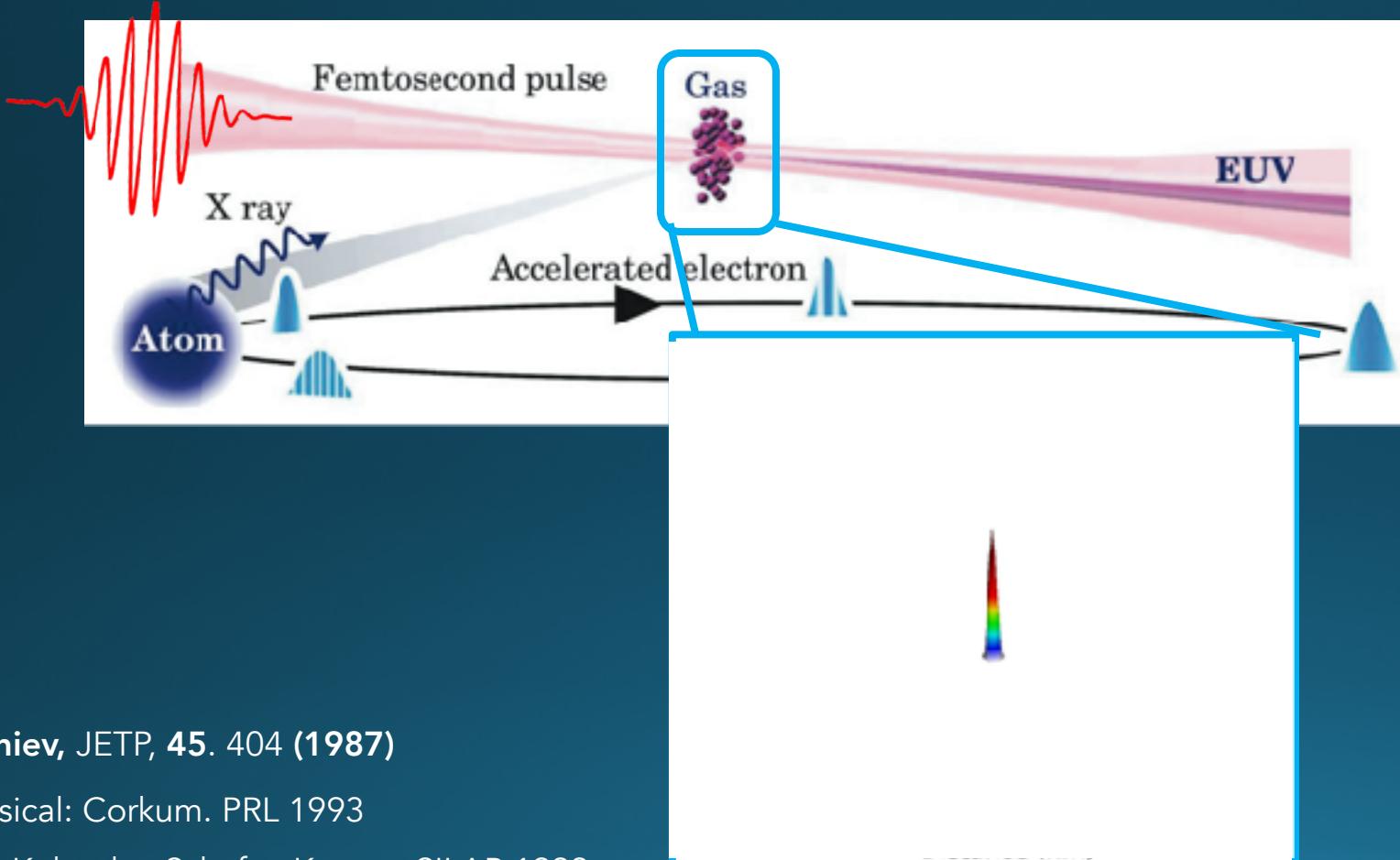
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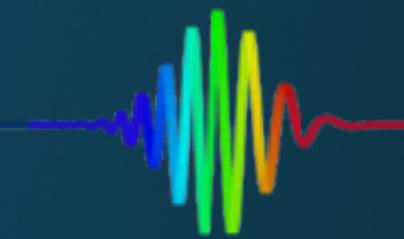


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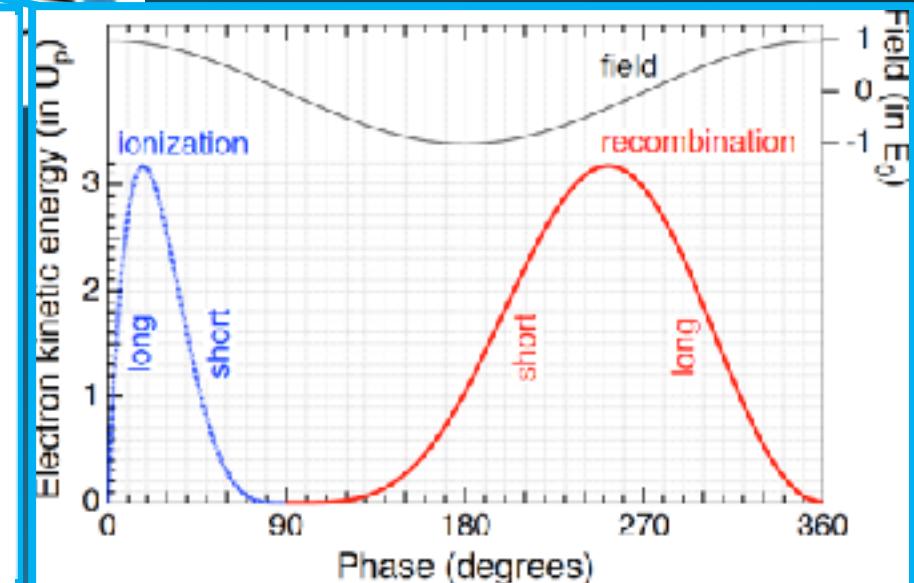
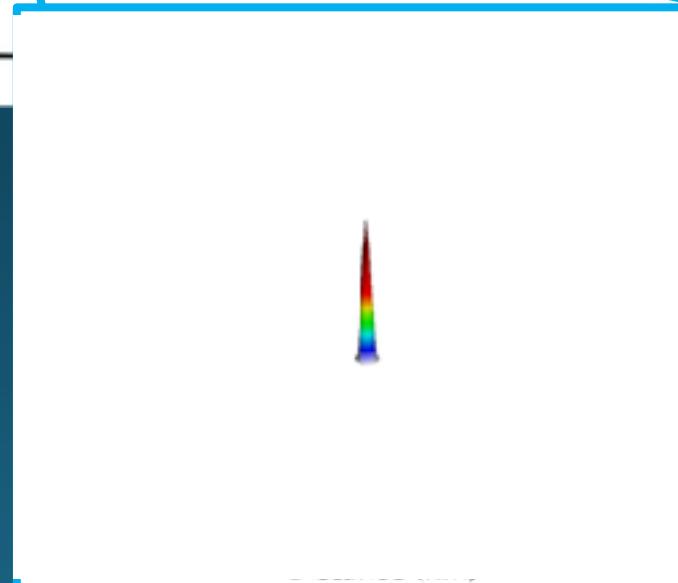
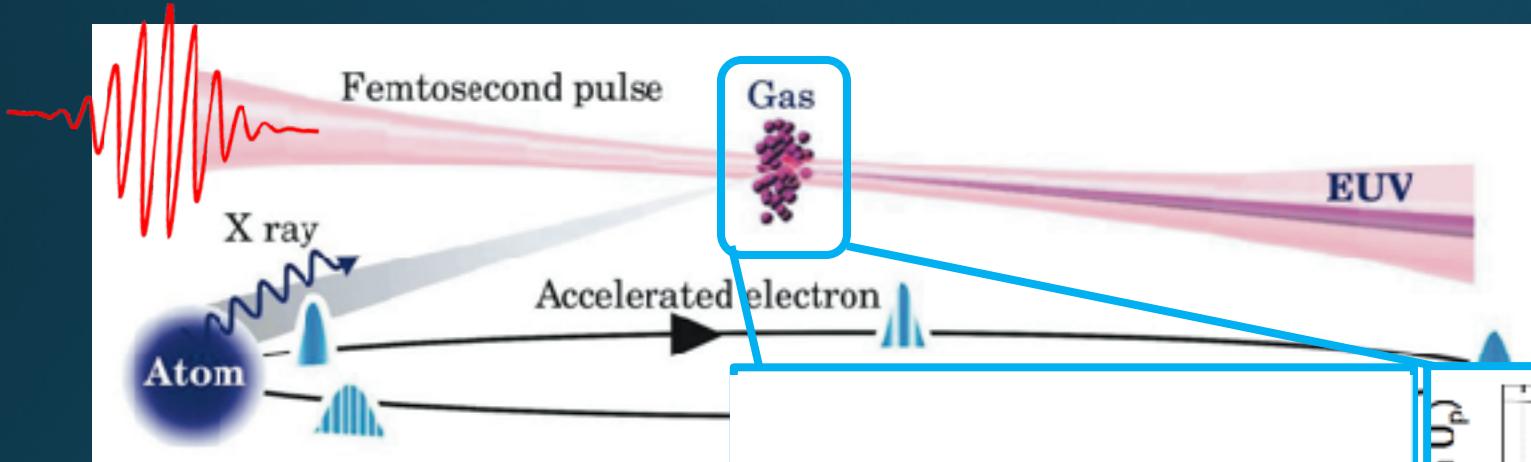
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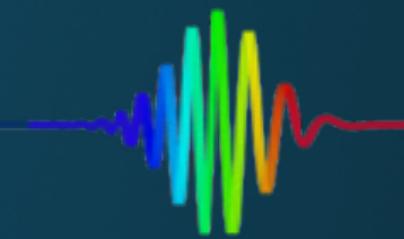


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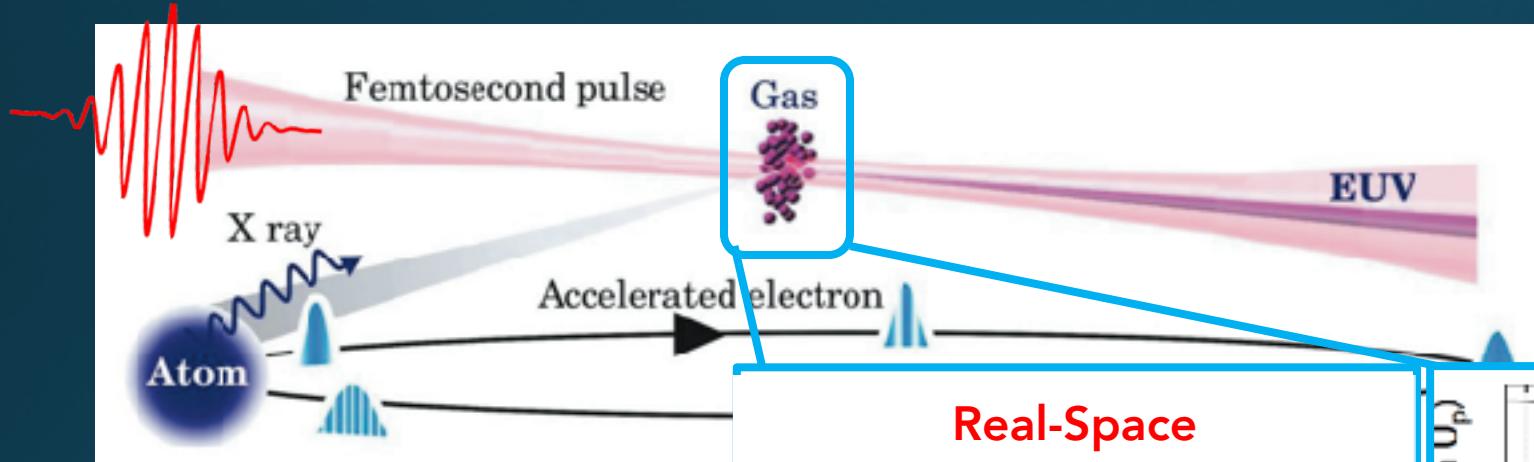
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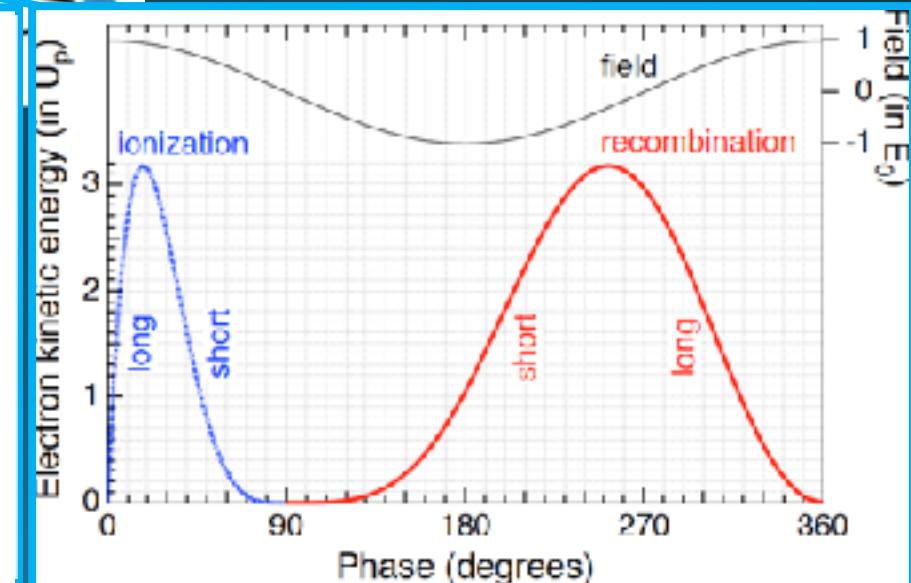
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Real-Space
Wavepacket Motion

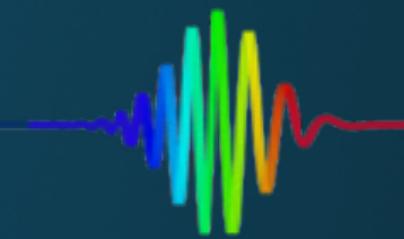


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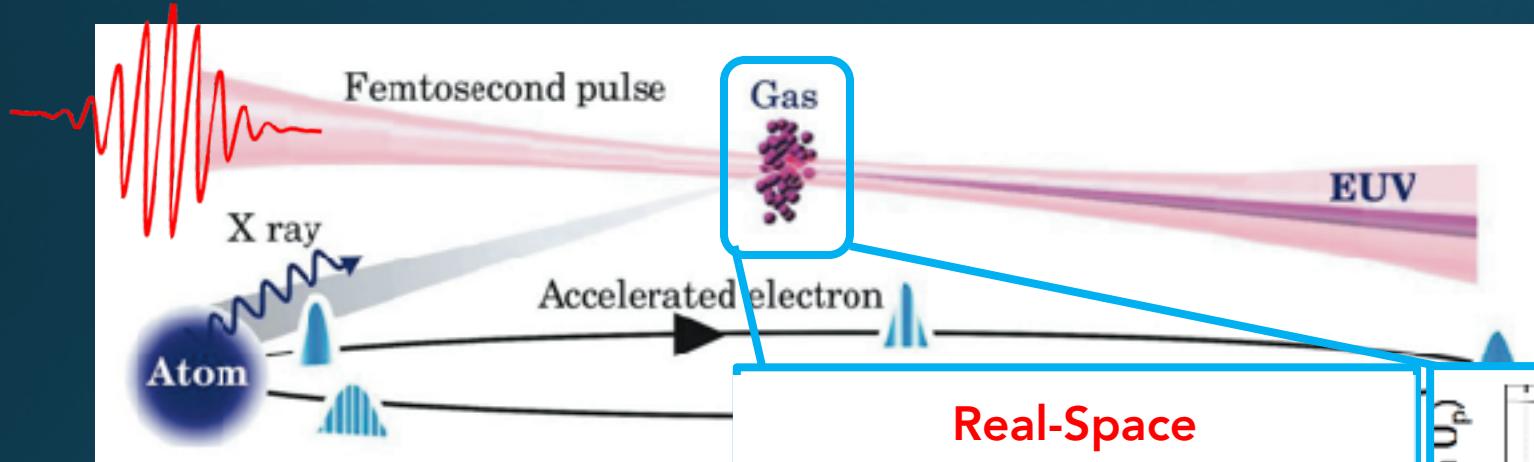
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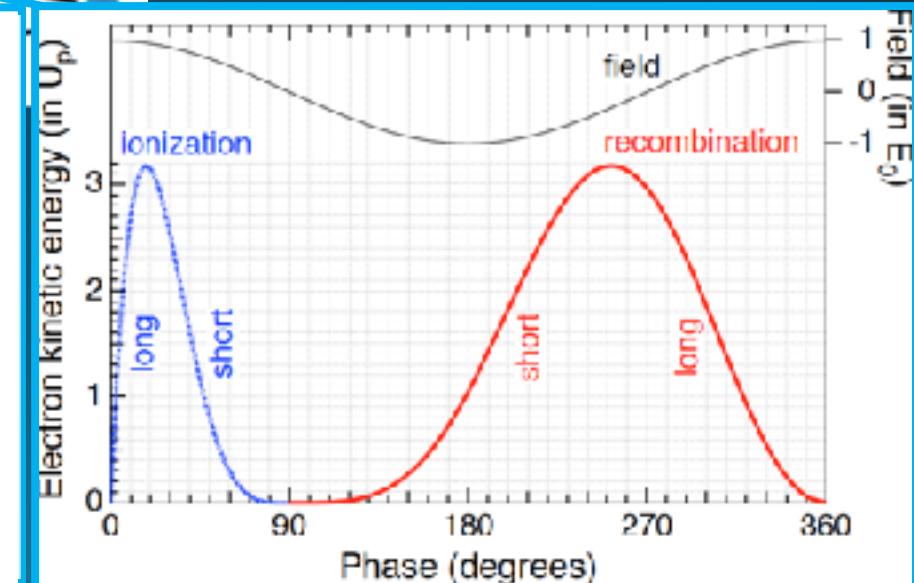
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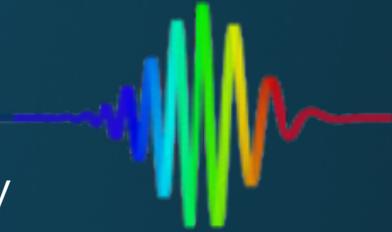


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Macroscopic HHG: Attosecond Nonlinear Optics in a K (Nut) Shell



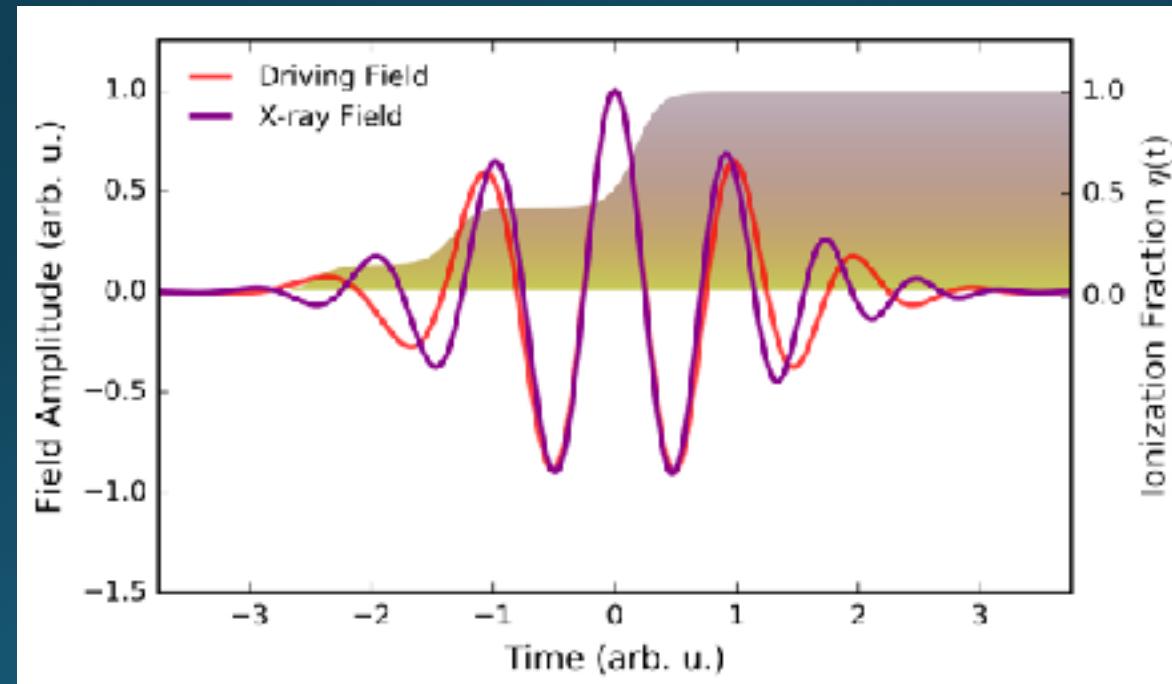
- The generation of bright, coherent beams of X-ray light demands that we solve the currency exchange problem inherent to nonlinear optics.
- Single atom yield $\sim \lambda^{-6.5}$

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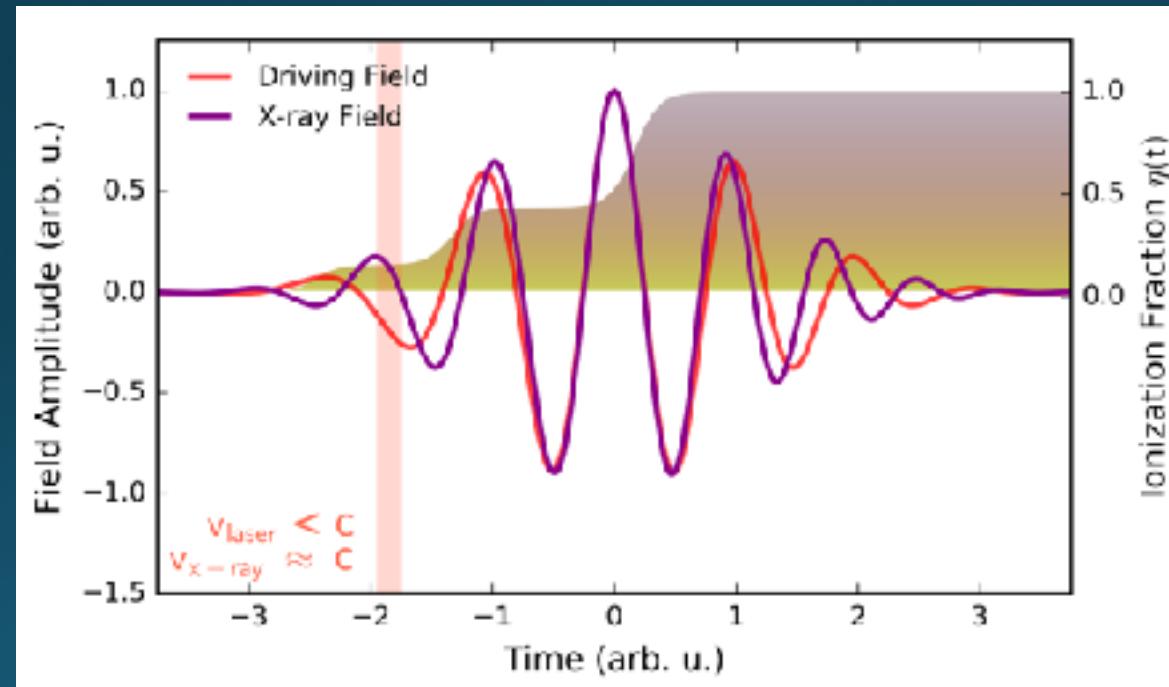


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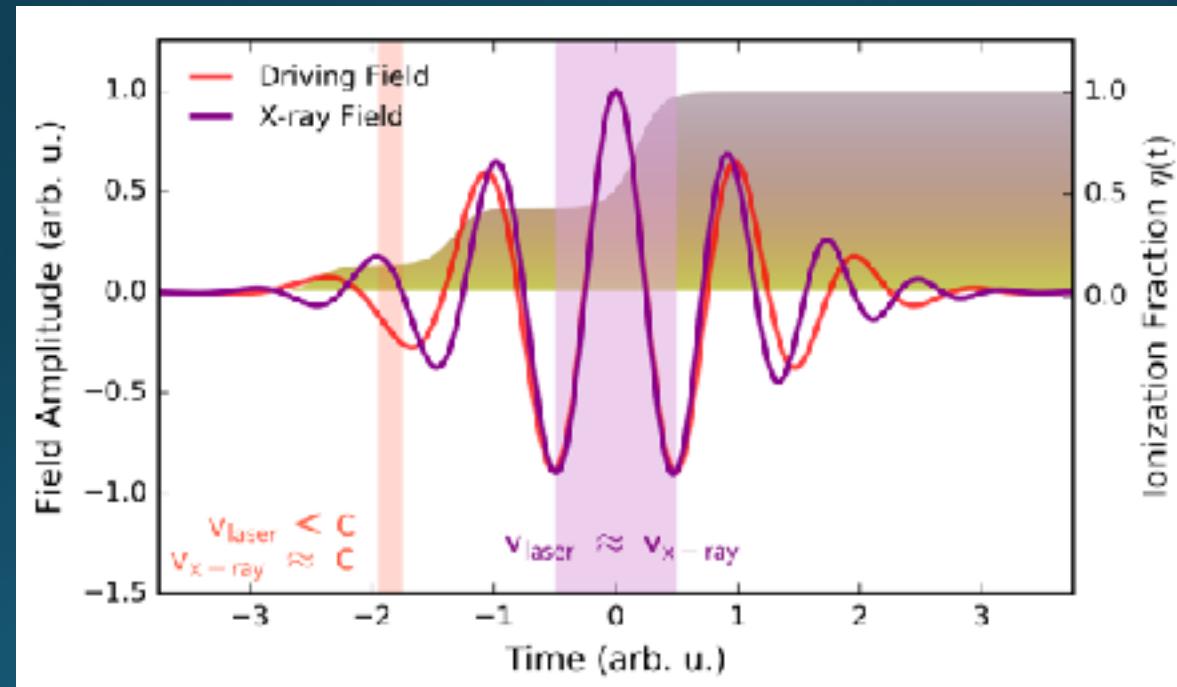


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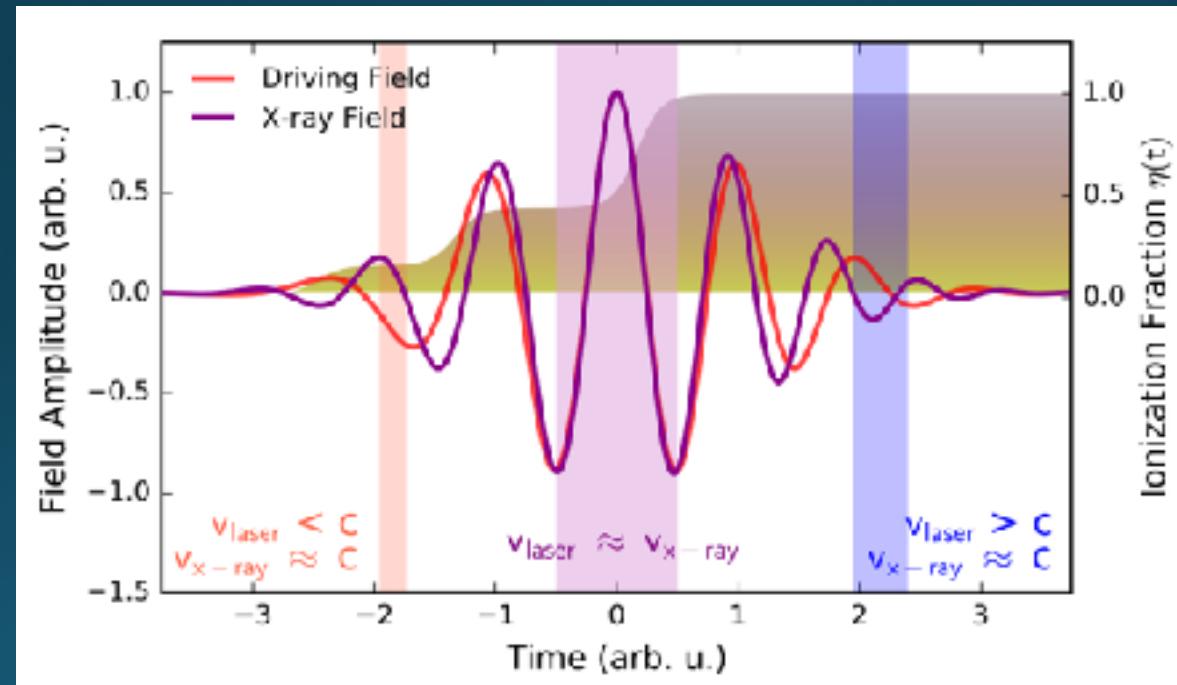


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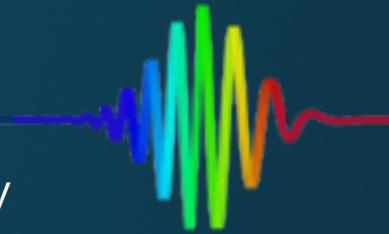


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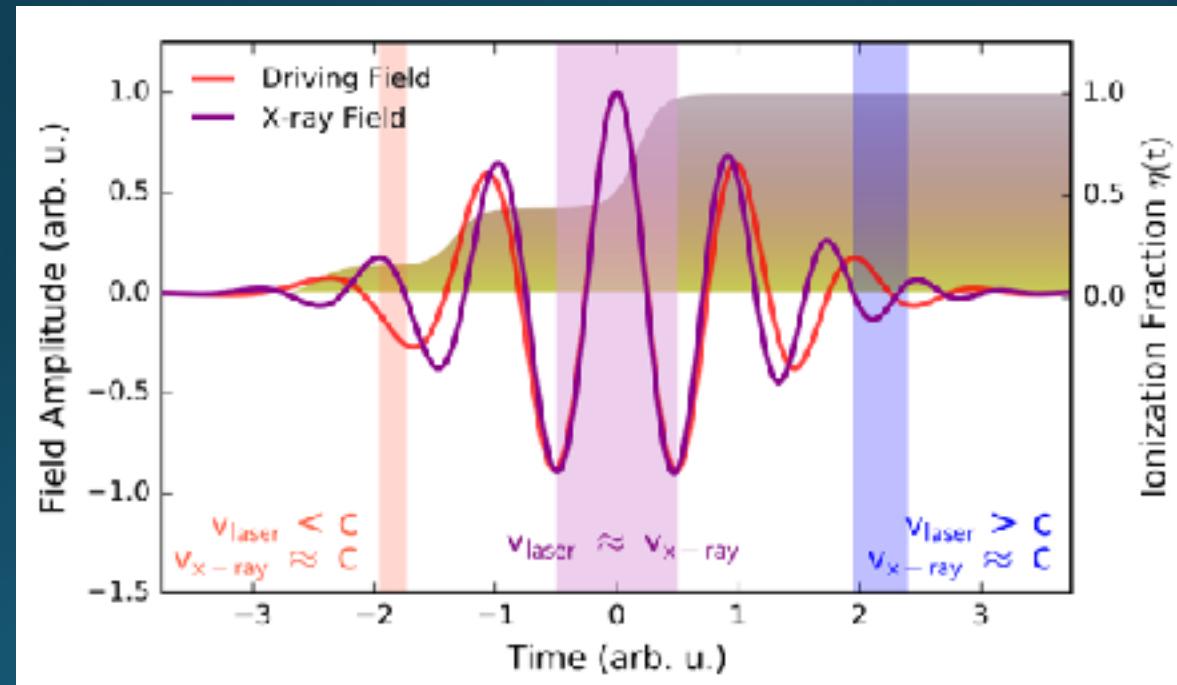
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$$\Delta k(t) = k_{q\omega} - qk_\omega$$

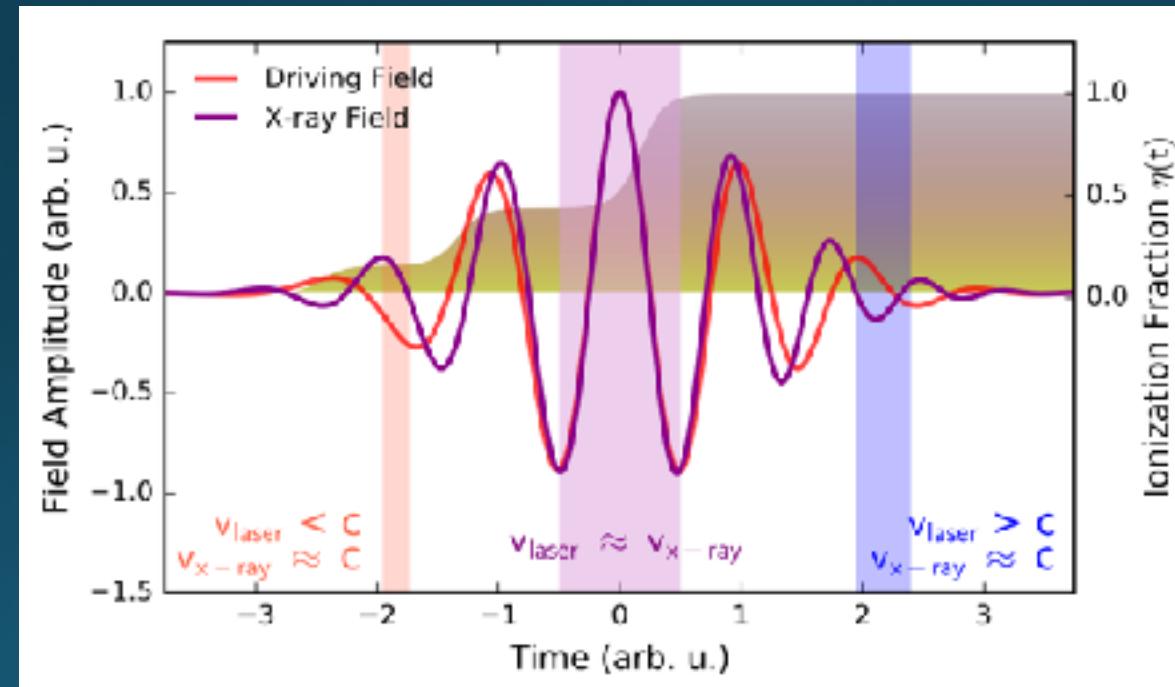


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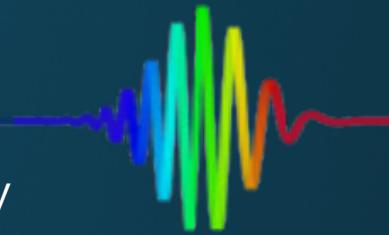


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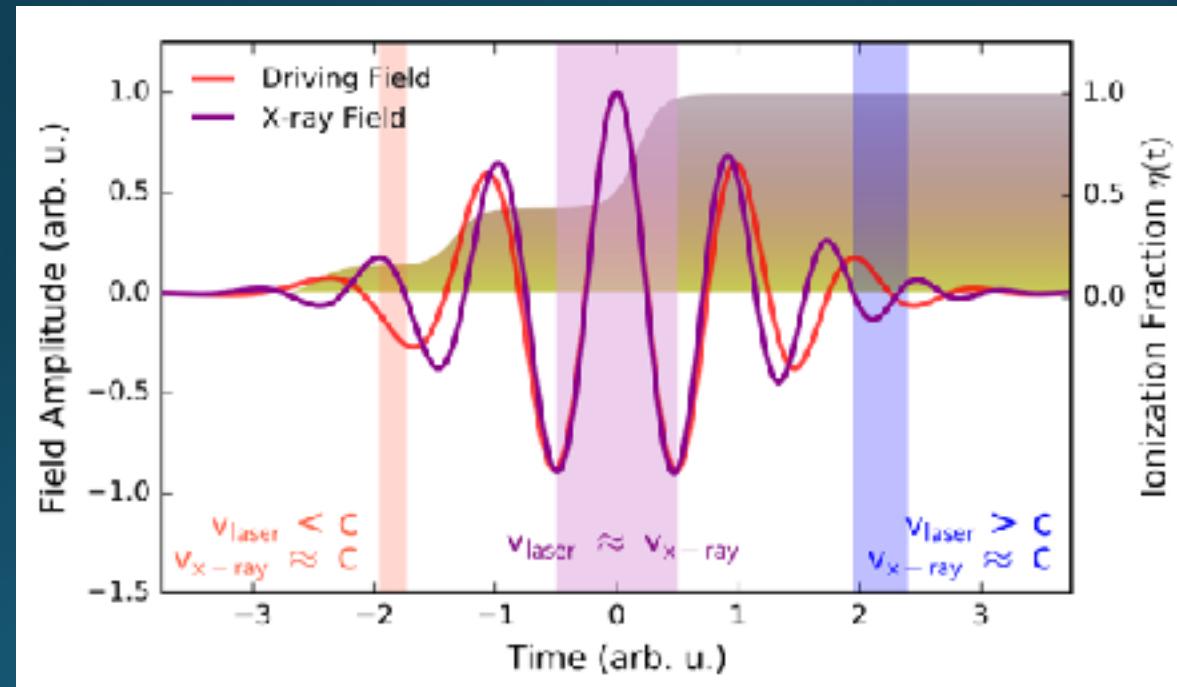
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$$\Delta k(t) = -qP \left\{ [1 - \eta(t)] \frac{2\pi}{\lambda_L} \delta n - \eta(t) N_{atm} r_e \lambda_L \right\} + \Delta k_{geom} + \Delta k_{quantum}(t)$$



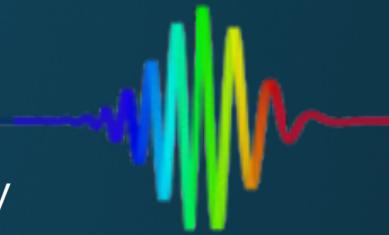
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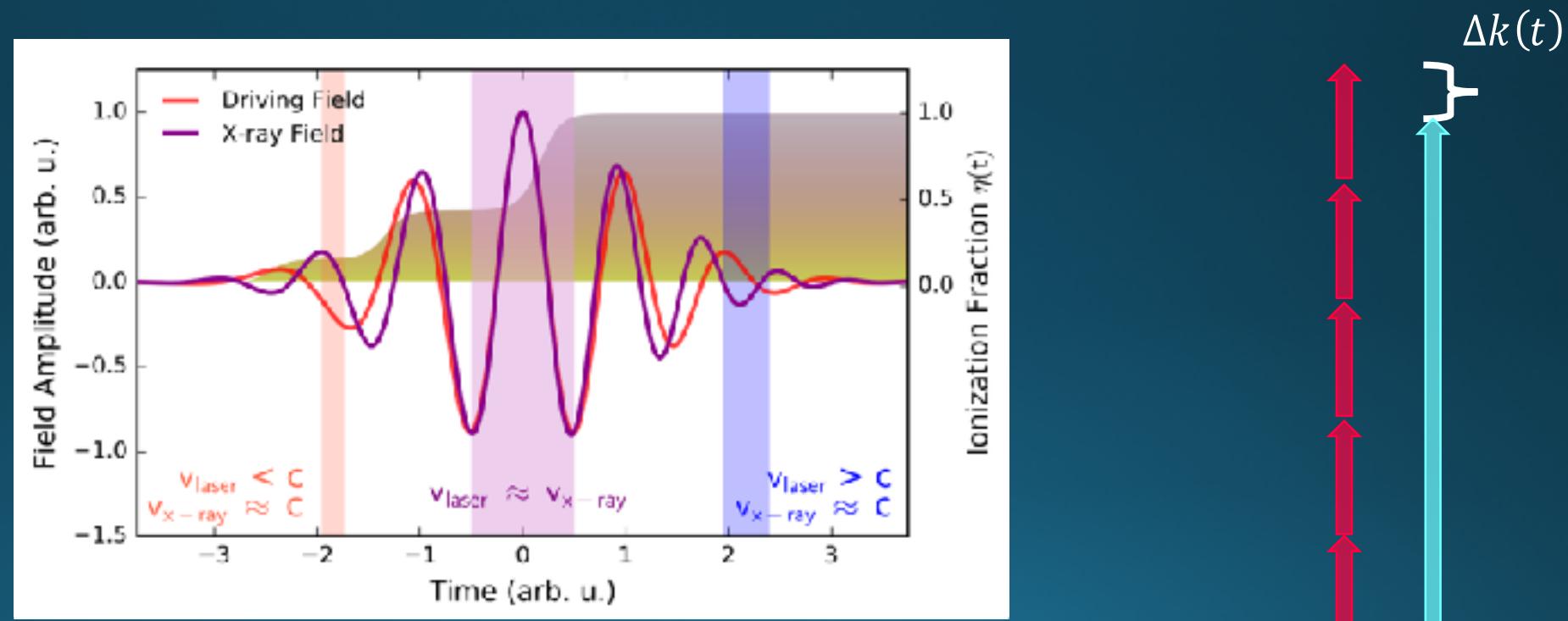
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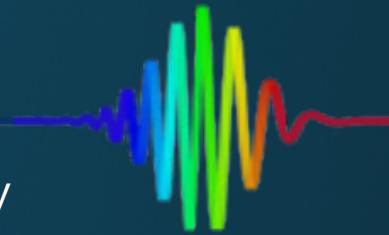
- Single atom yield $\sim \lambda^{-6.5}$

$$\Delta k(t) = -qP \left\{ [1 - \eta(t)] \frac{2\pi}{\lambda_L} \delta n - \eta(t) N_{atm} r_e \lambda_L \right\} + \Delta k_{geom} + \Delta k_{quantum}(t)$$



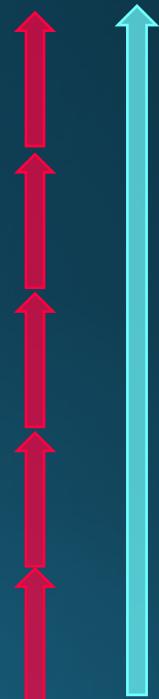
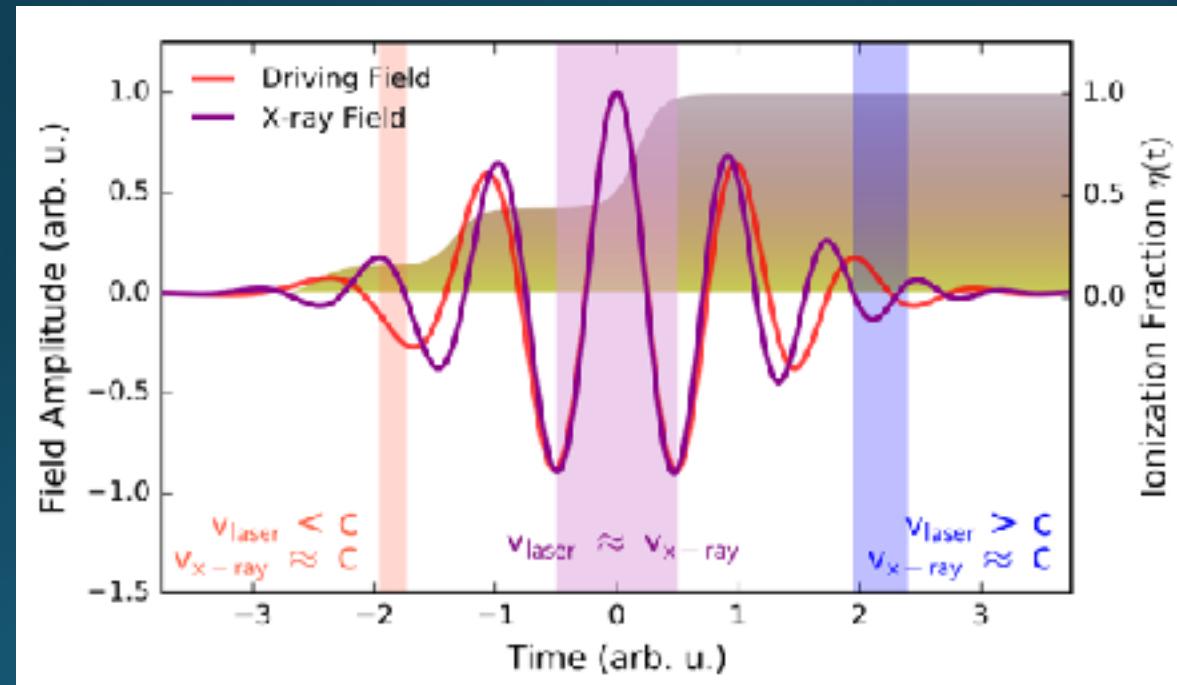
Rundquist, Science, 5368, 1998
 Popmintchev, PNAS, 106, 2009
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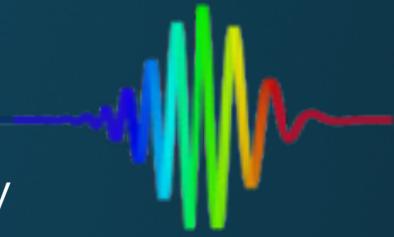
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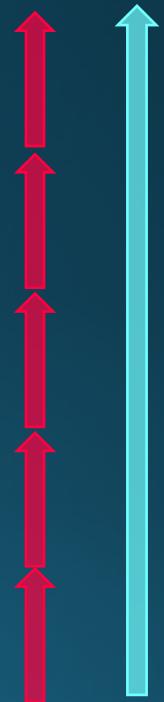
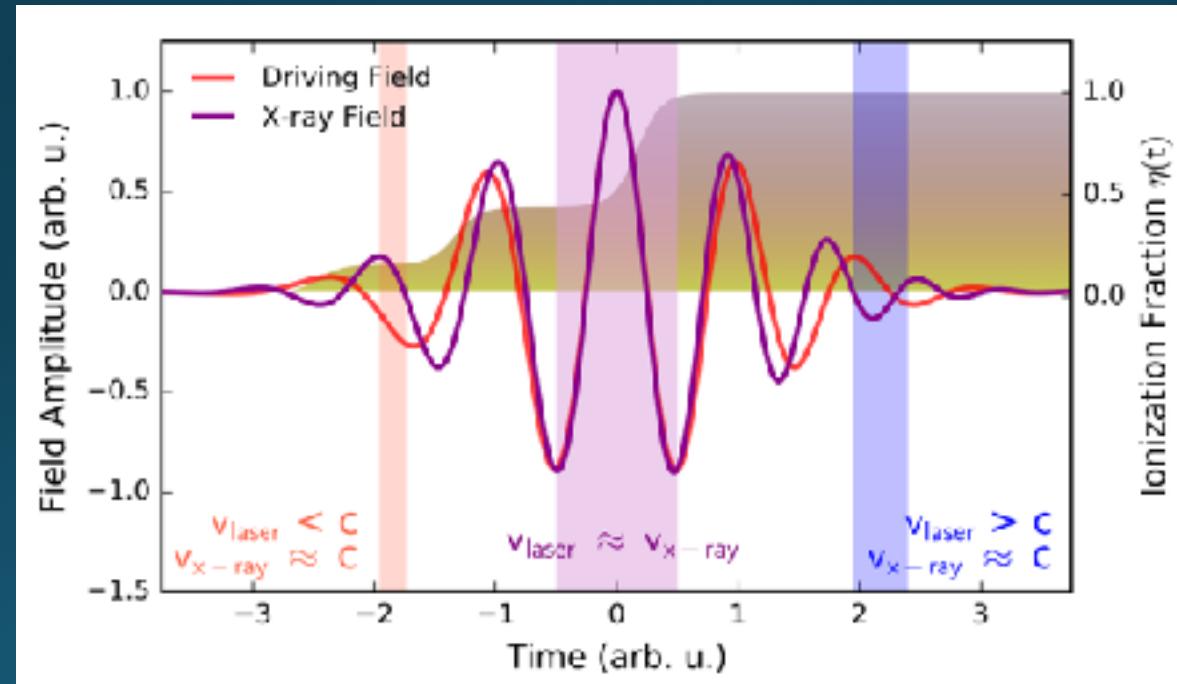


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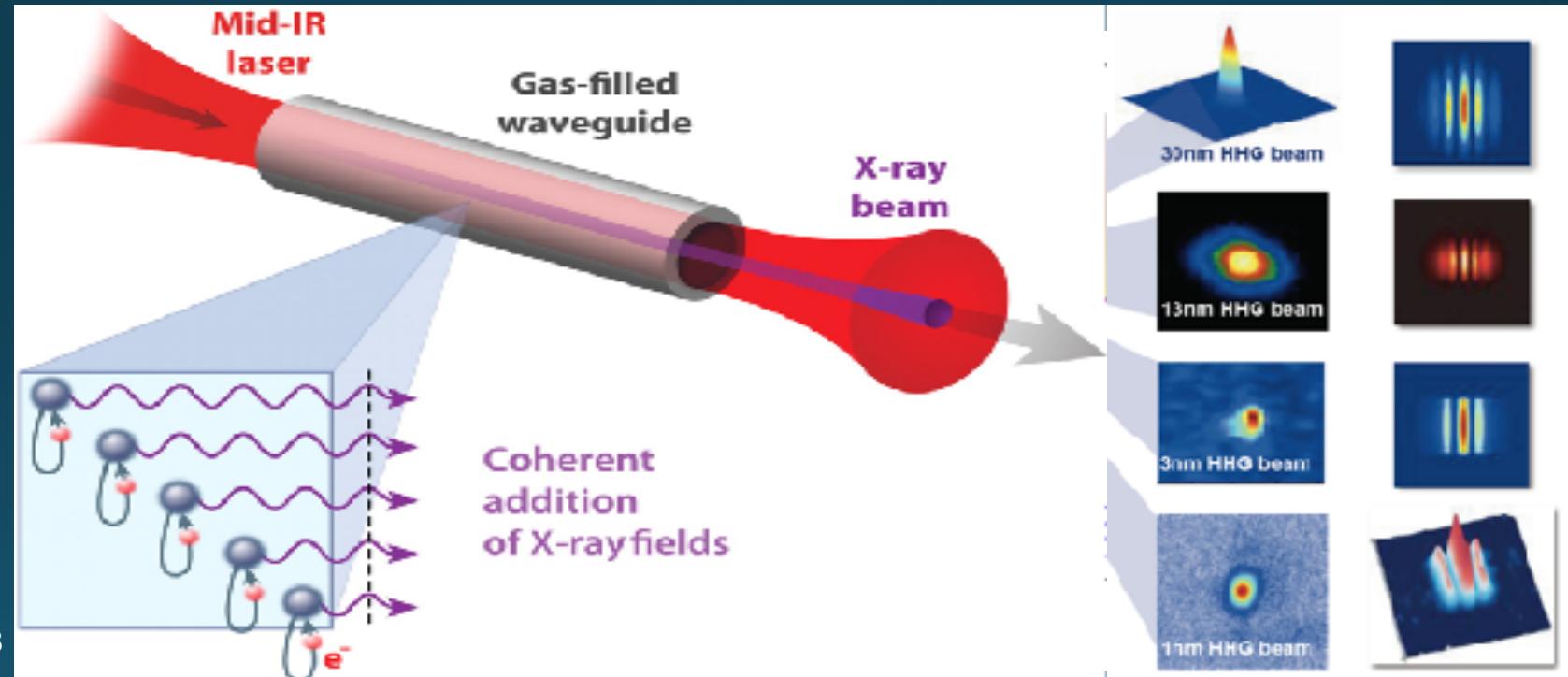


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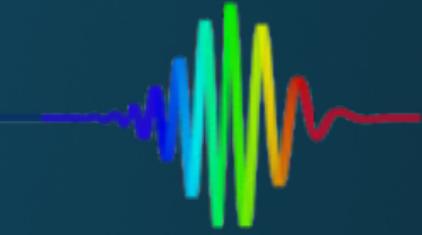
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Custom Tailored EUV and X-ray Light From a Table-Top: Exquisite Control Over the Entire Up-Conversion Process



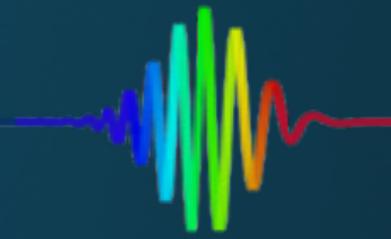
Chen, PNAS, 111, 2014

Hernandez-Garcia, Opt Exp, 106, 2017

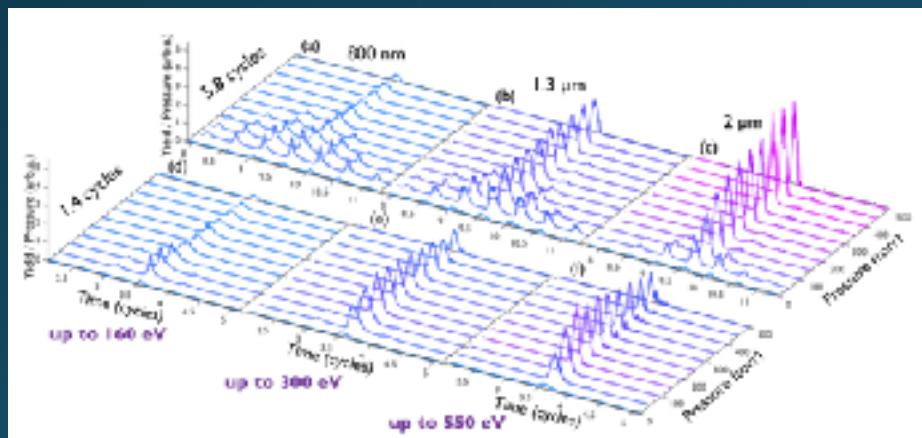
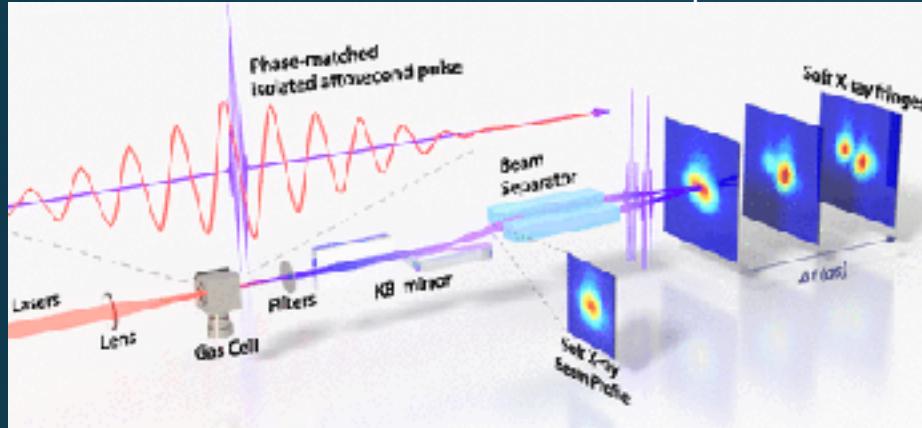
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Phase-matched isolated as pulses



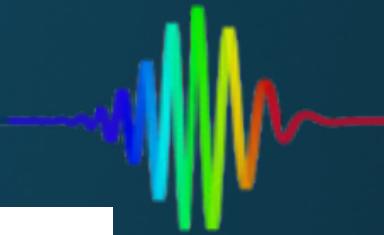
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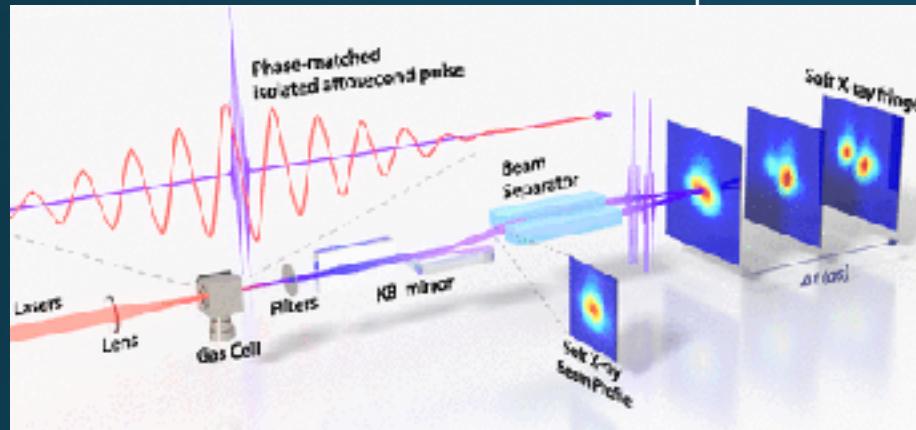
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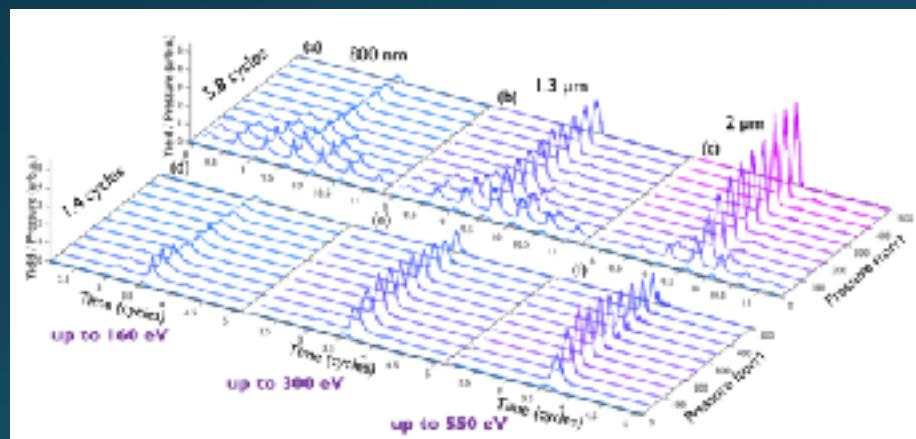
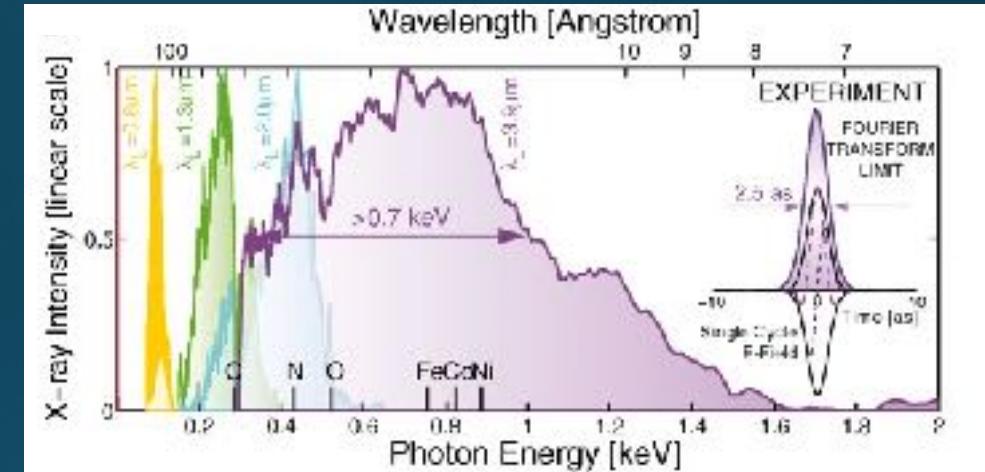
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Coherent, zeptosecond x-rays



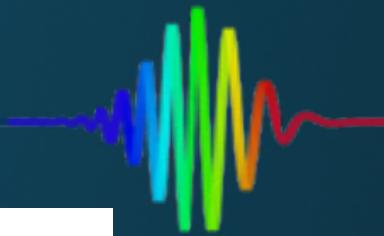
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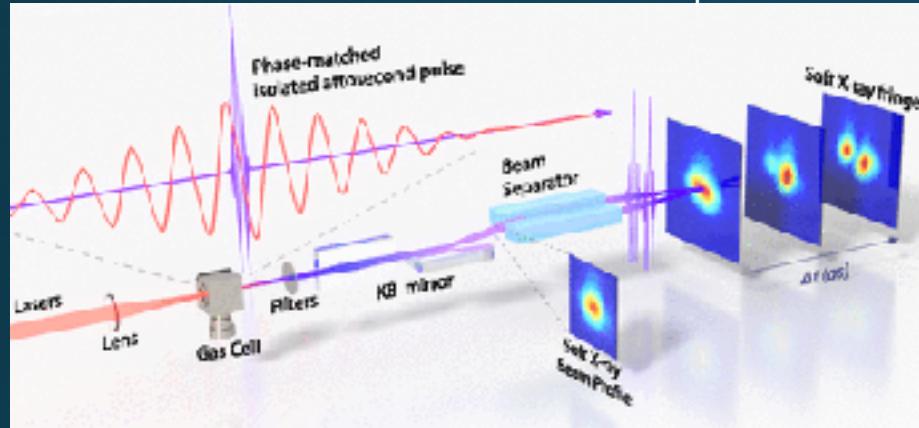
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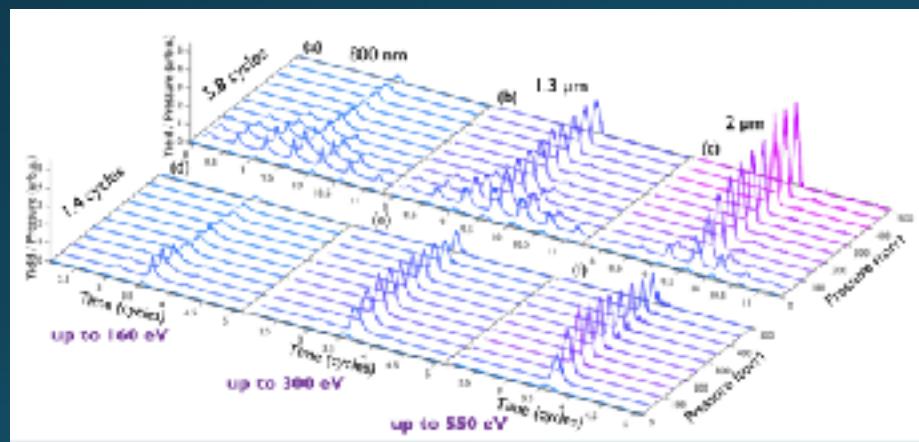
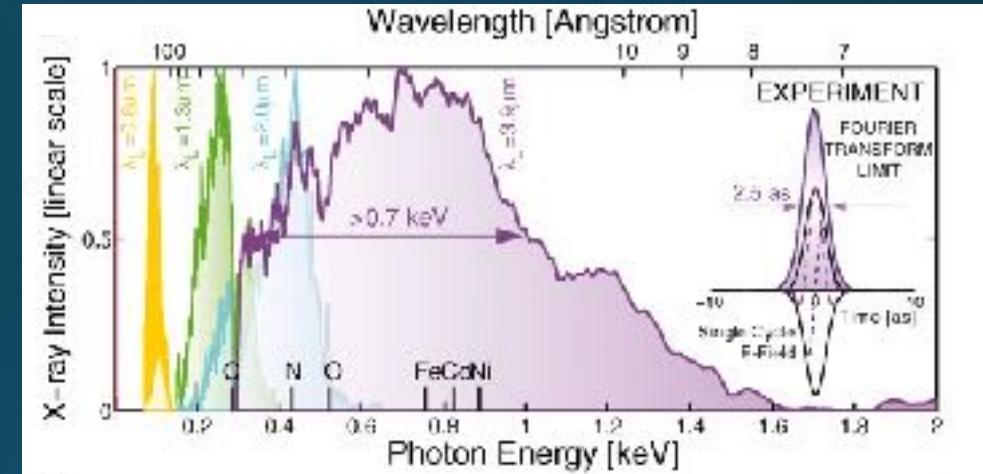
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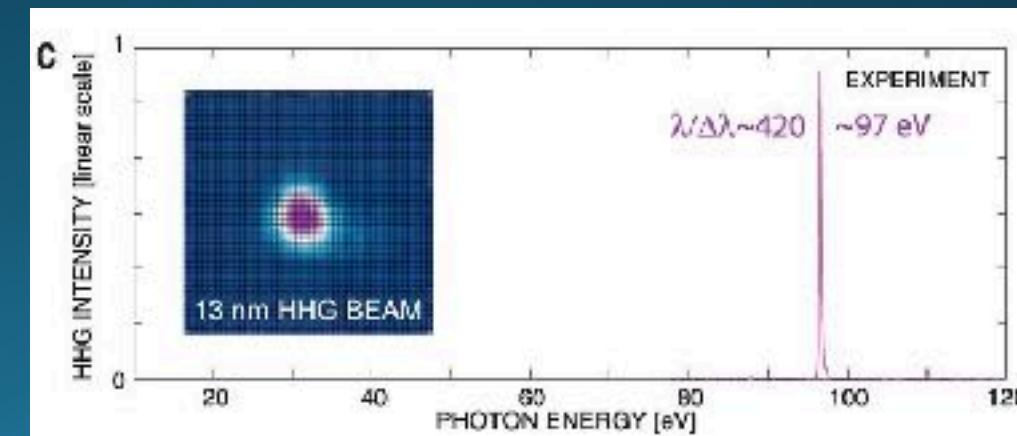
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Bright, isolated harmonics



Chen, PNAS, 111, 2014

Hernandez-Garcia, Opt Exp, 106, 2017

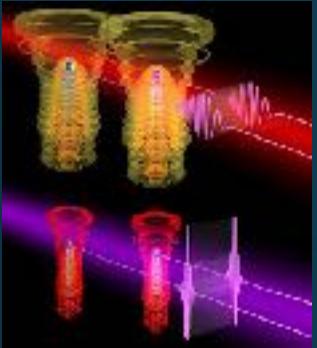
Popmintchev, Science, 6086, 2012

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

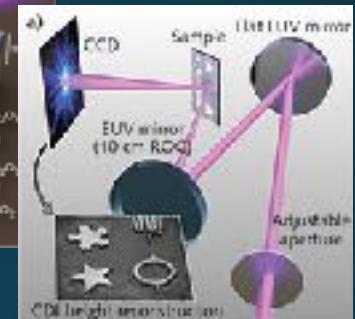
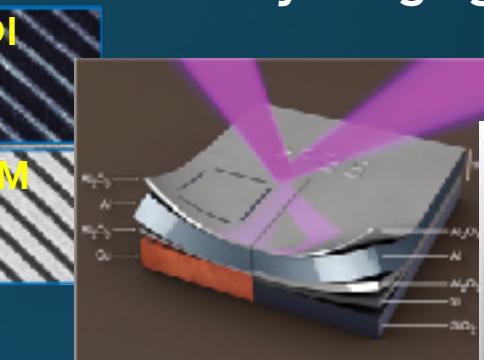


Attosecond Extreme Nonlinear Optics



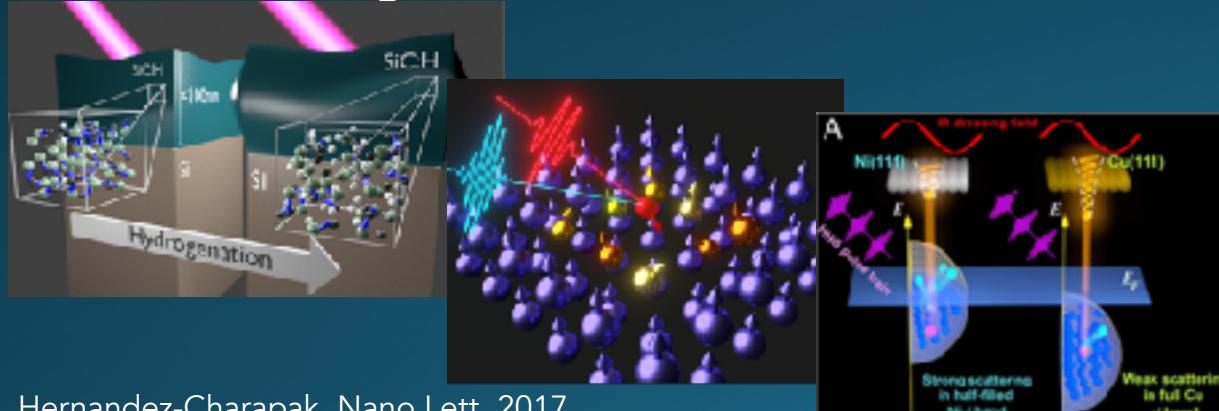
Popmintchev, Science 6265, 2015
Dorney, PRL 118, 2017

Coherent X-ray Imaging



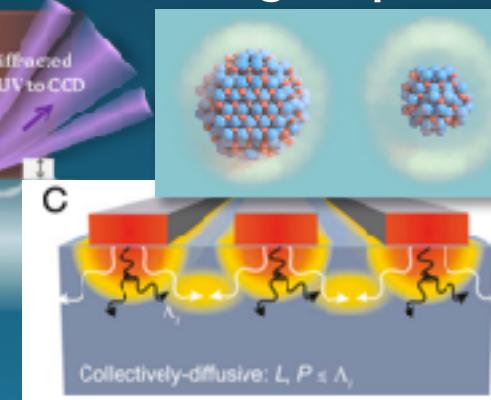
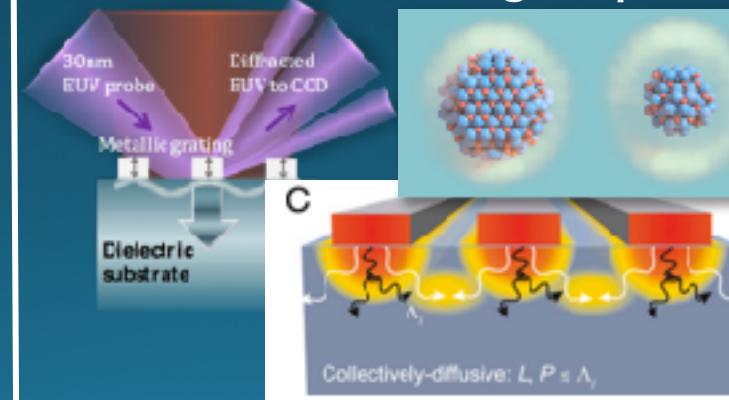
Gardener, Nat. Photon. 11, 2017
Shanblatt, Nano Lett. 16, 2016

Uncovering New Ultrafast Materials Science



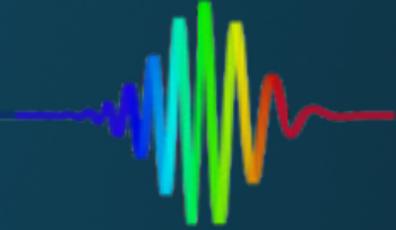
Hernandez-Charapak, Nano Lett. 2017
Tao, Science 353, 2016
Chen, PNAS 114, 2017

Nanoscale Charge, Spin, Energy Transport



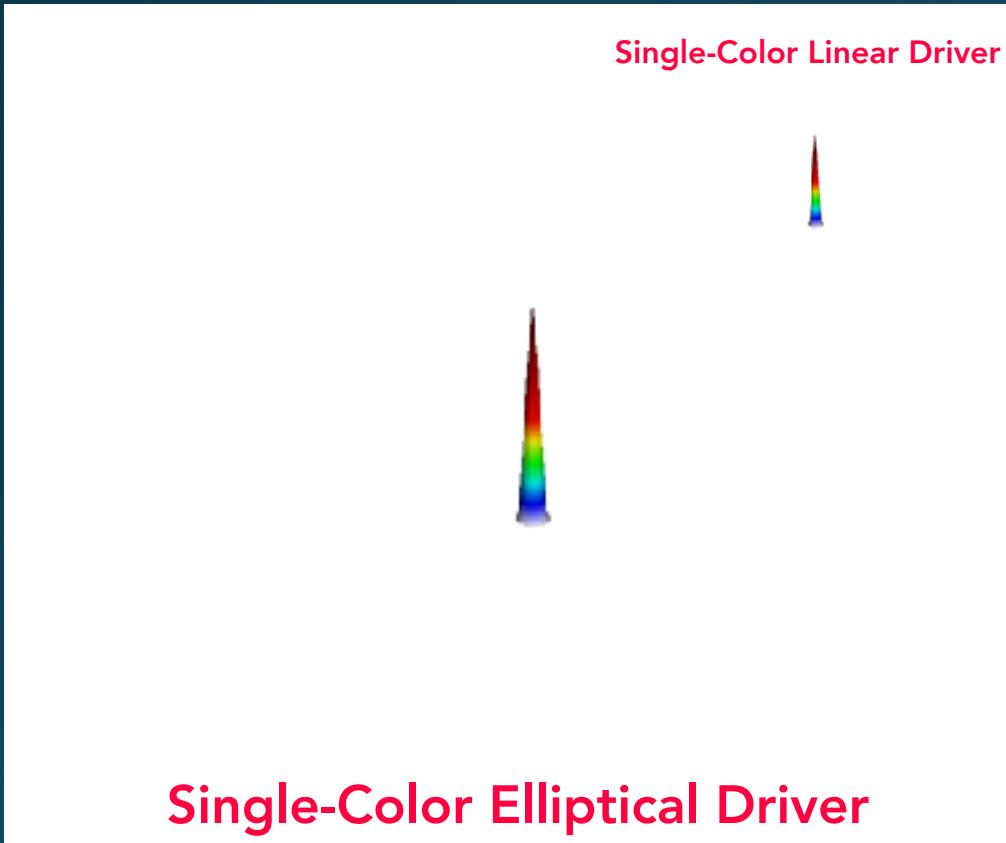
Hoogeboom-Pot, PNAS 112, 2015
Ellis, JACS, 11 2015

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



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

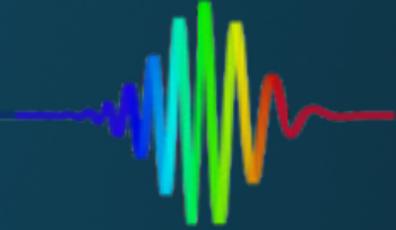
Video: Courtesy of Carlos Hernandez-Garcia



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

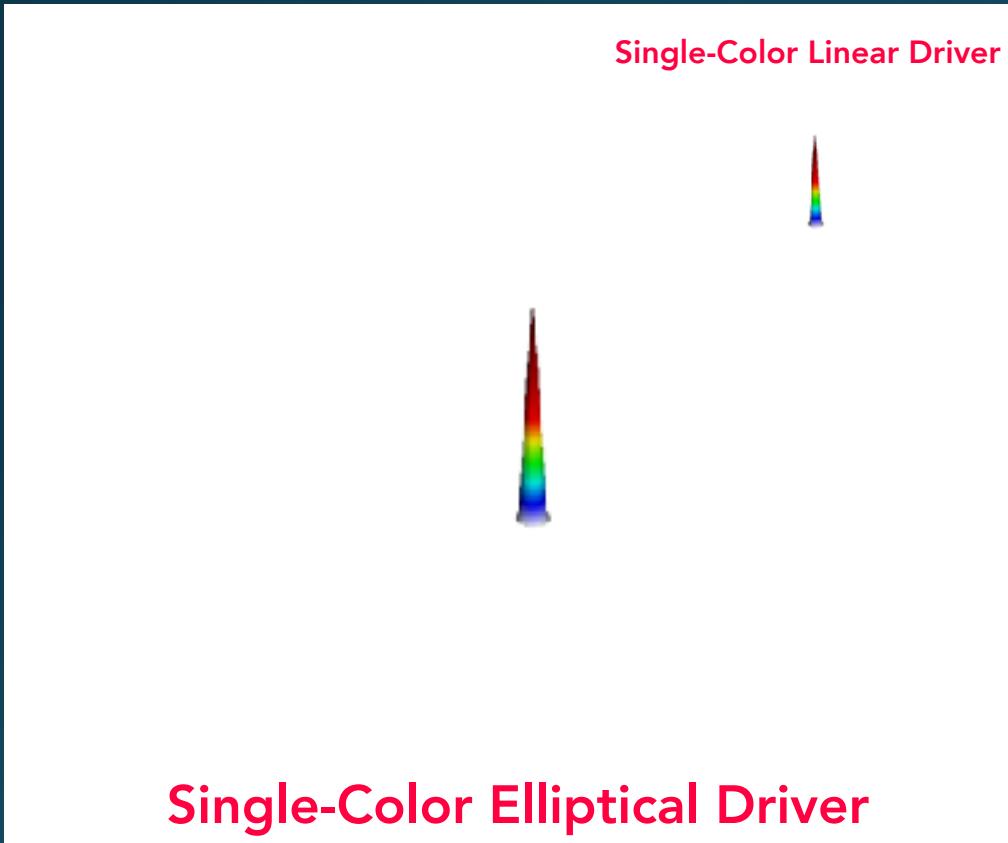
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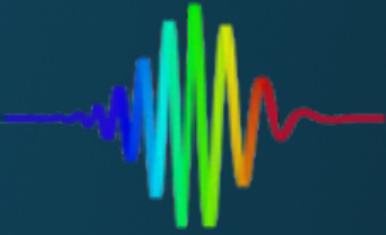
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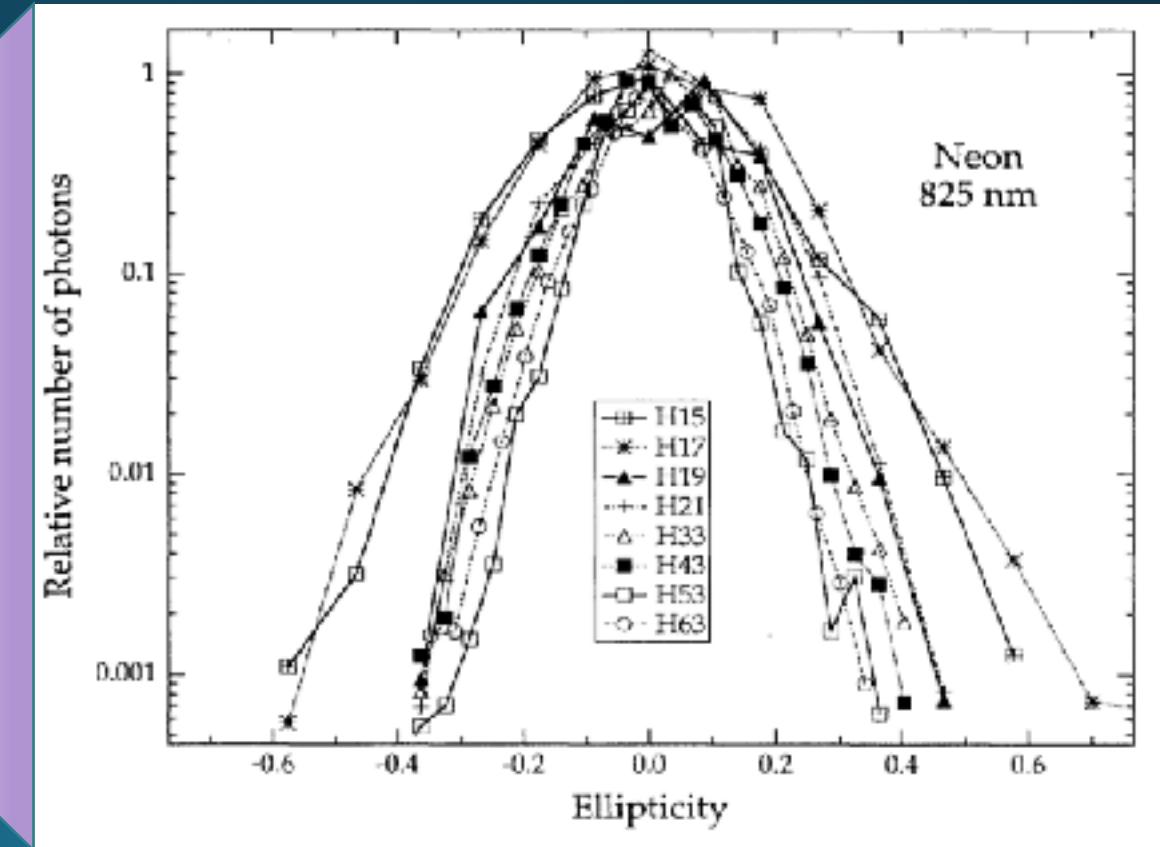
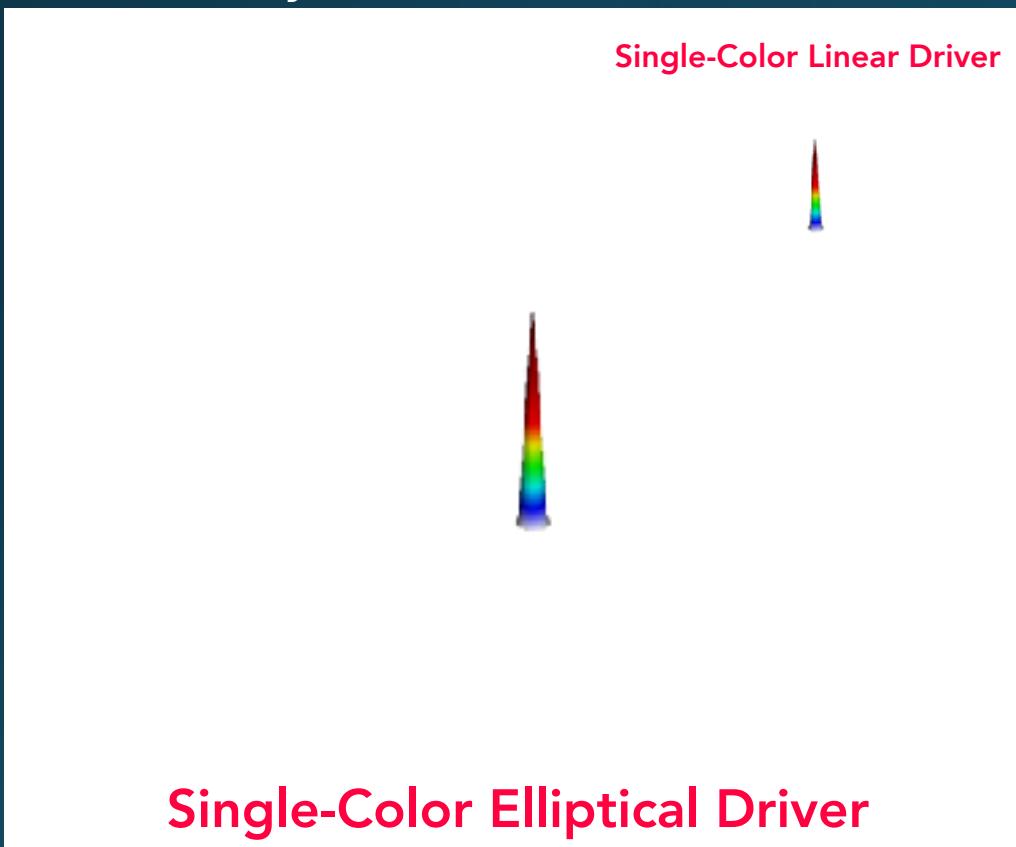
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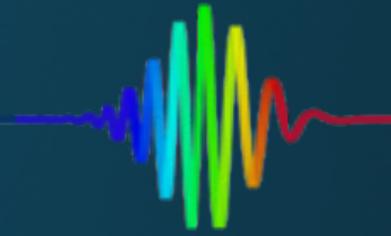
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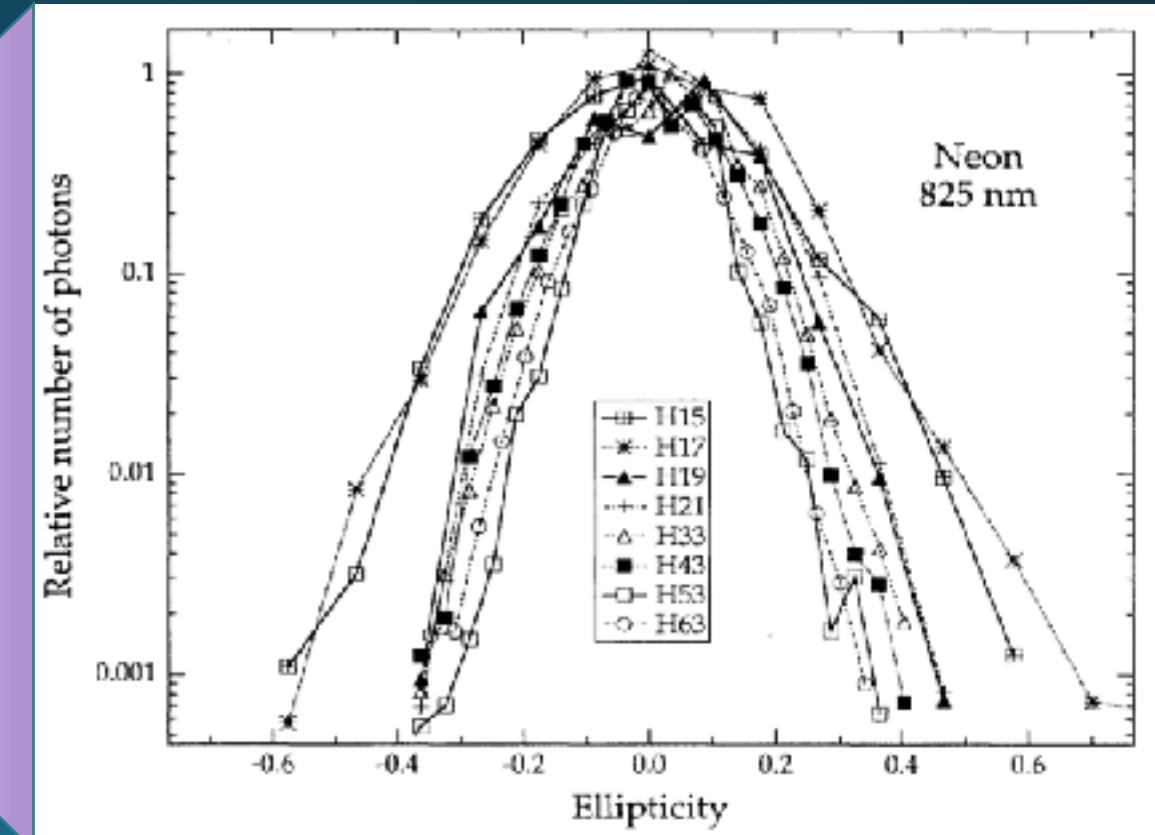
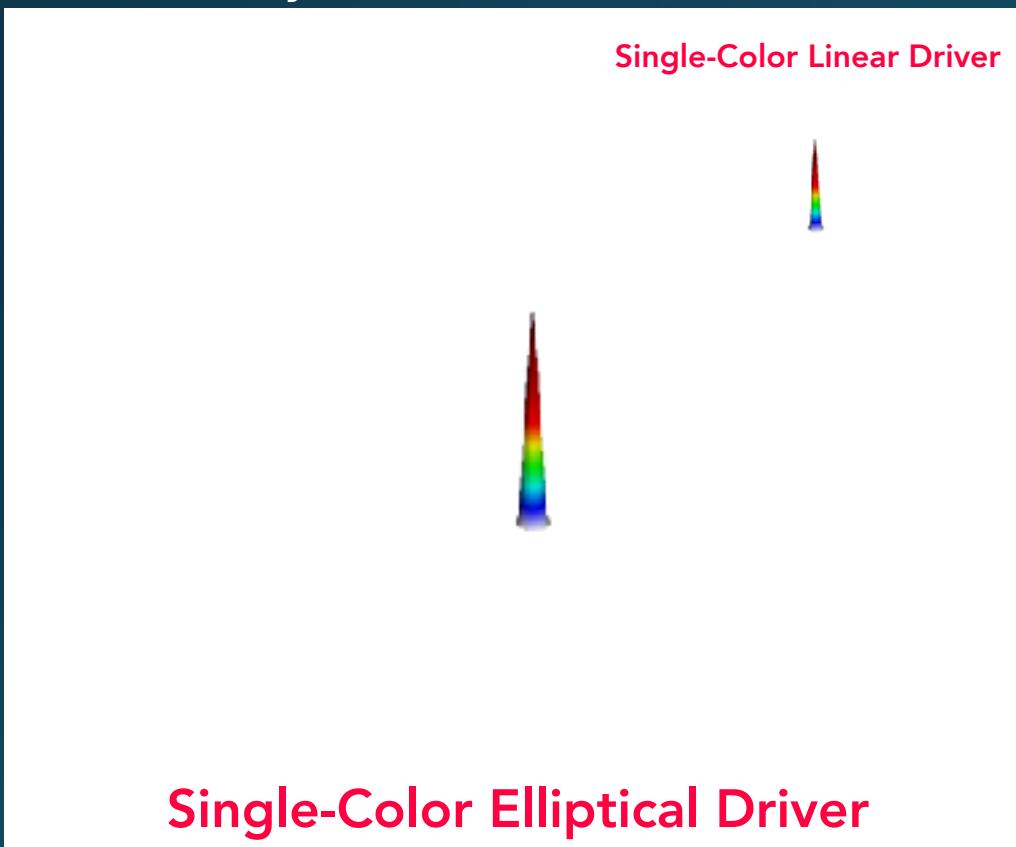
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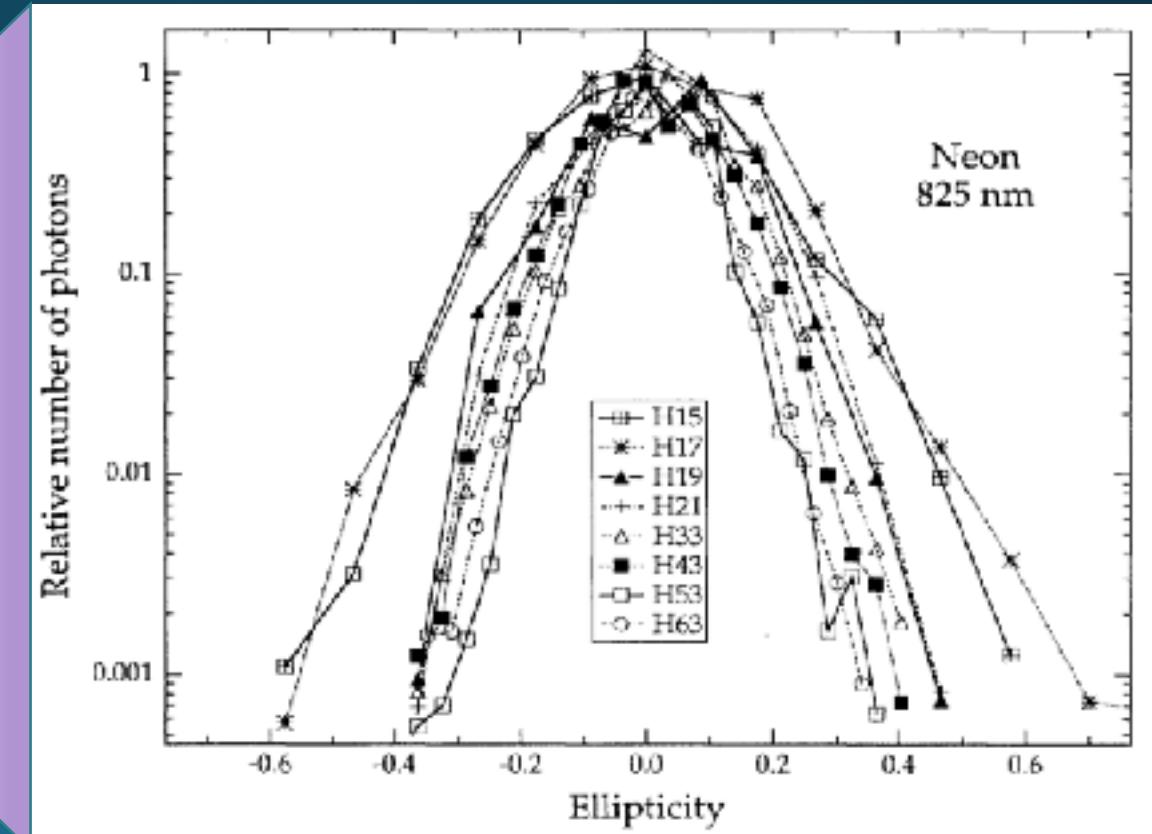
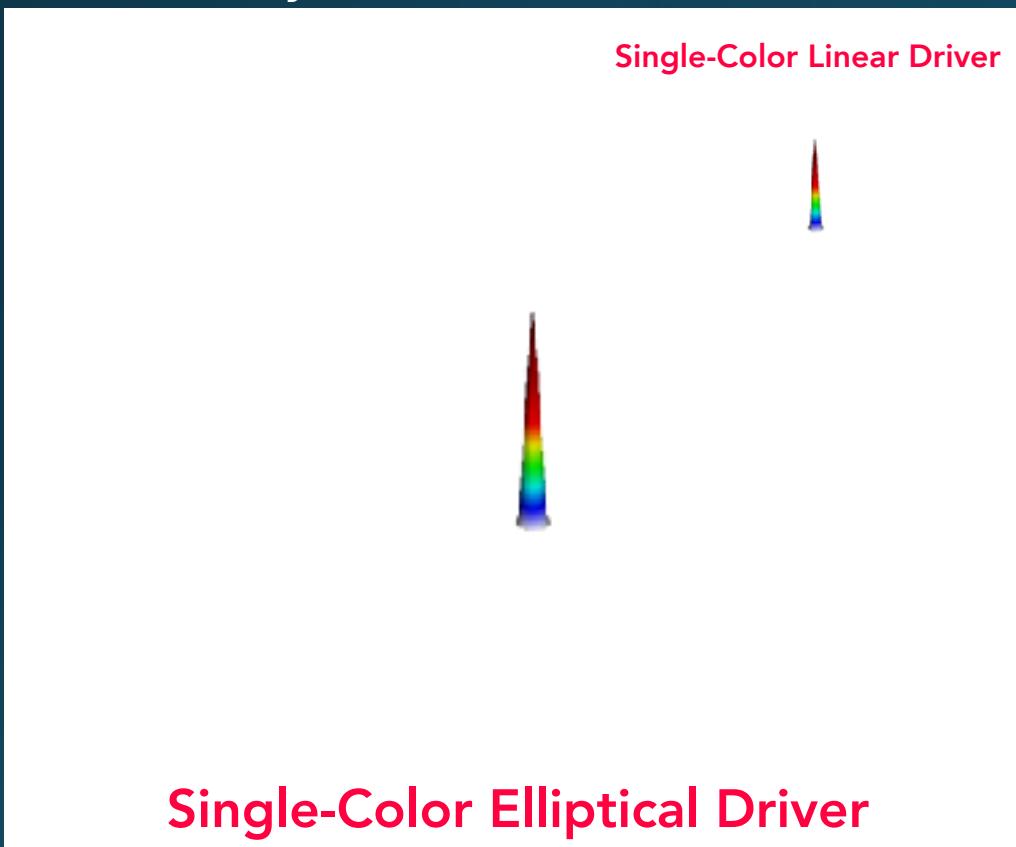
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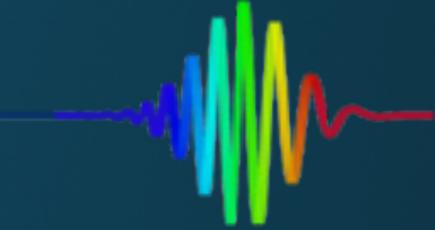
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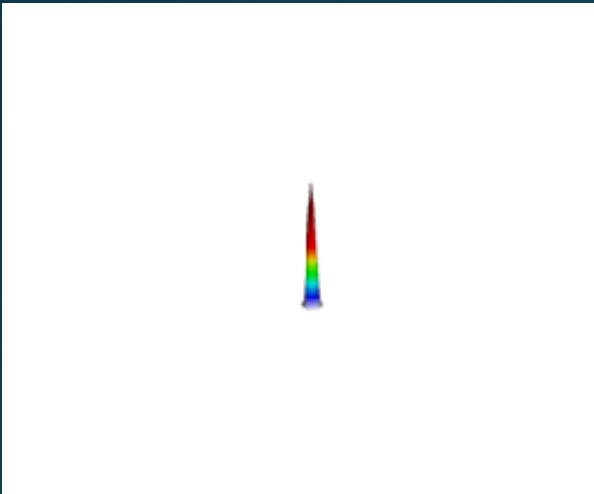


HHG in a Whole New Dimension: Reshaping the Frontier of Attosecond Science

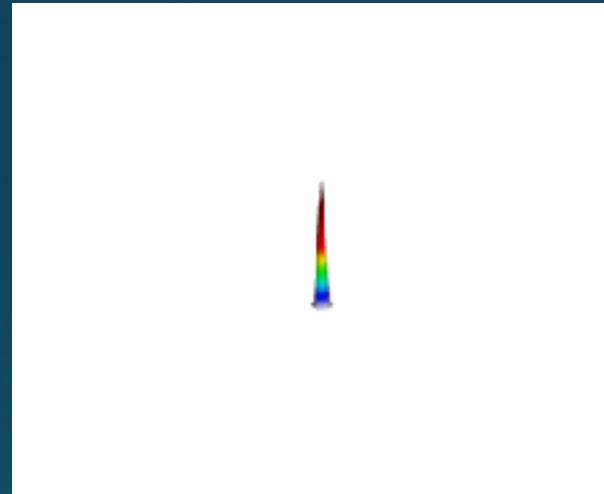
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One Color Linear Driver



One-Color Circular Driver



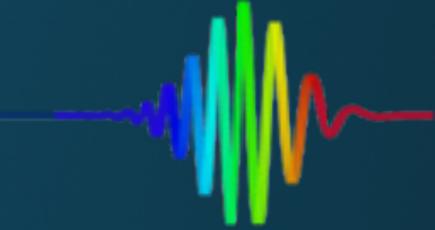
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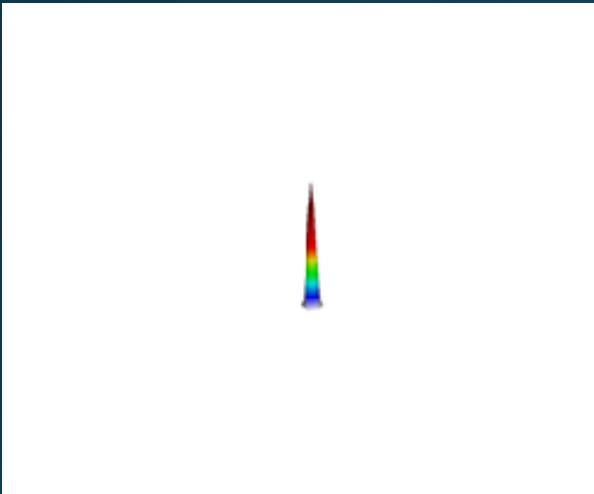


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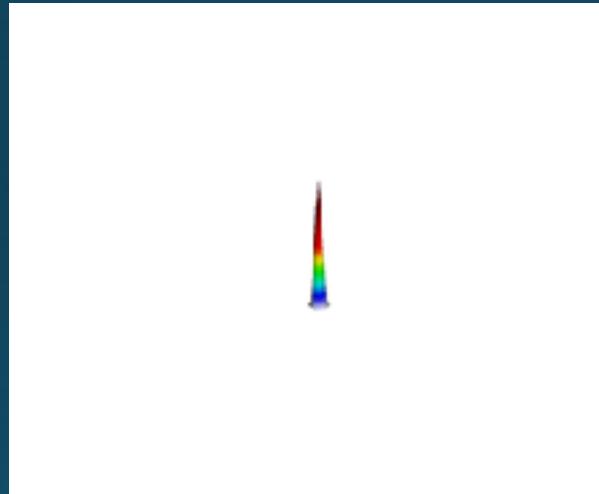
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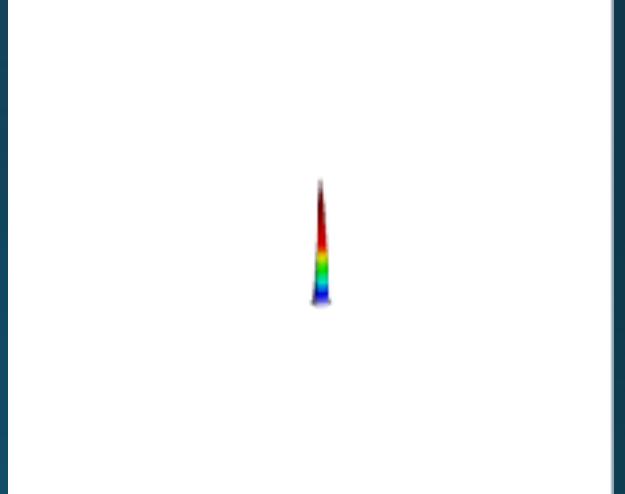
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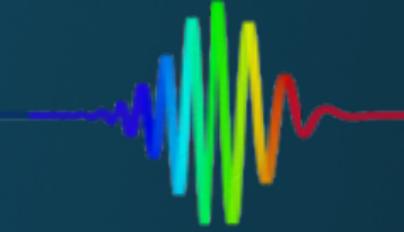
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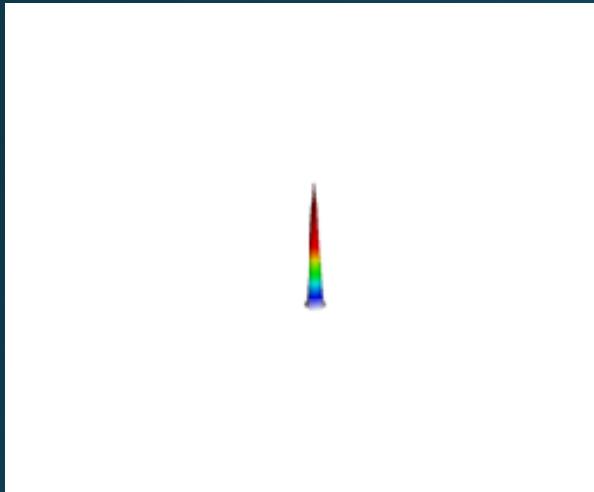


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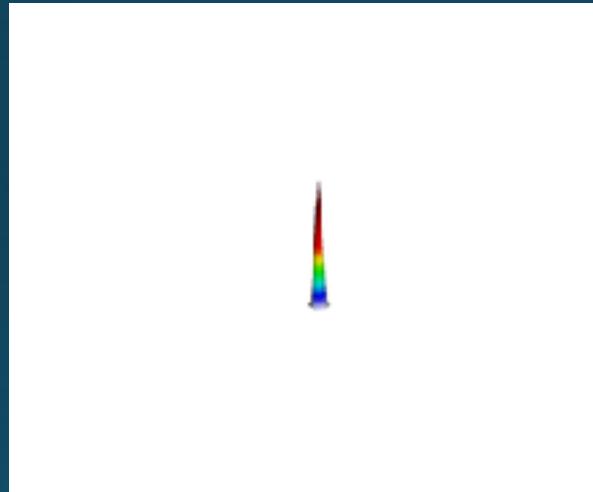
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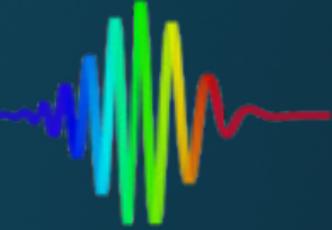


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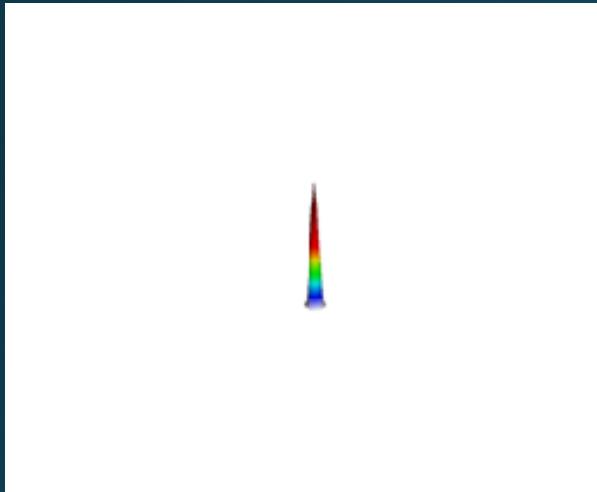


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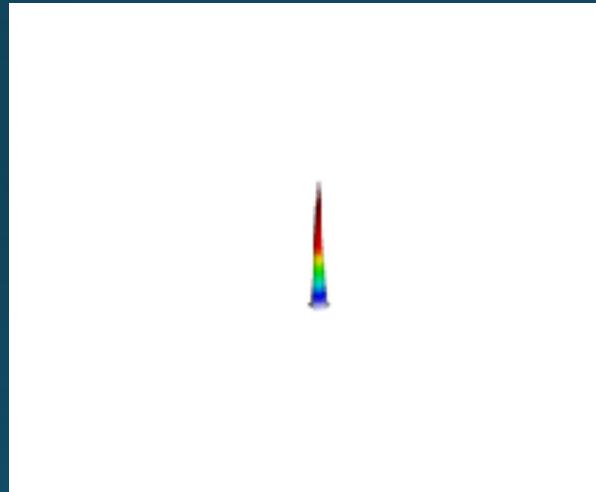
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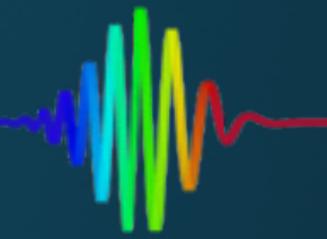


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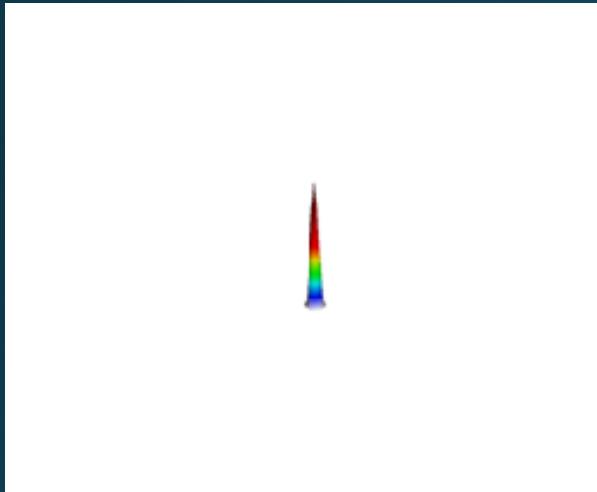


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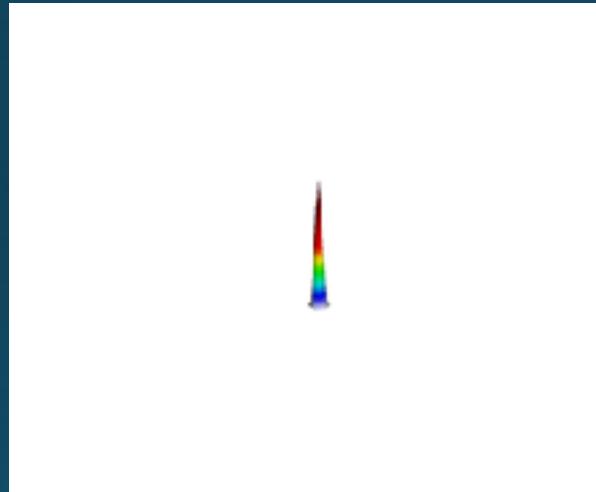
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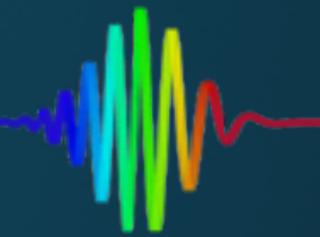


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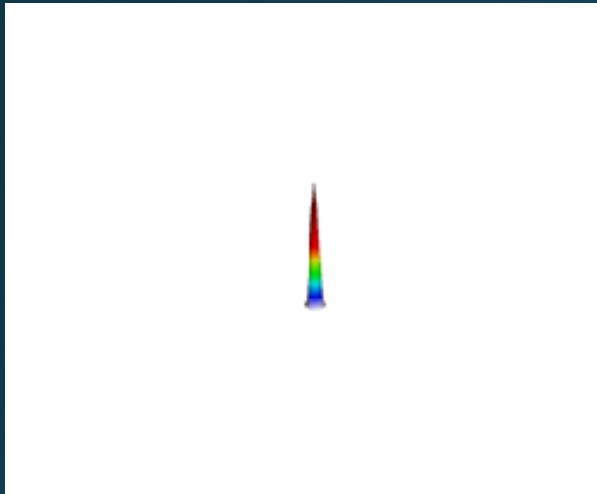


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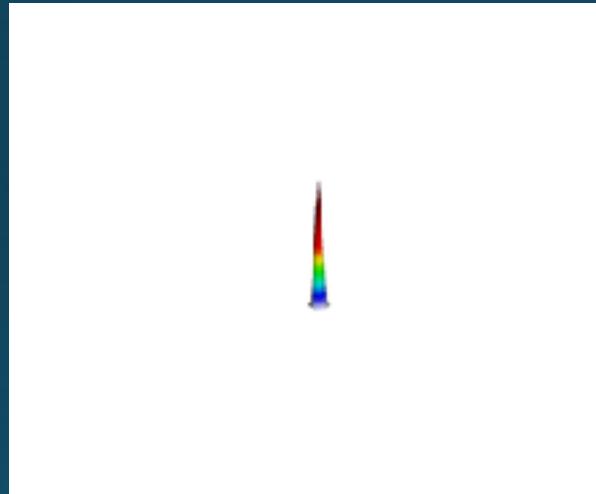
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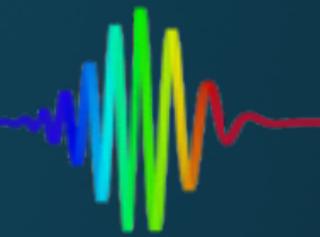


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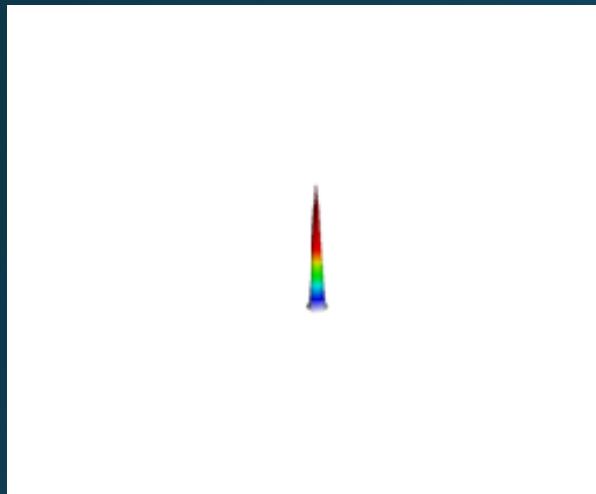


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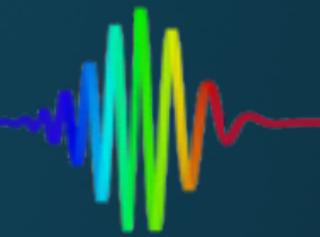


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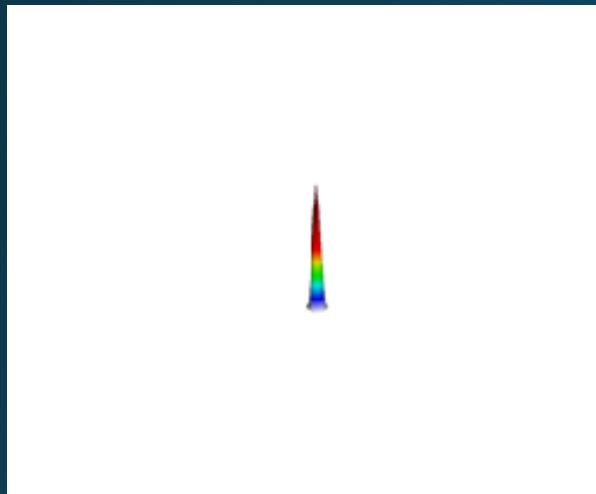


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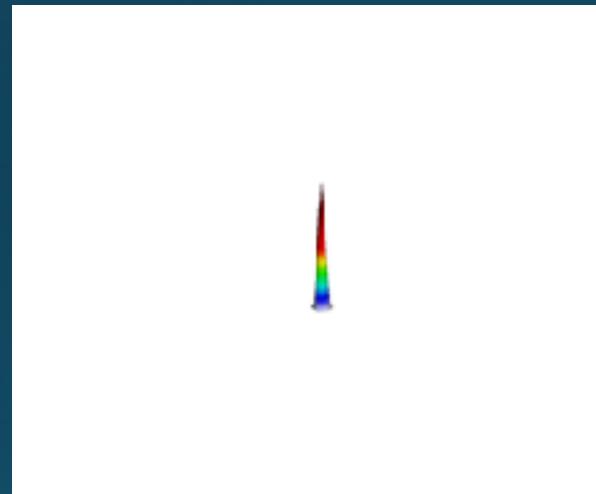
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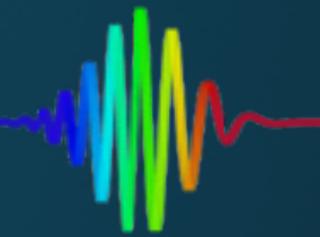


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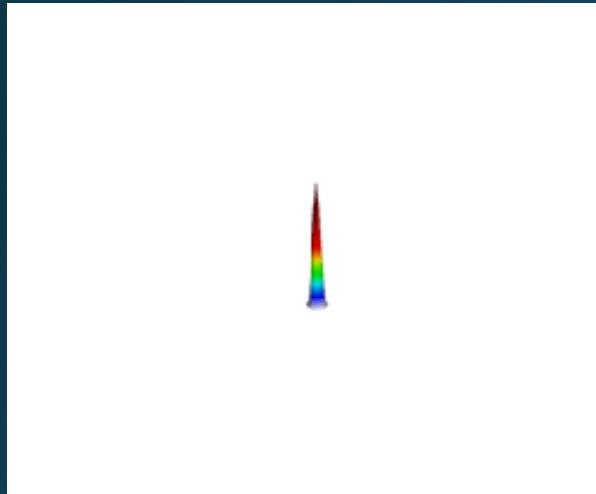


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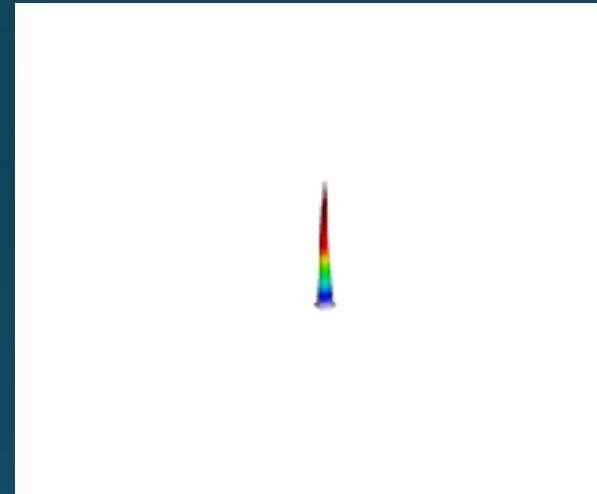
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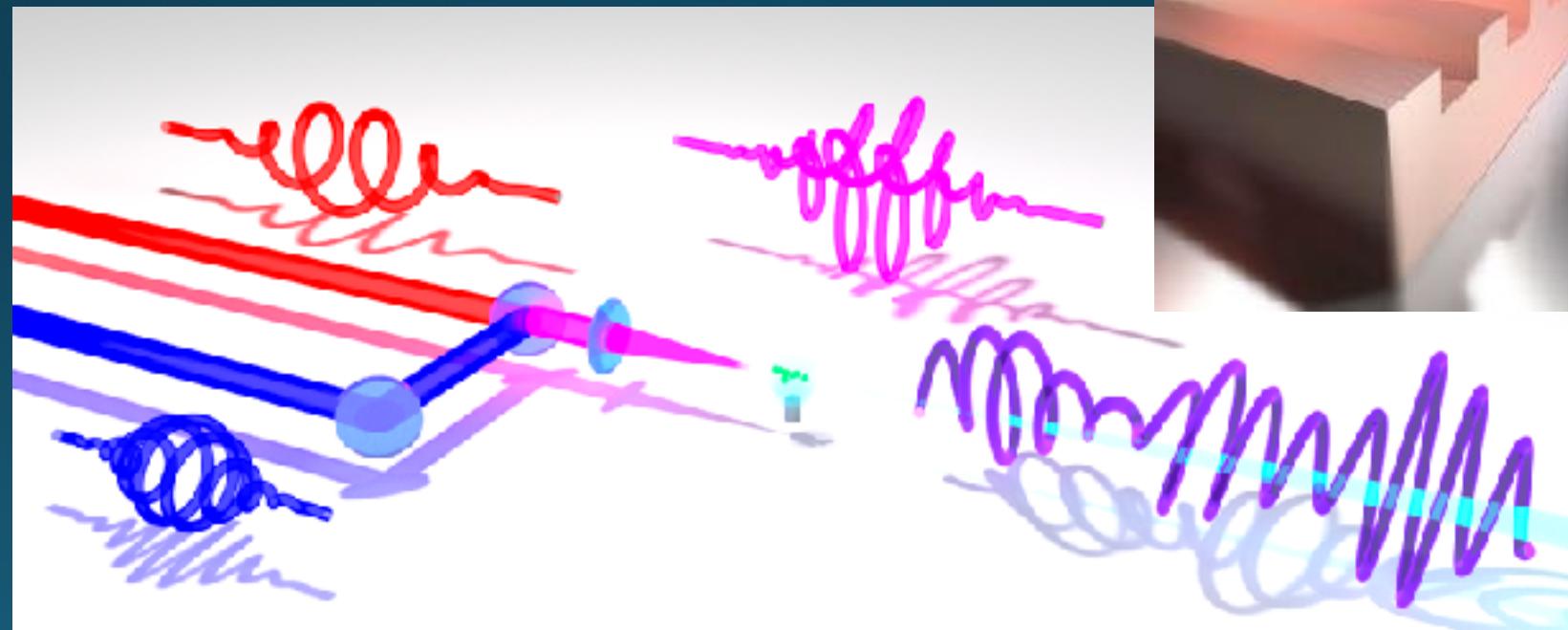
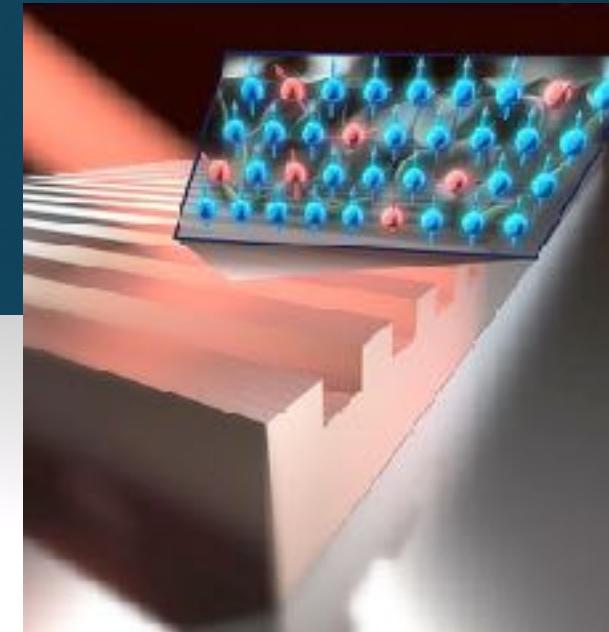
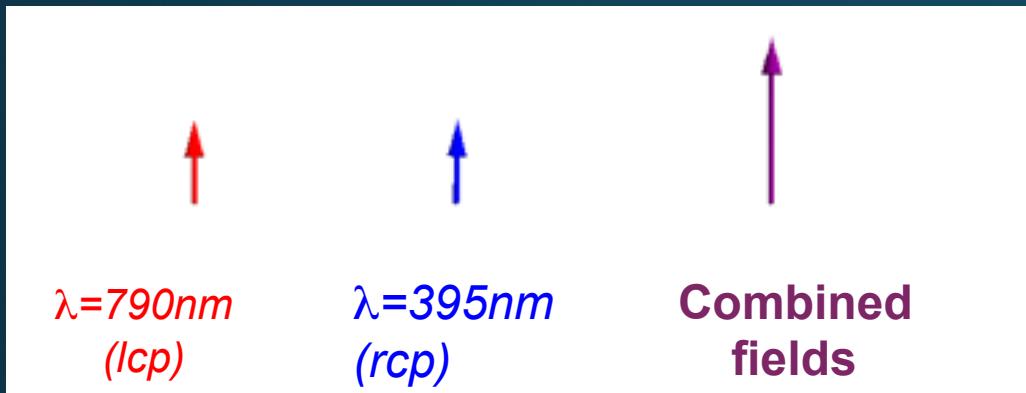
One-Color Circular Driver



Two-Color Counter-Rotating

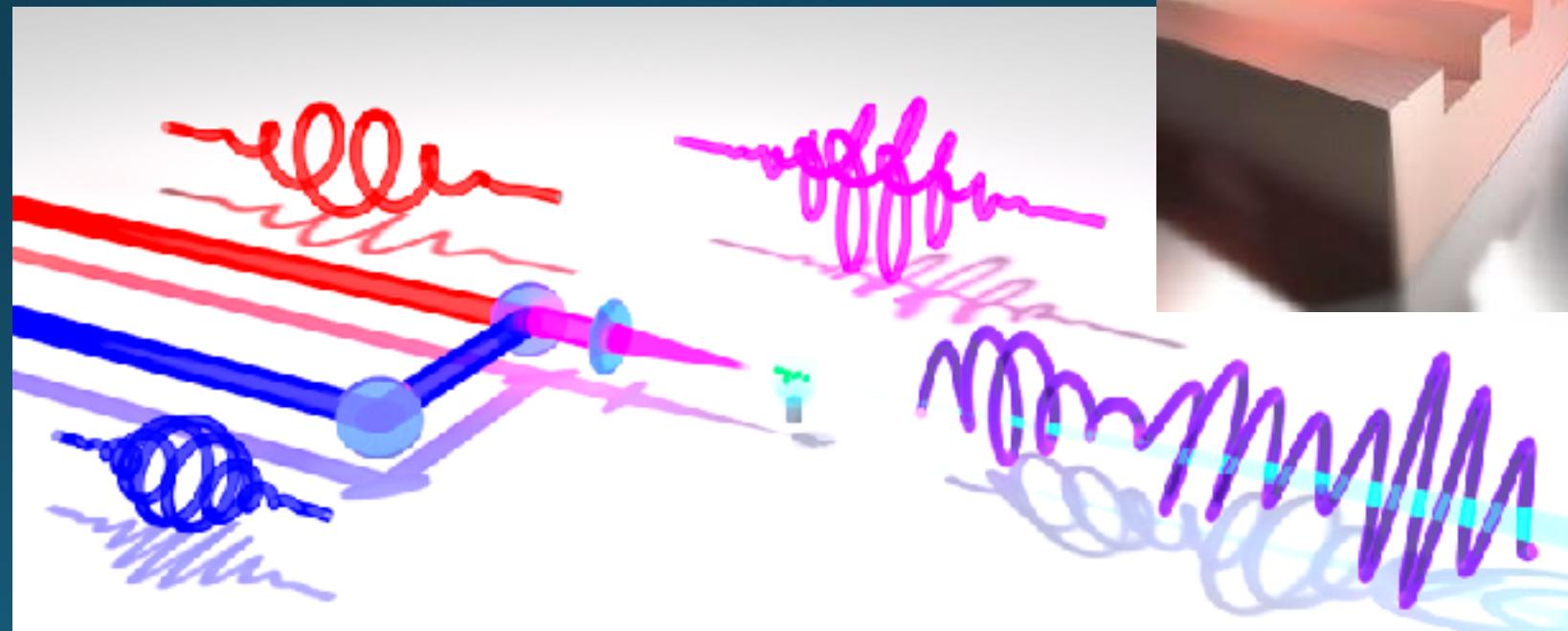
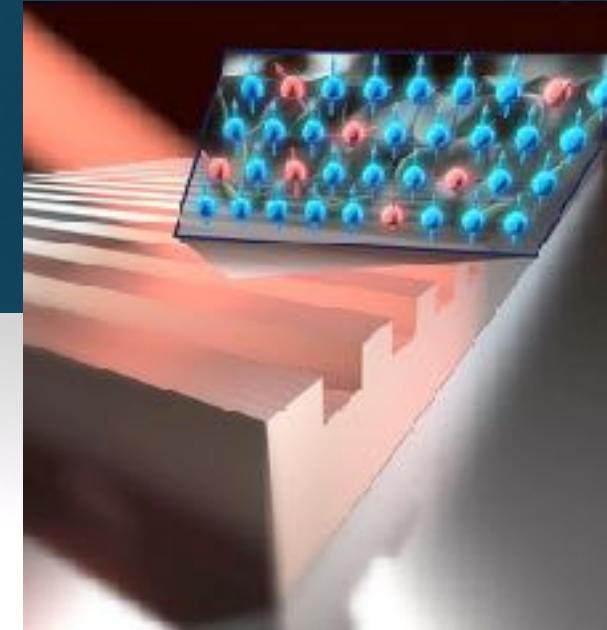
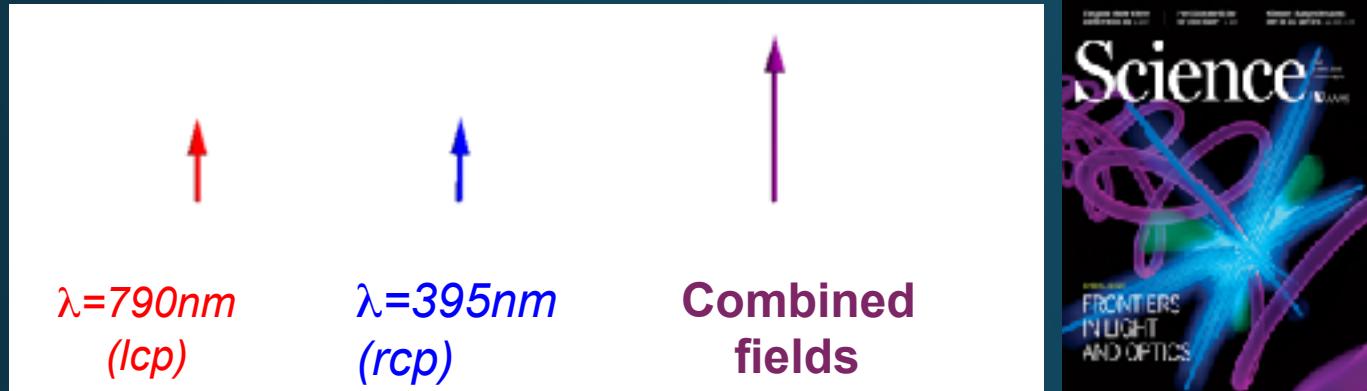
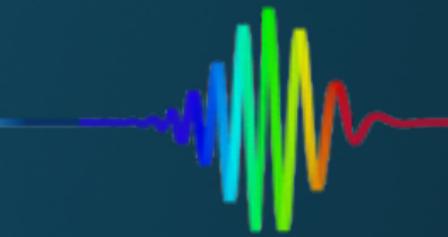


Circularly Polarized High Harmonic Generation (CPHHG): The Attosecond Blast(s) From the Past!



Eichmann, Phys. Rev. A. 51, 1995
Long, Phys. Rev. A. 52, 1995
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Fan, PNAS, 112, 2015
Dorney, PRL, 119, 2017
Many, many, more

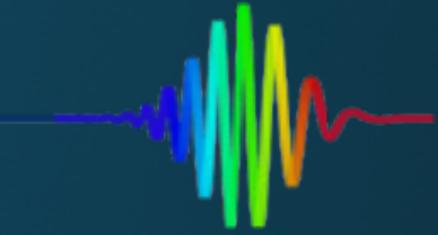
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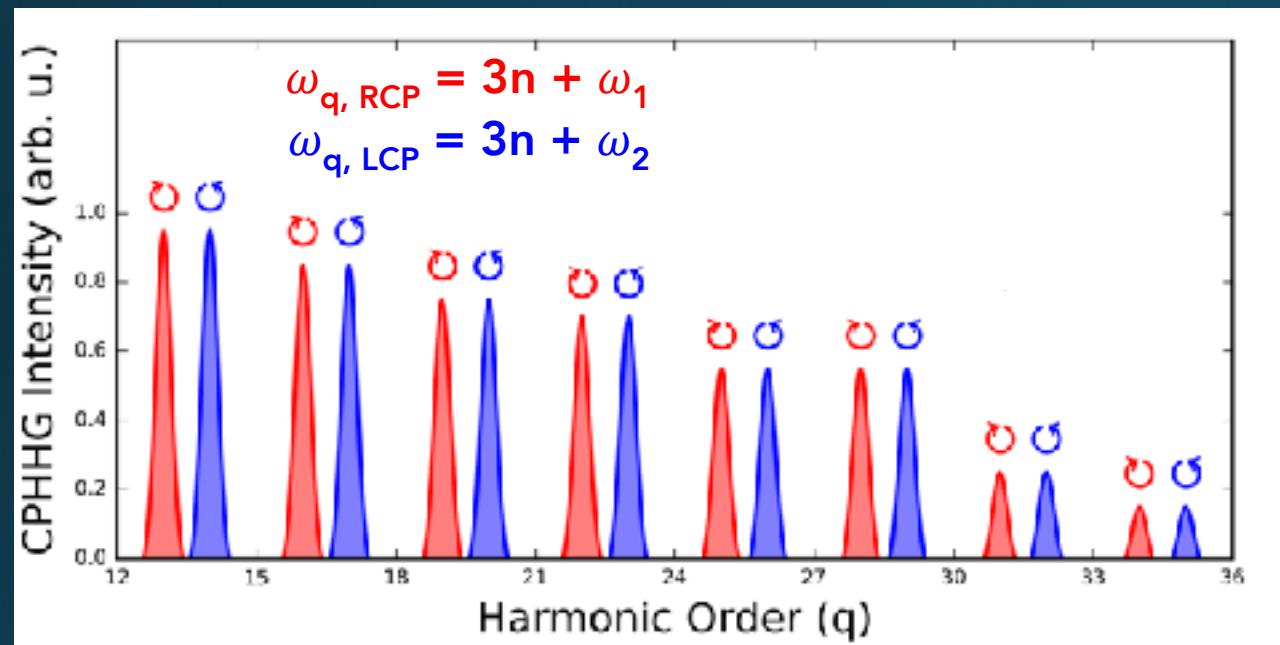
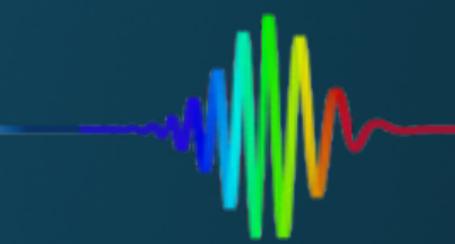
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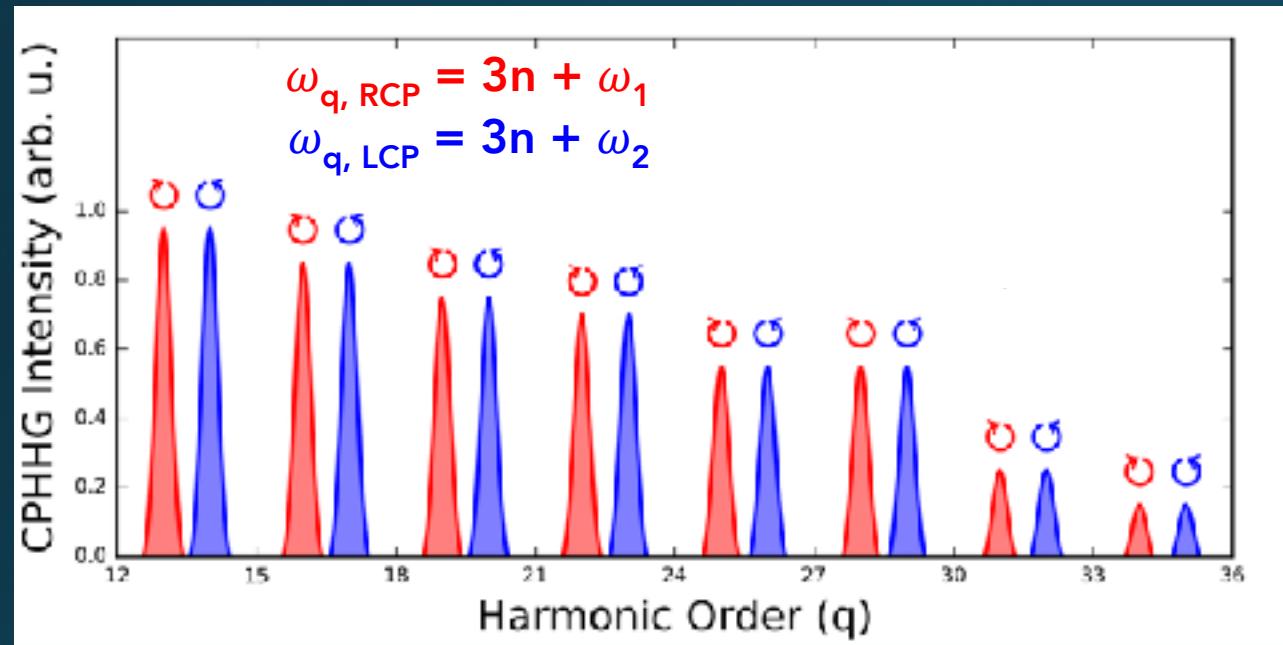
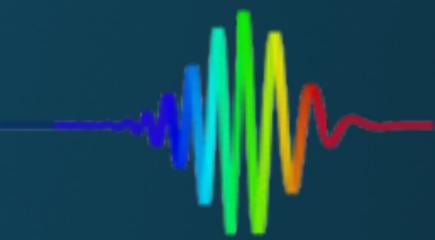
Circularly Polarized Harmonics, You Say?
Well, Surely We Have Circular Attosecond Pulses!



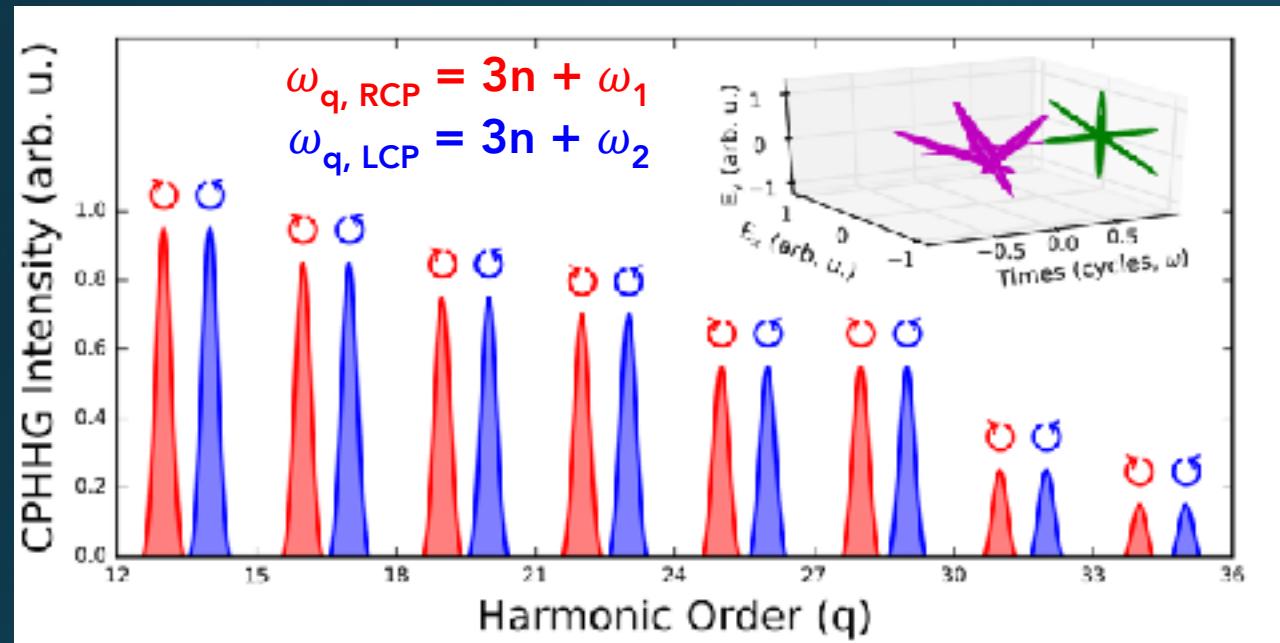
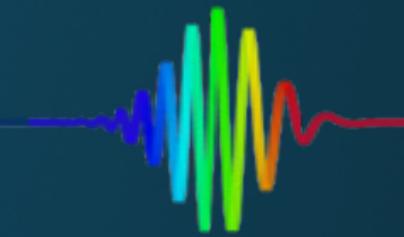
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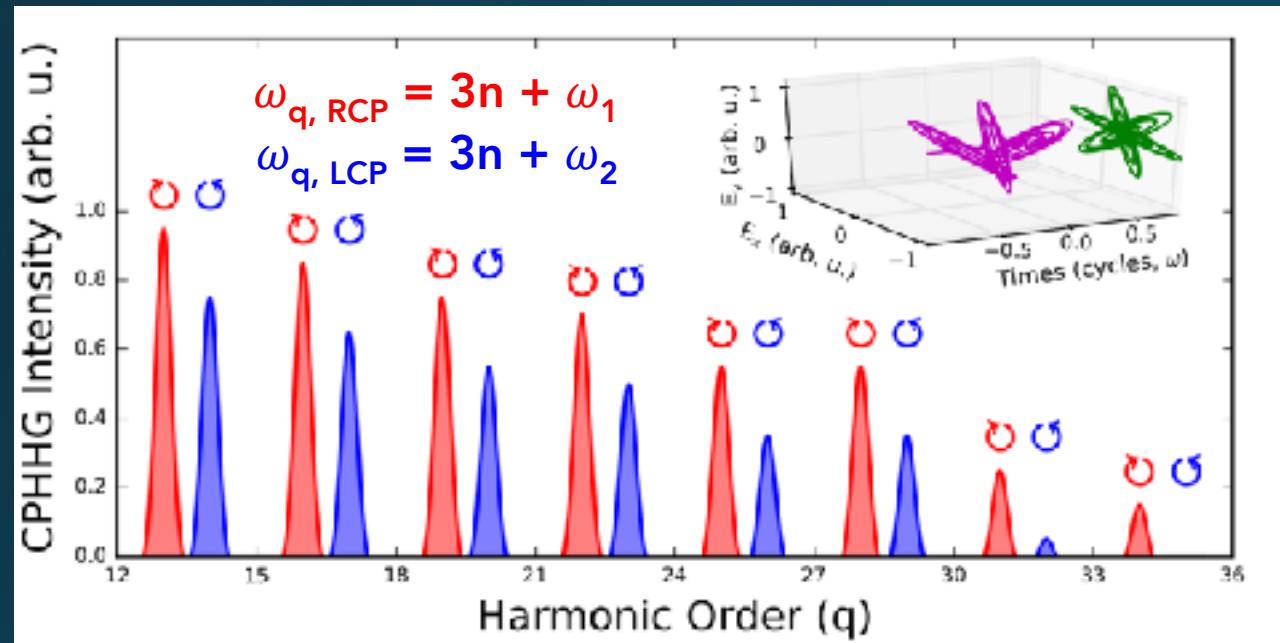
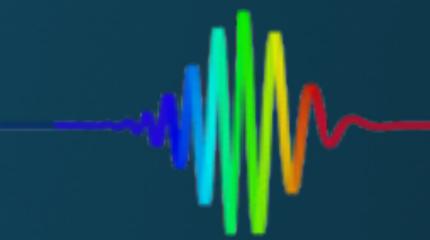
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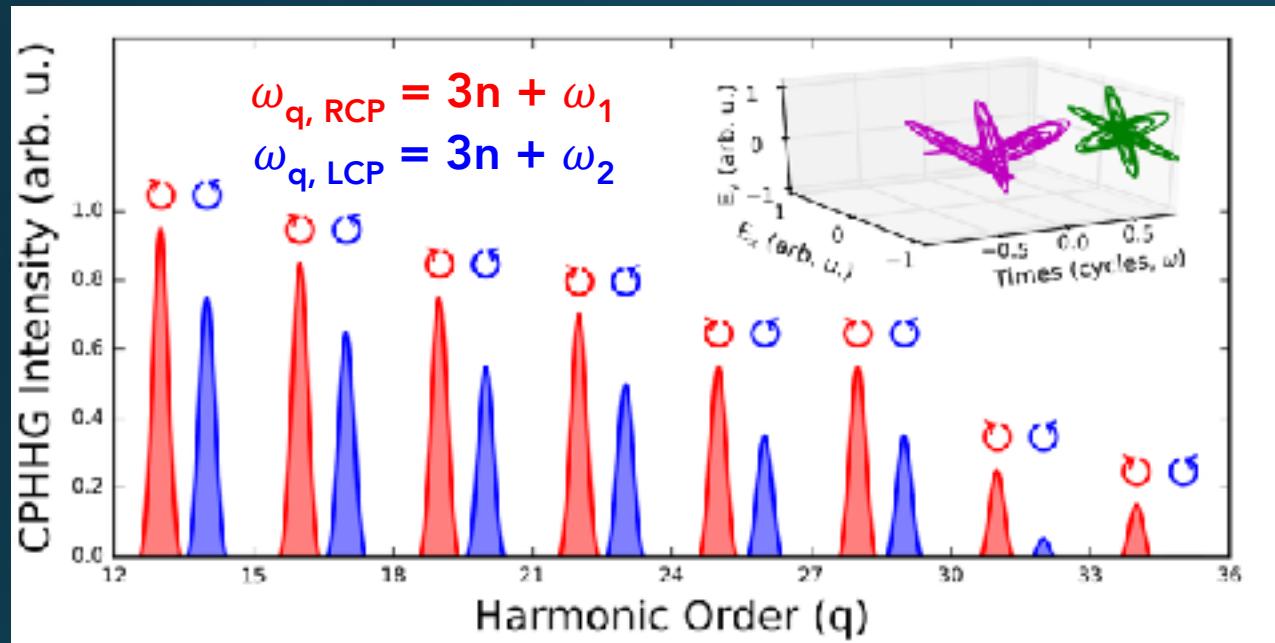
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Kfir, J. Phys. B., 49, 2016

Li, Opt. Quant. Electron. 49, 2017

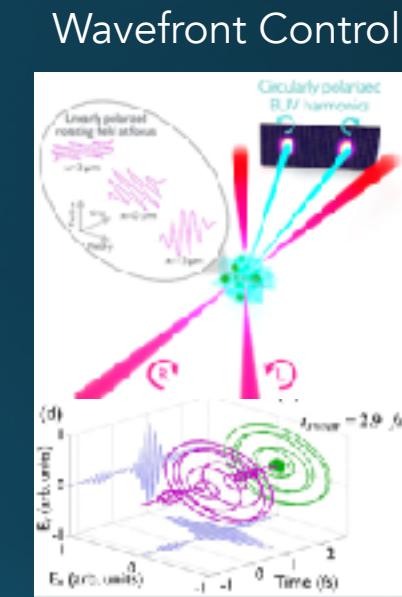
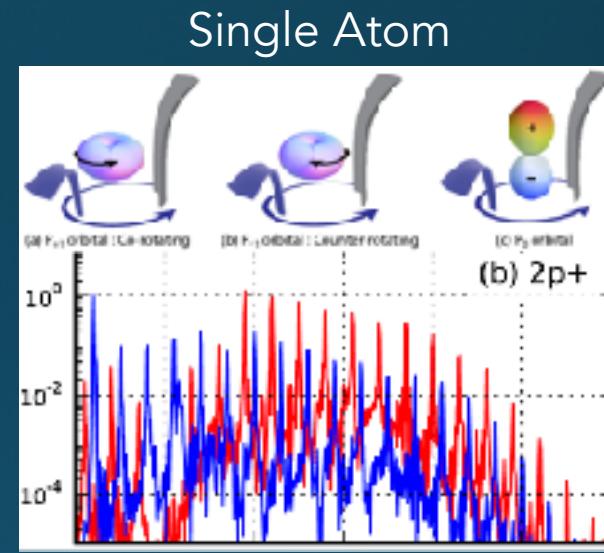
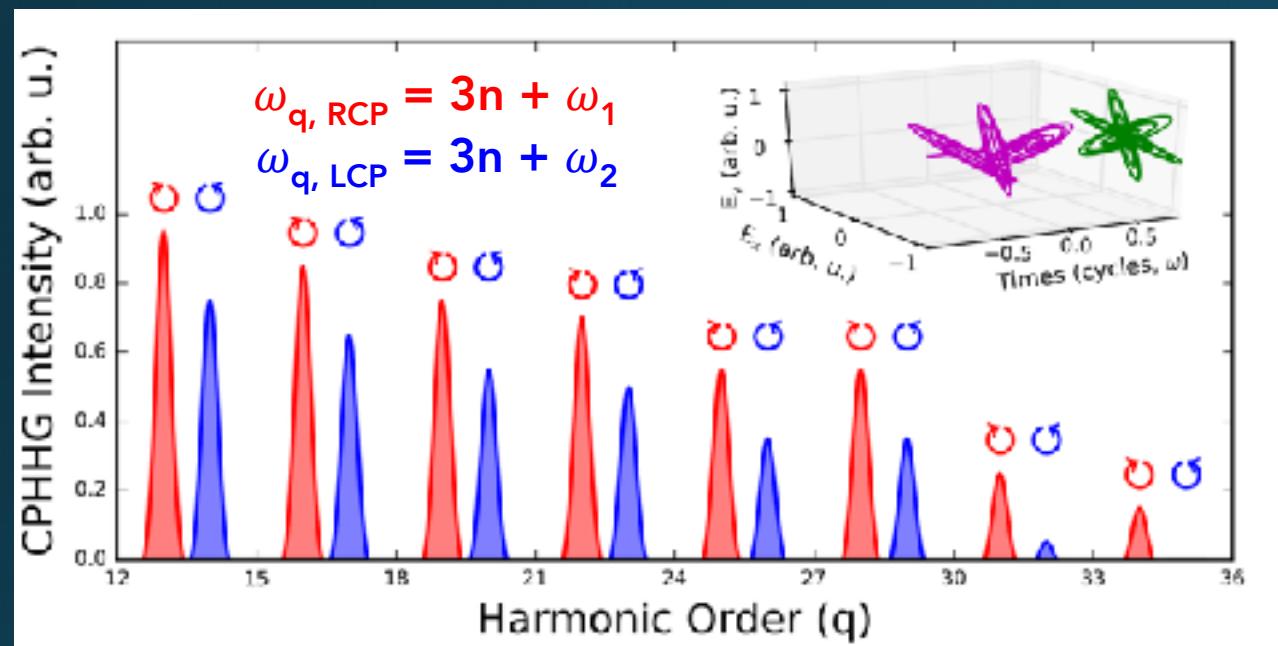
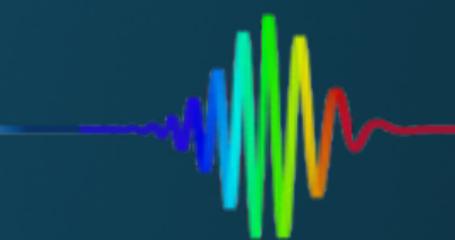
Lerner, Opt. Lett. 42, 2017

Skantzakis, Sci. Rep., 6, 2016

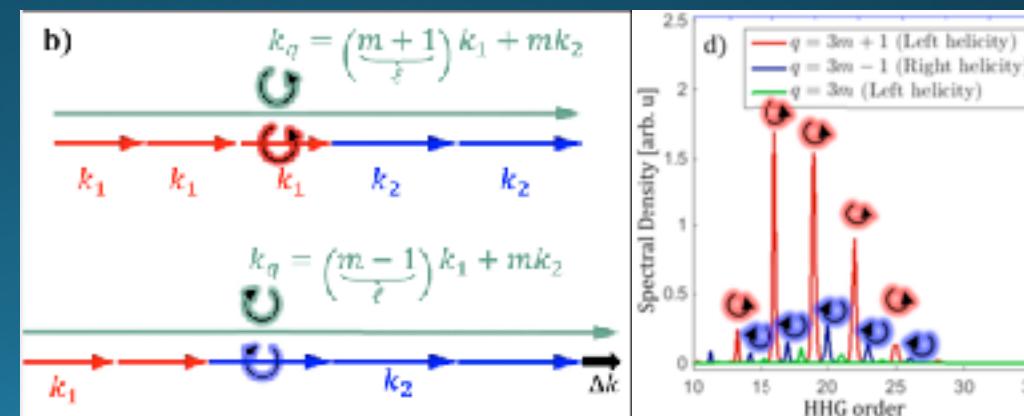
Yuan, Phys. Rev. Lett. 110, 2013

Zhang, Opt. Lett. 42, 2017

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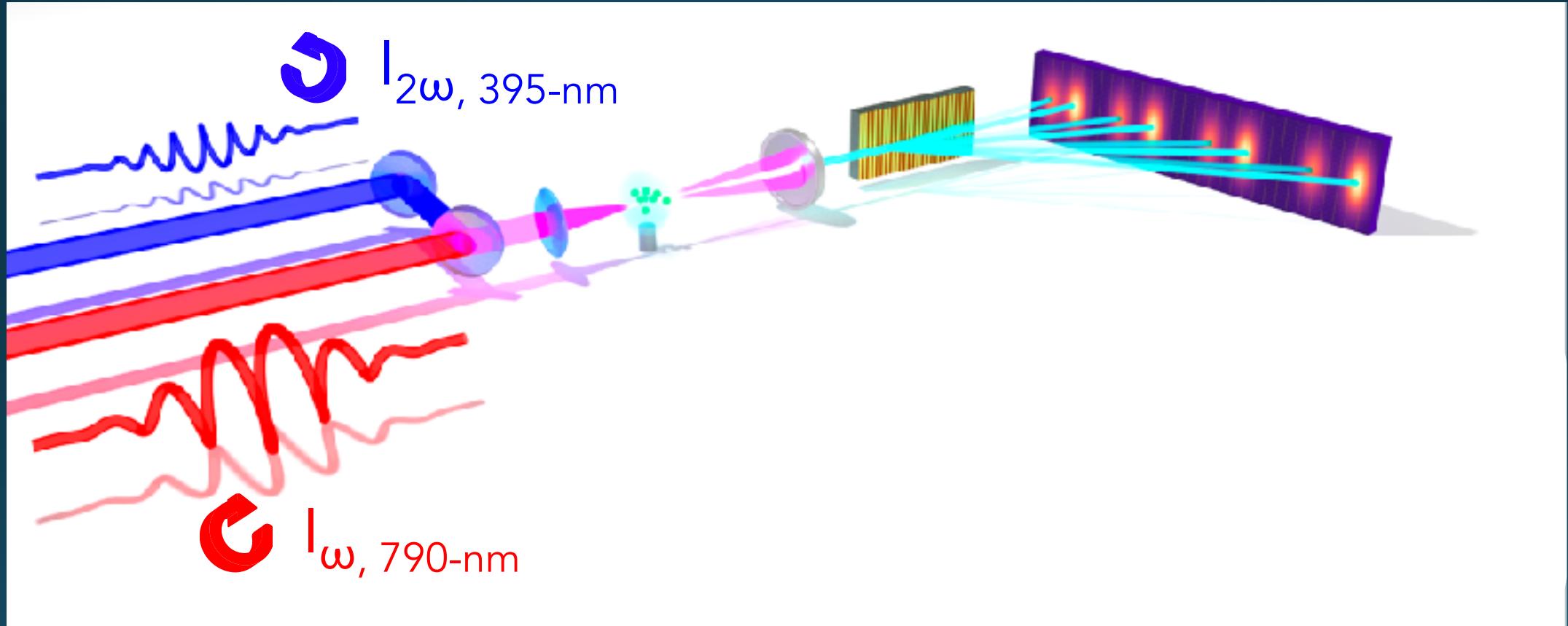
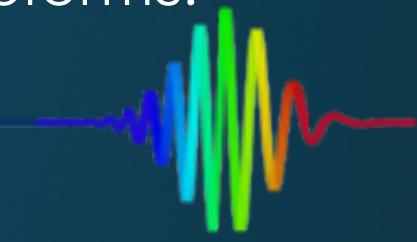


Macroscopic Control

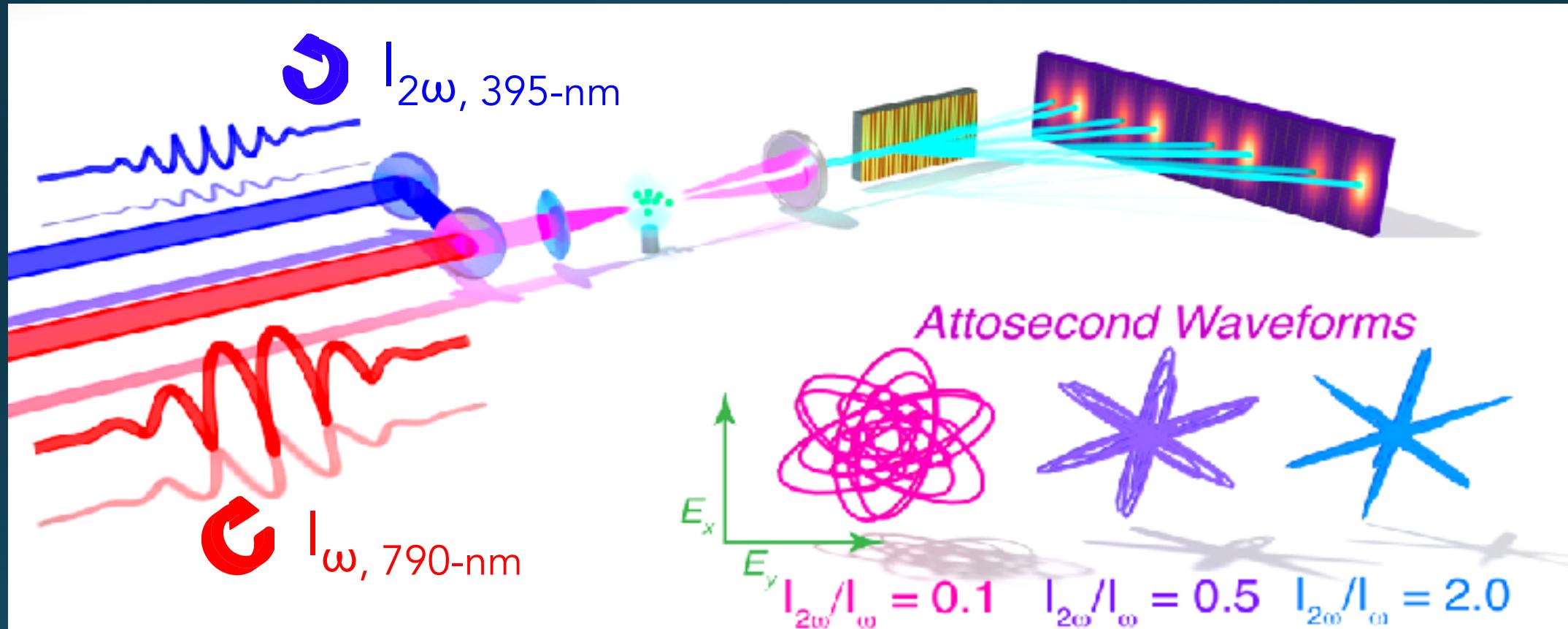
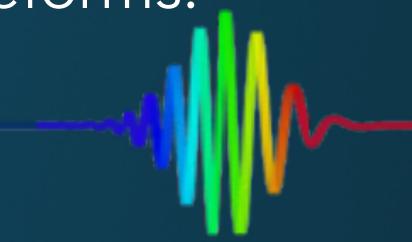


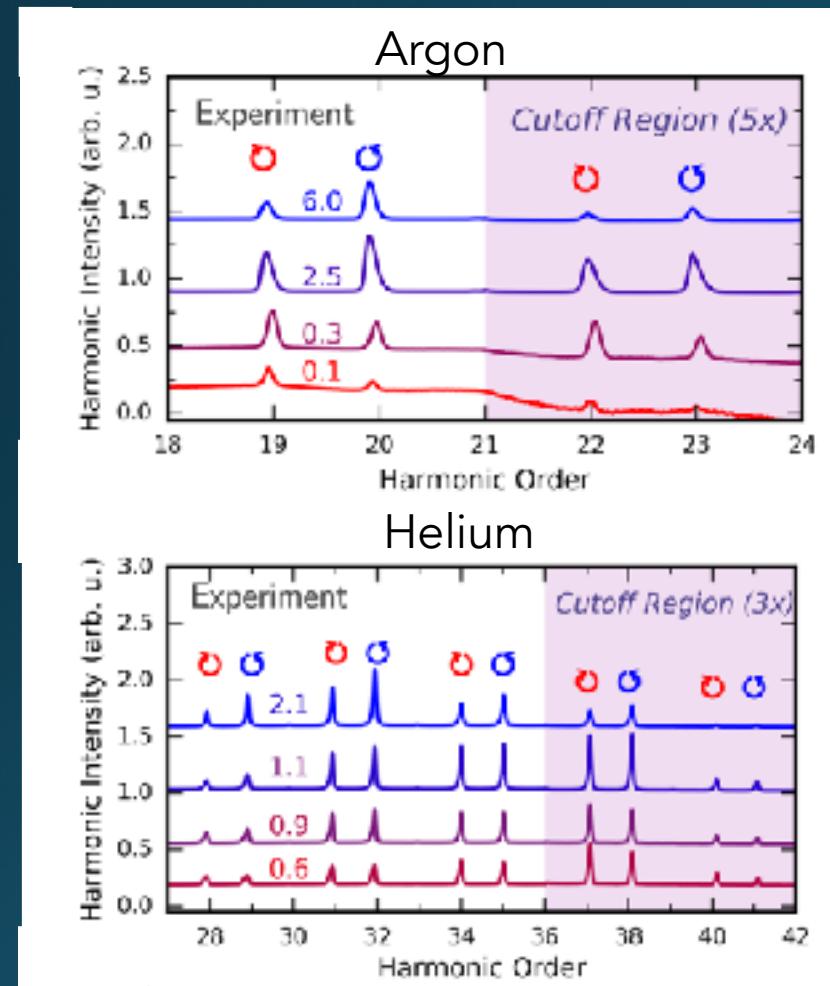
- Medisauskas, PRL, 115, 2015
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Active Control Over the Polarization of High-Harmonic Waveforms: Production of Elliptically Polarized Attosecond Pulse Trains!



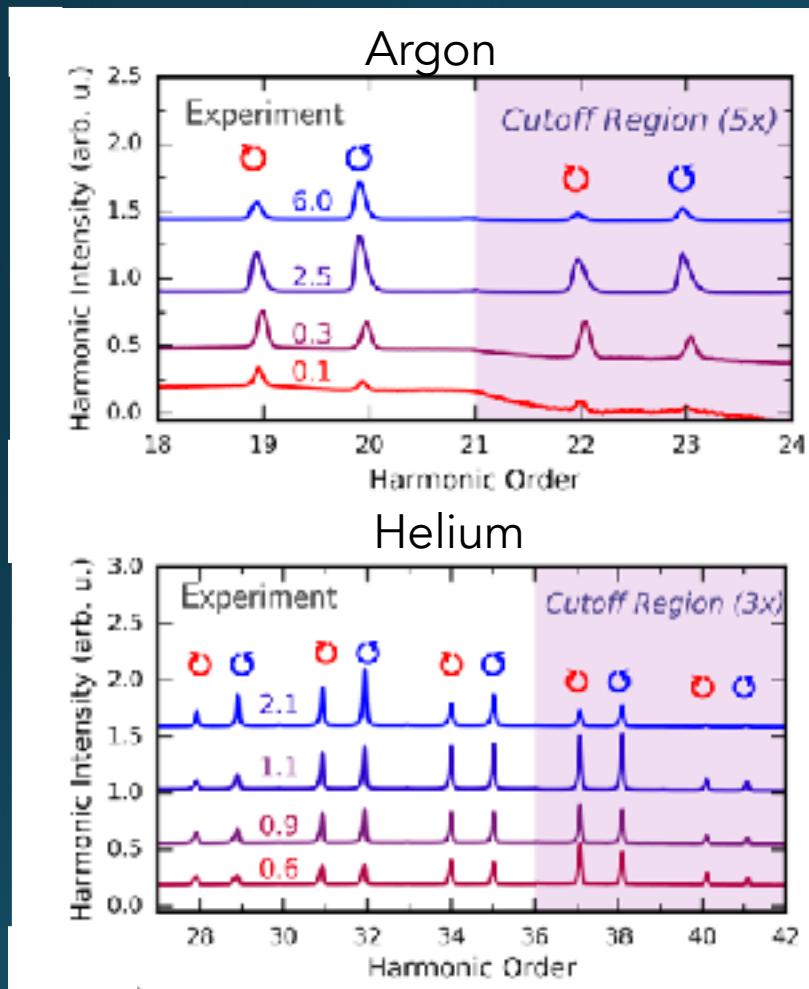
Dorney et. al., Phys. Rev. Lett., 119, 2017



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

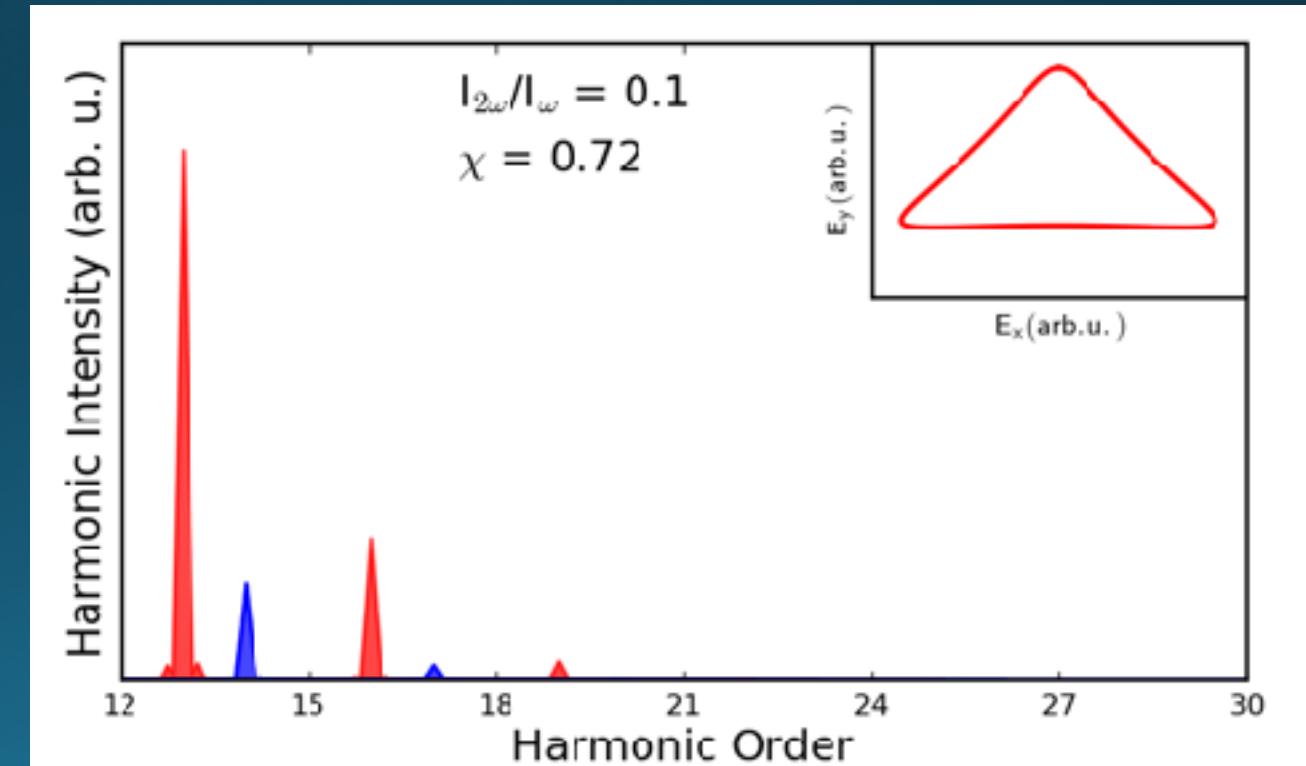
Dorney et. al., Phys. Rev. Lett., 119, 2017

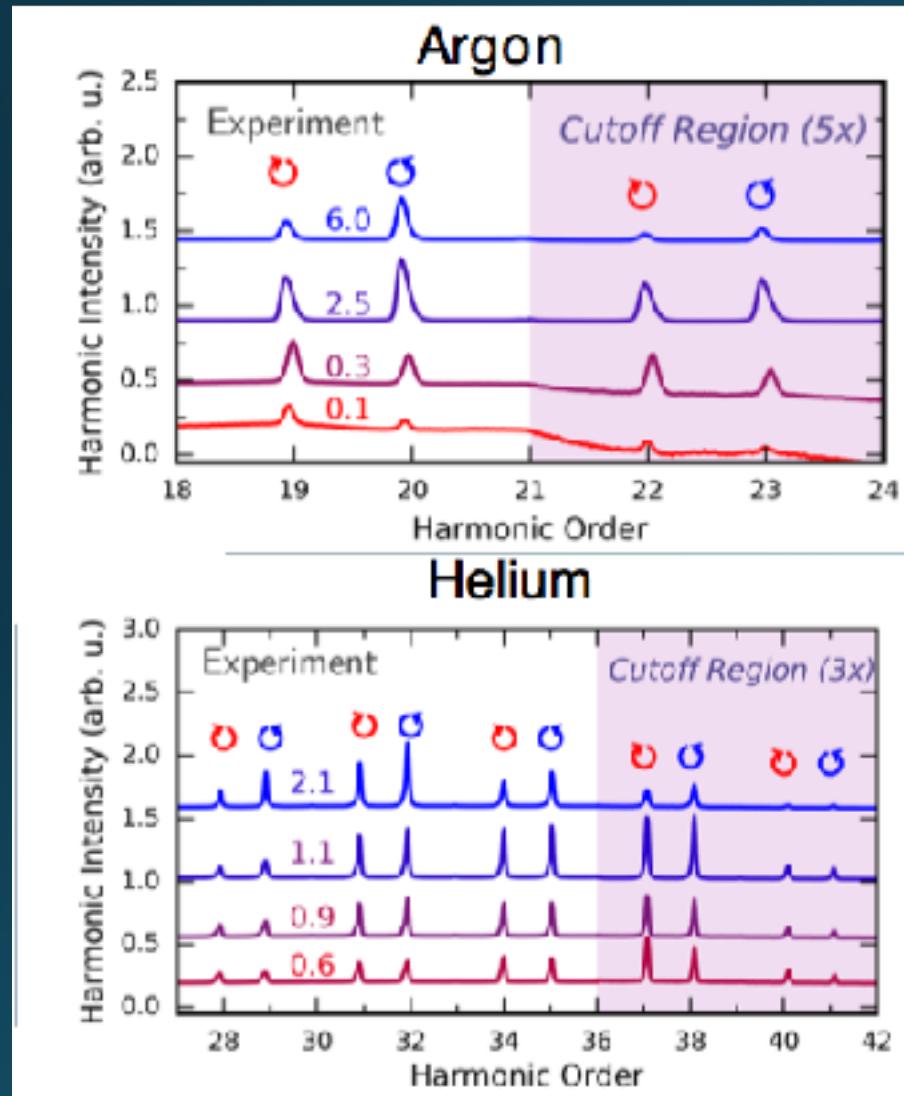
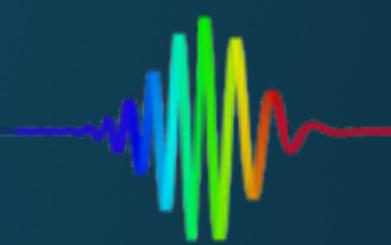
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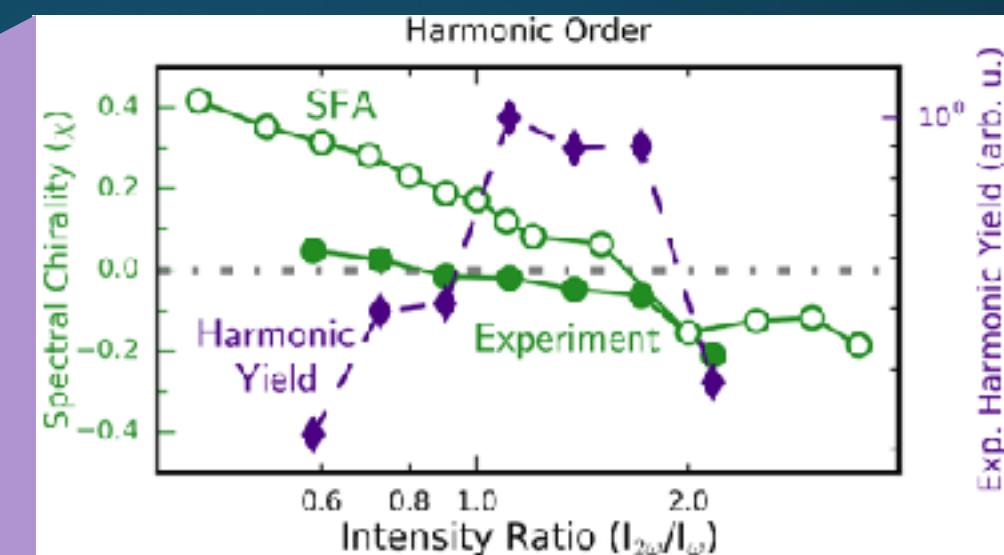
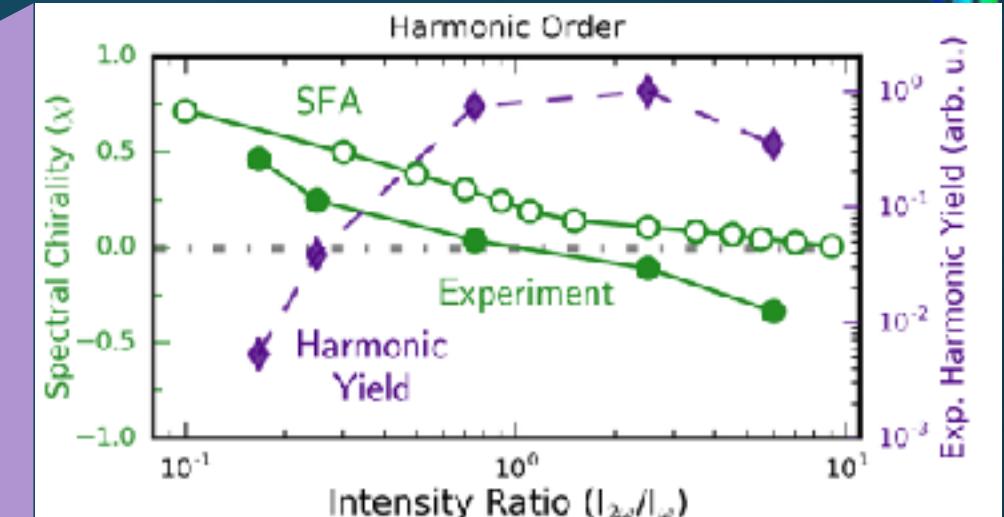
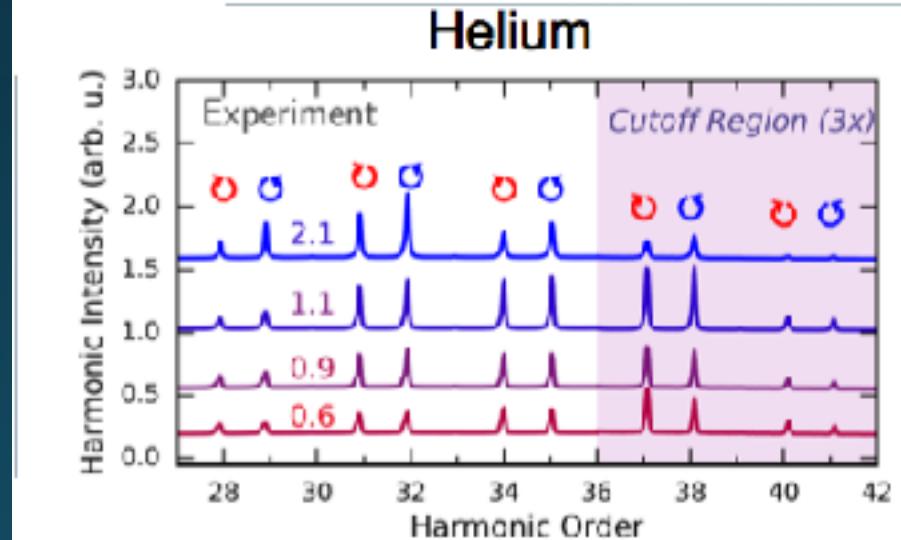
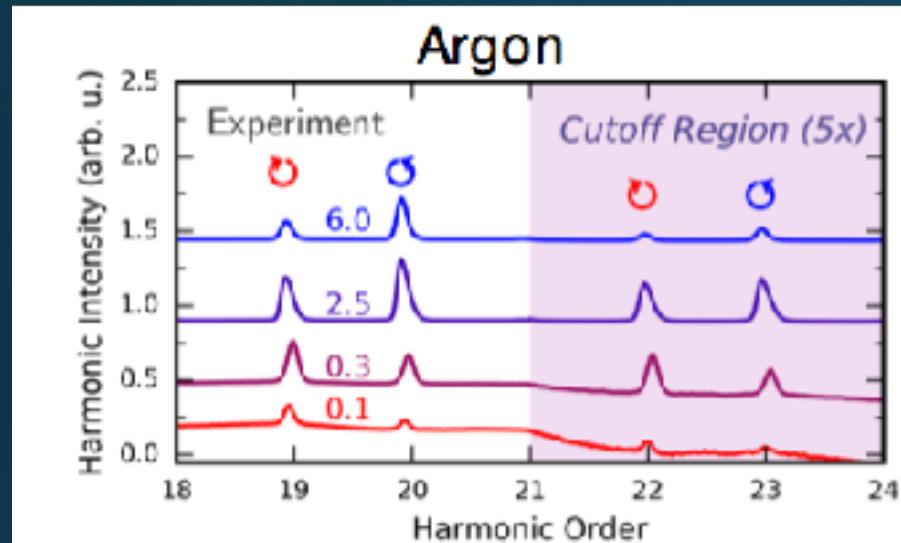


$$\chi = \frac{(I_{RCP} - I_{LCP})}{(I_{RCP} + I_{LCP})}$$

SFA Simulation in Ar

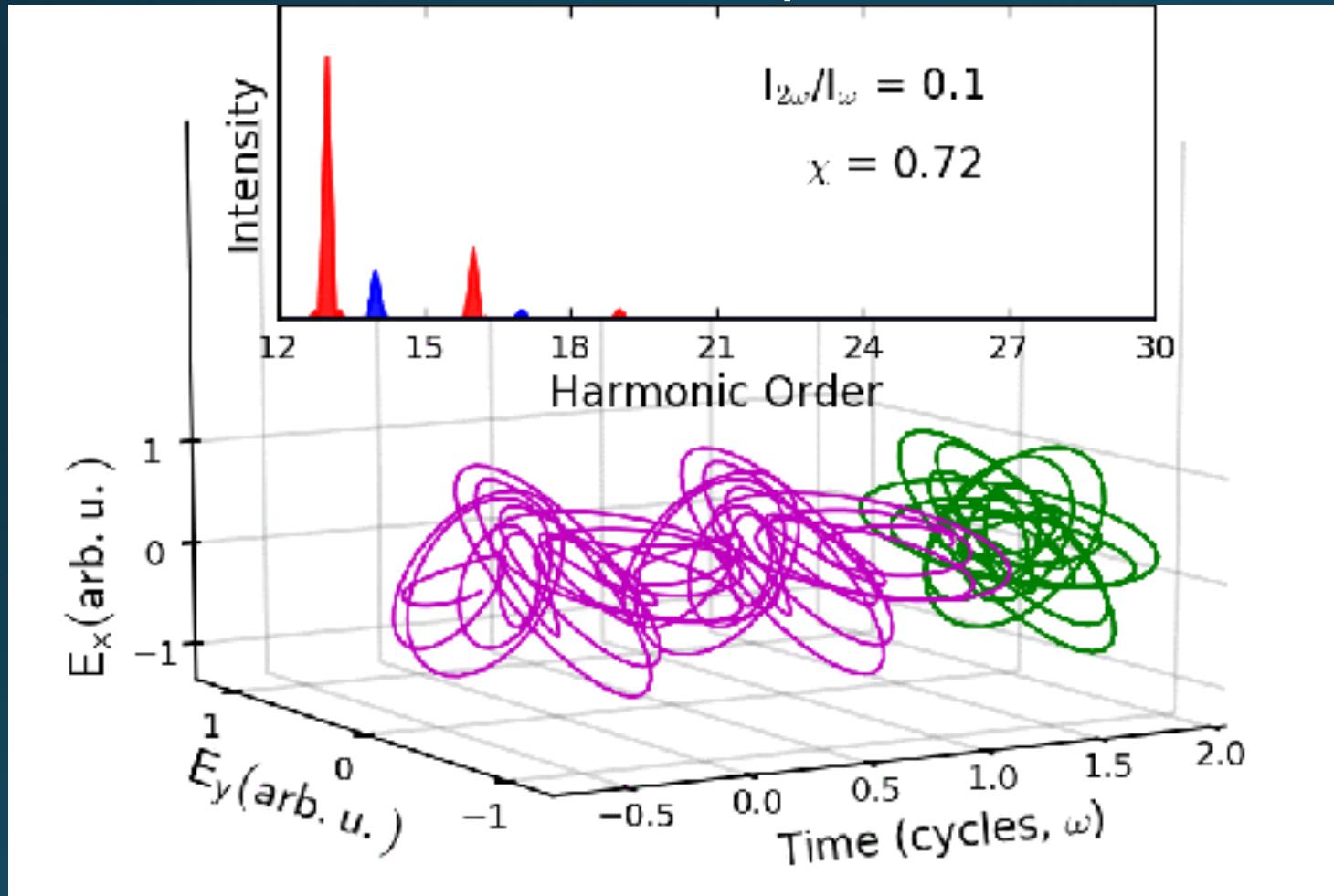






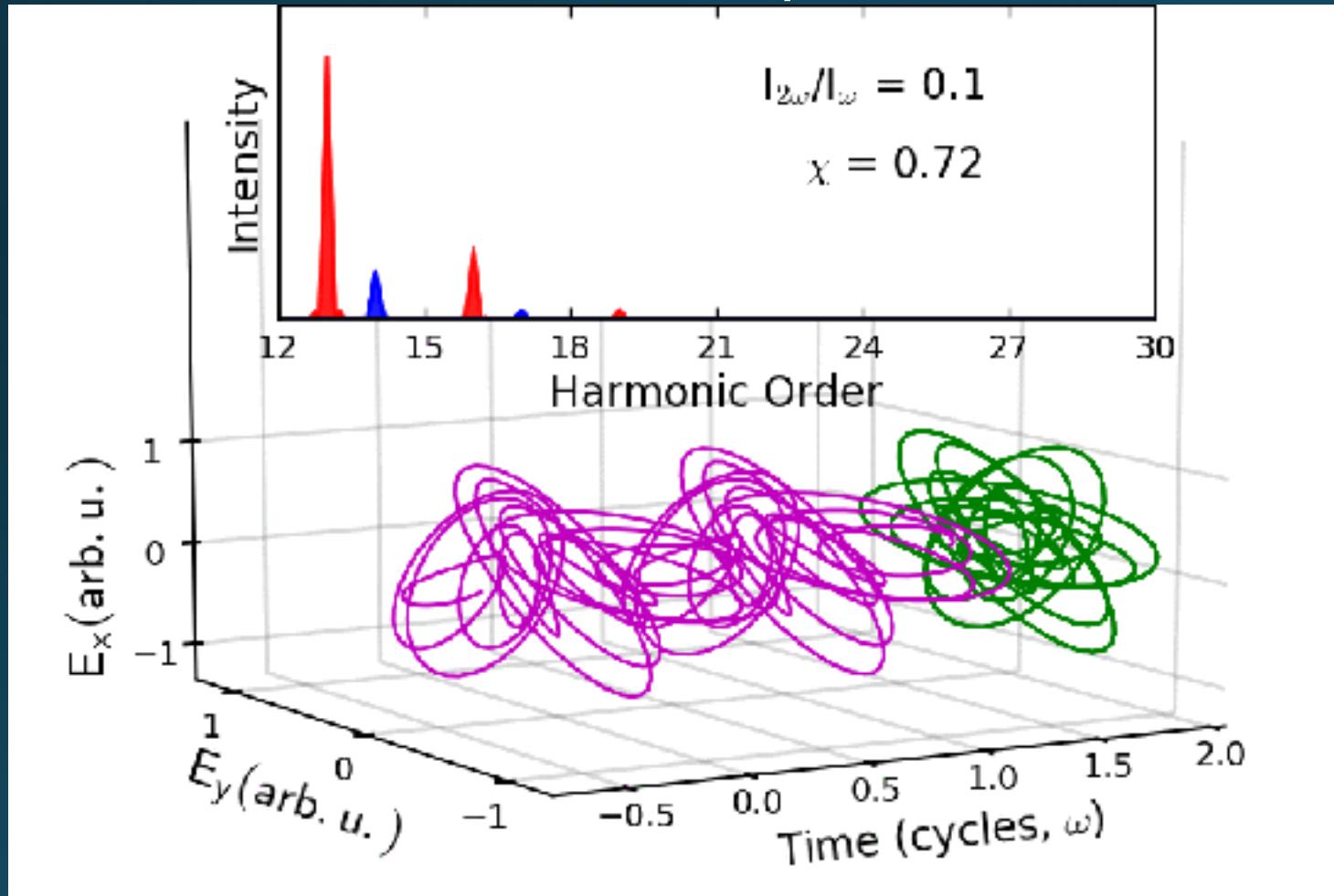


➤ First demonstration of real-time polarization control of attosecond pulse trains in CPHHG!

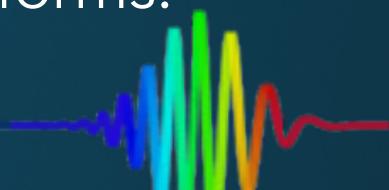




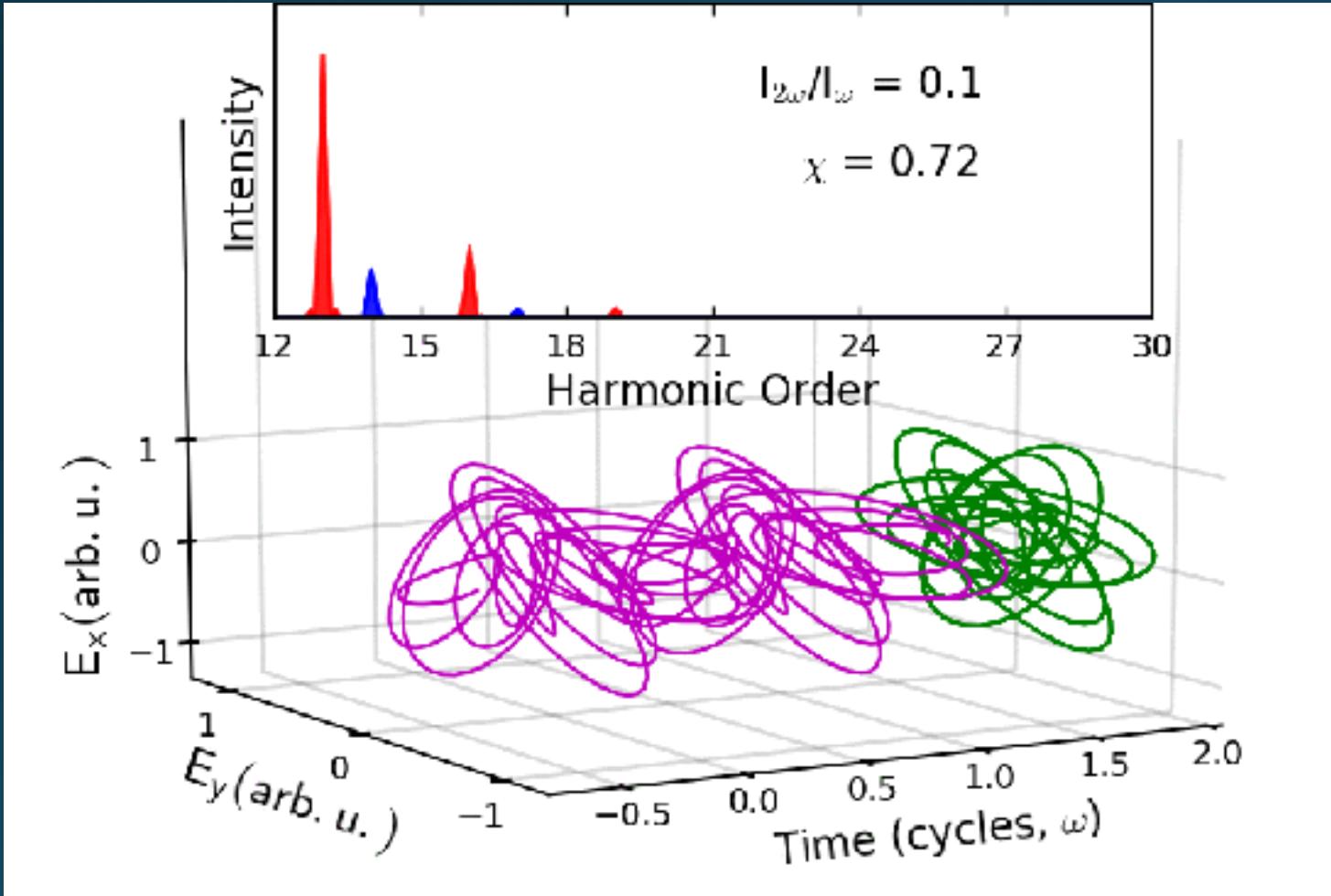
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Active Control Over the Polarization of High-Harmonic Waveforms: Production of Elliptically Polarized Attosecond Pulse Trains!



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Neufeild, et al., arXiv:1709.06261, 2017

Instantaneous Optical Chirality

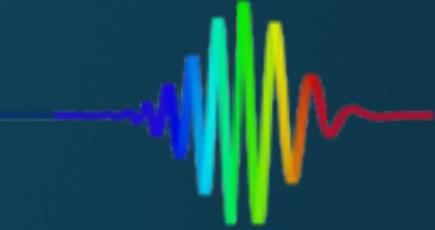
$$C_P = \frac{1}{c_0^2} \dot{\phi}(t) I(t)$$

Non-Instantaneous Optical Chirality

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



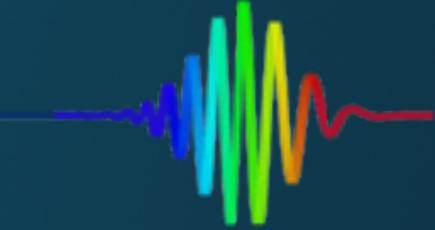
Simple Interpretation of Elliptical Control in CPHHG: Perturbative-(ish) Photon Absorption Model



Dorney et al., Phys. Rev. Lett., 119, 2017
Pisanty et al., Phys. Rev. A, 90, 2014
Li, et al., arXiv:1702.04084, 2017



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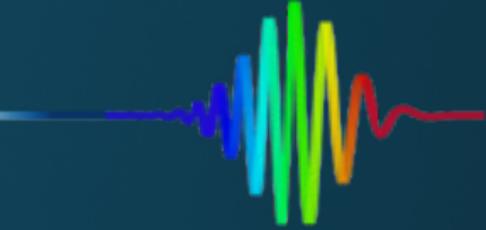


$$I_q \propto P(\Omega) \propto \sum_{i=0}^{\infty} p_1^{|n_1^i|} p_2^{|n_2^i|}$$

Dorney et al., Phys. Rev. Lett., 119, 2017
Pisanty et al., Phys. Rev. A, 90, 2014
Li, et al., arXiv:1702.04084, 2017



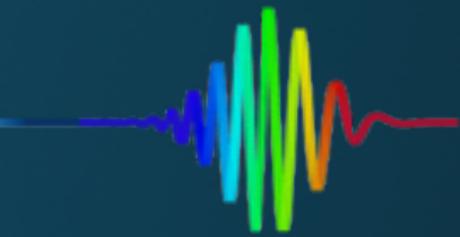
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$$I_q \propto P(\Omega) \propto \sum_{i=0}^{\infty} p_1^{|n_1^i|} p_2^{|n_2^i|} \quad p_{\omega} = \frac{I_{\omega}}{I_{\omega} + I_{2\omega}} = \frac{1}{1 + I_{ratio}} \quad p_{2\omega} = \frac{I_{2\omega}}{I_{\omega} + I_{2\omega}} = \frac{1}{1 +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

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n = channel number

s = number of "emitted" photons

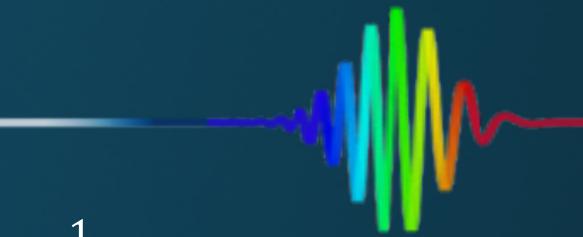
Channel (n, s)	$H_{19, RCP} (7\omega_1 + 6\omega_2)$	
	Total Photons	Statistical Scaling
(6, 0)	$7\omega_1 + 6\omega_2$	$p_{1\omega}$
(6, 1)	$7\omega_1 + 8\omega_2$	$p_{2\omega}$
(6, 2)	$9\omega_1 + 10\omega_2$	$p_{2\omega}$
(6, 3)	$11\omega_1 + 12\omega_2$	$p_{2\omega}$

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

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

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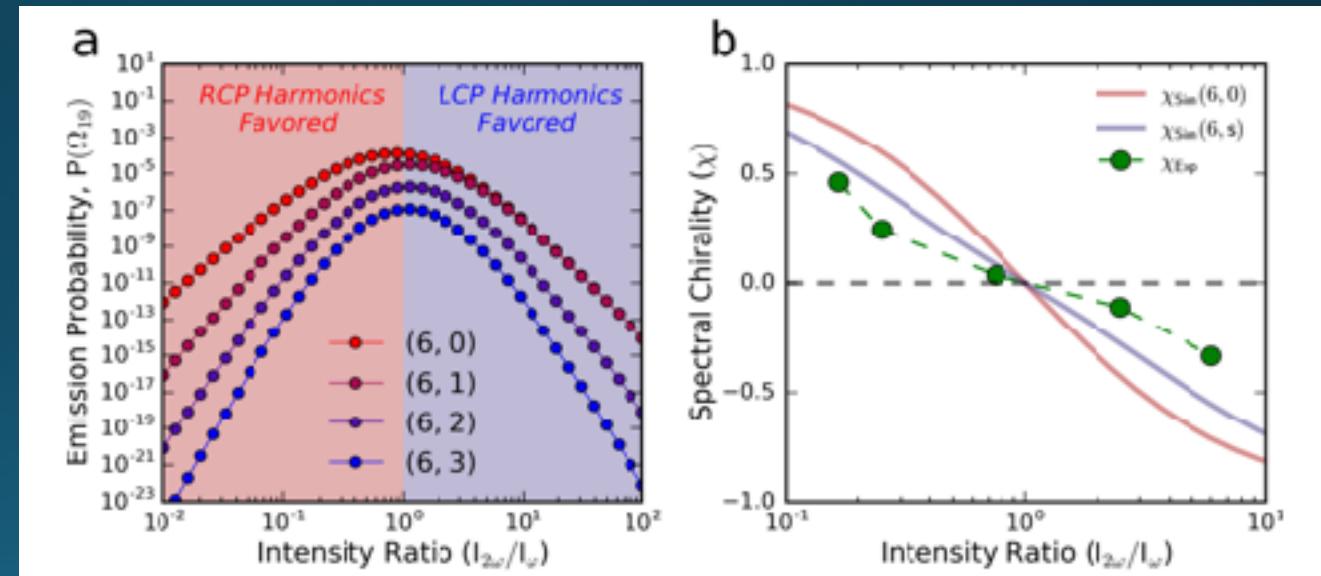


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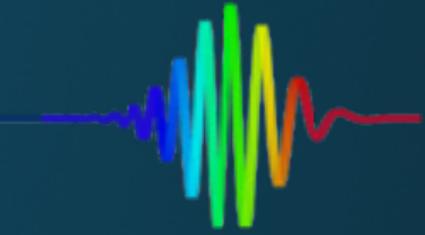
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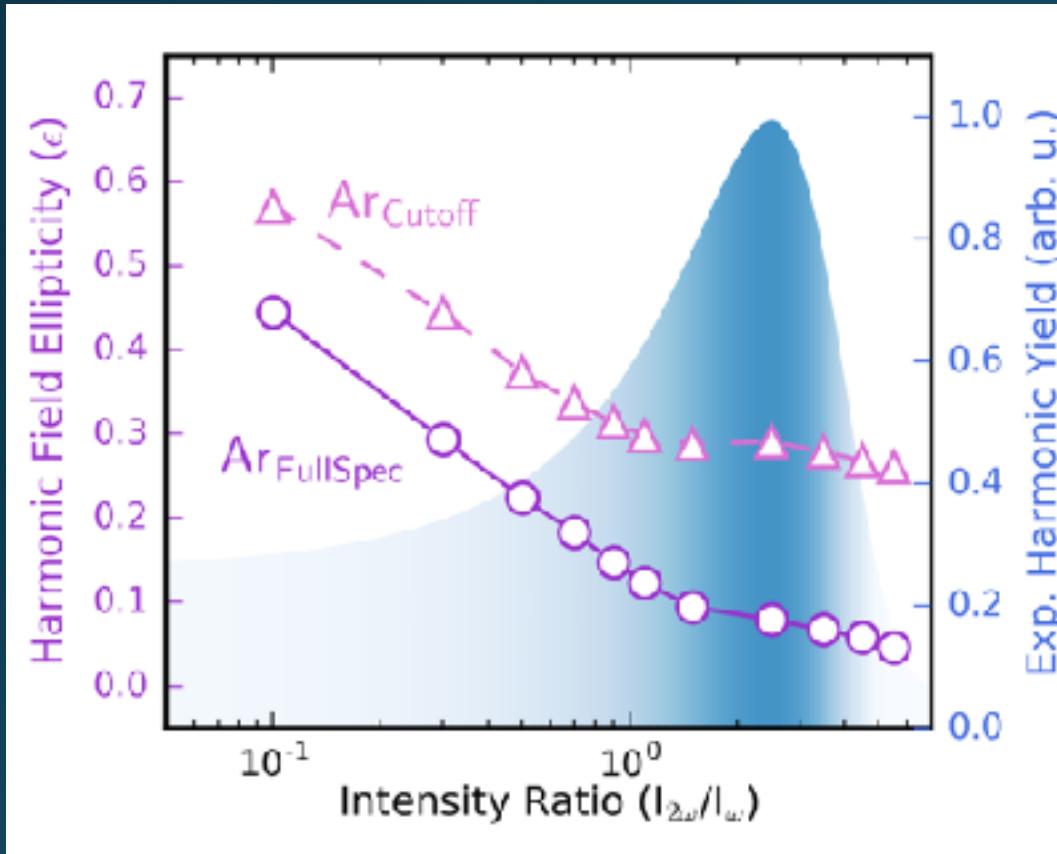
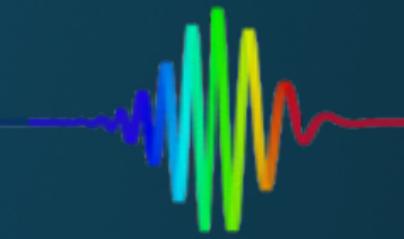
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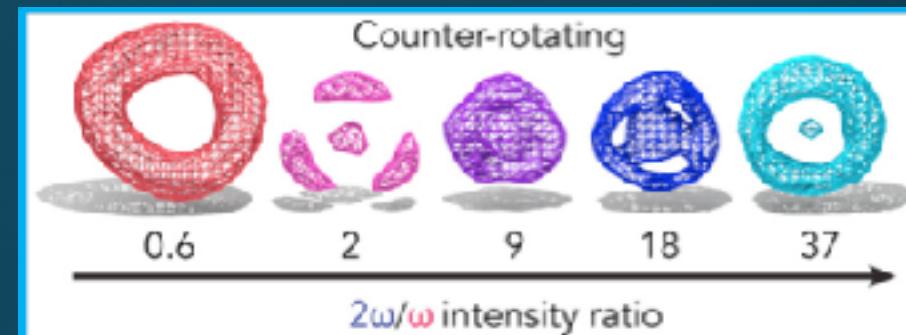
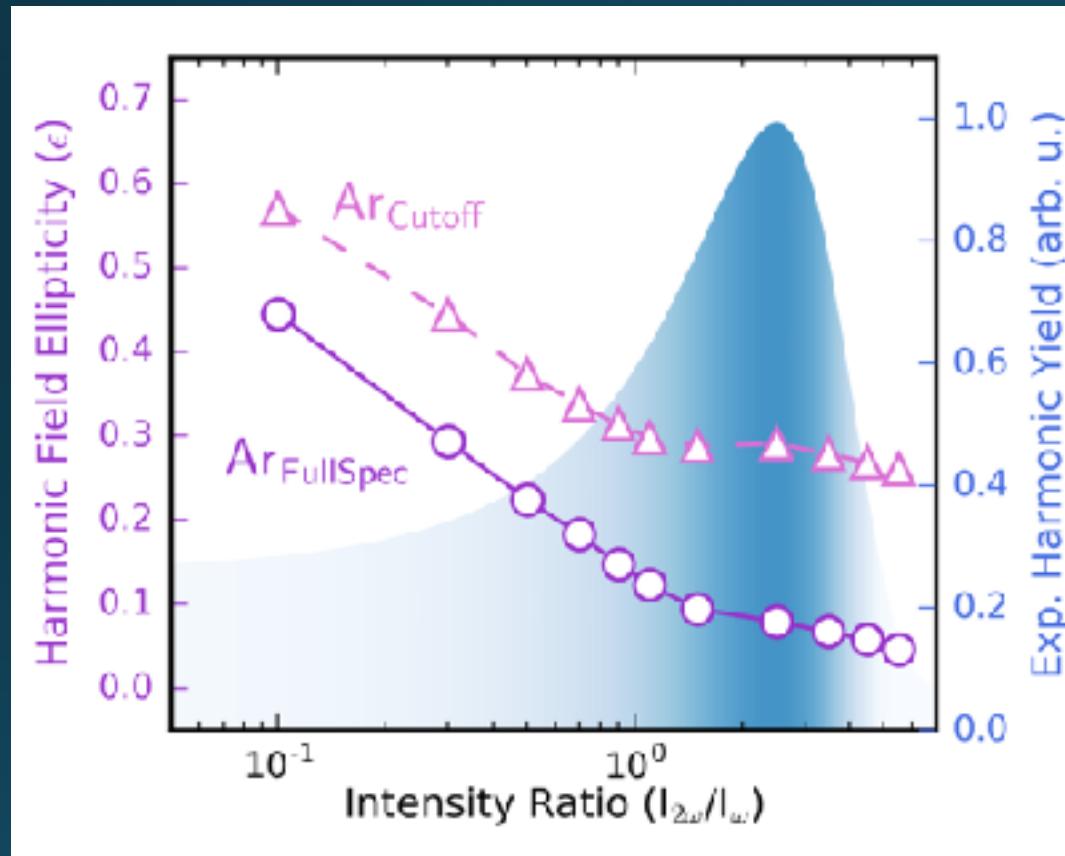
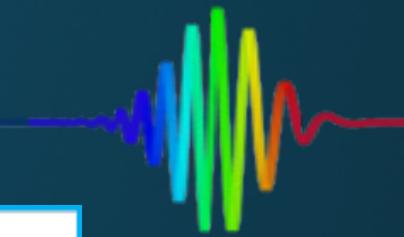


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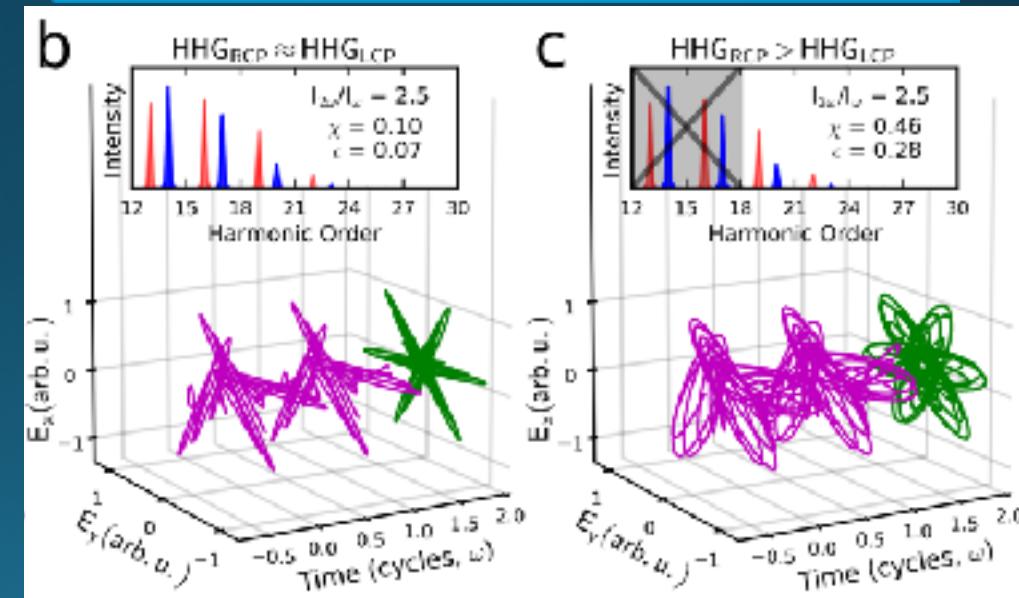
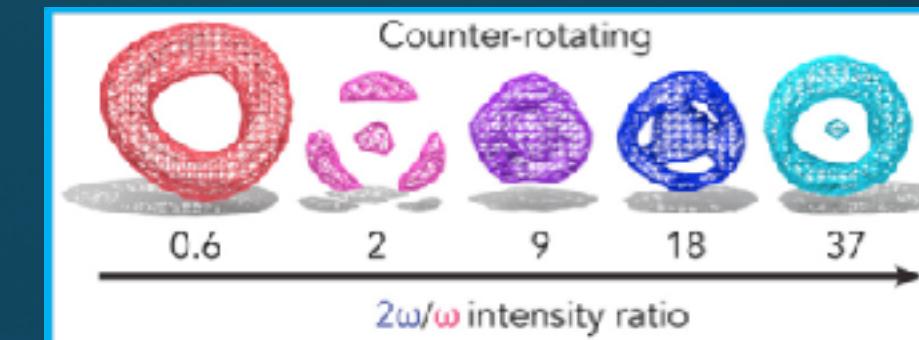
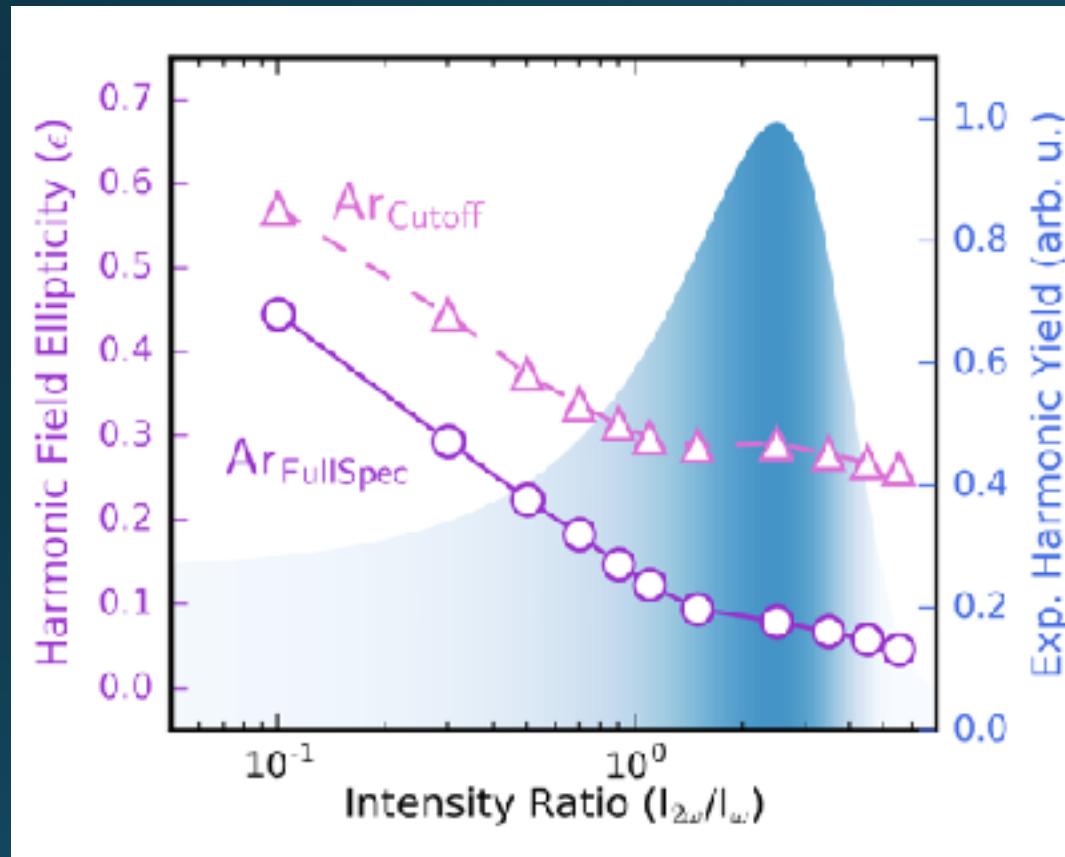
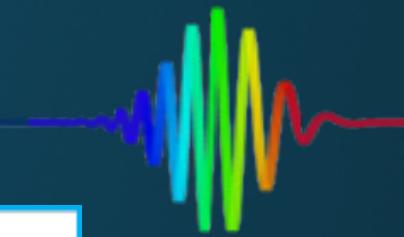
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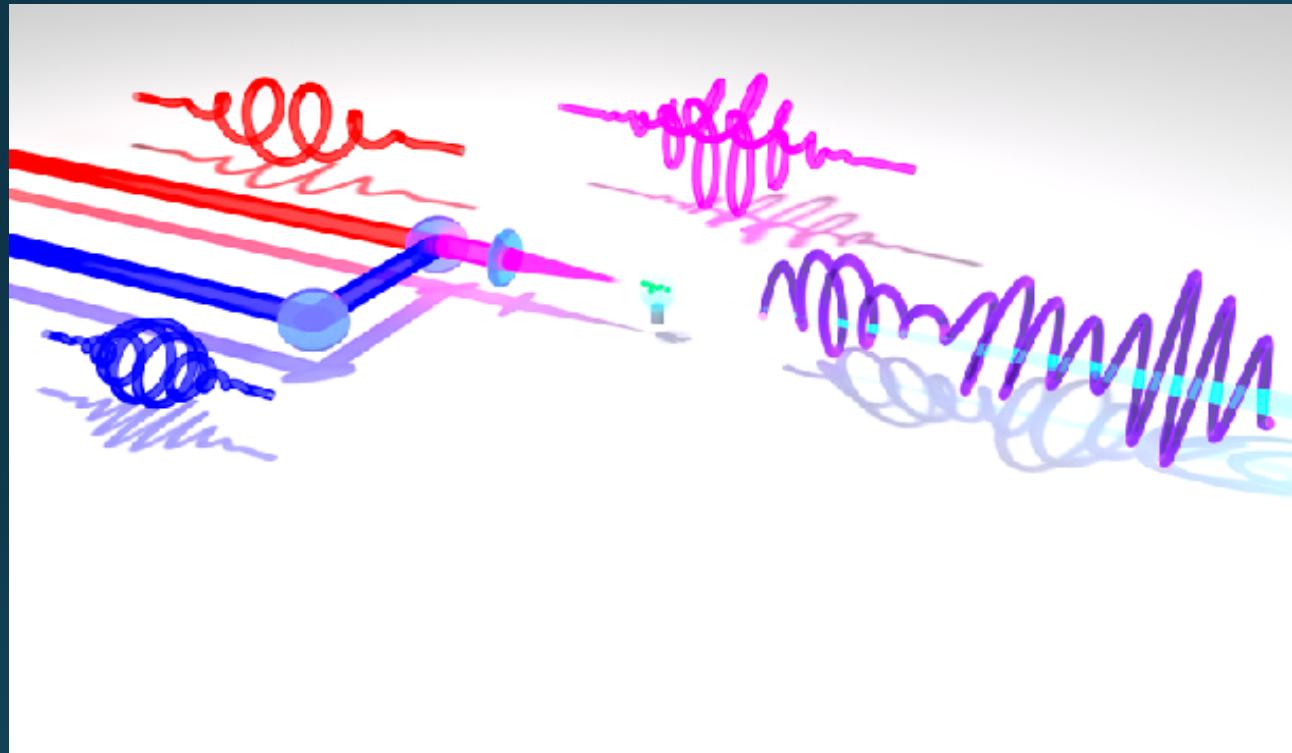
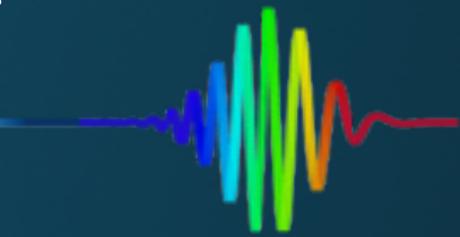
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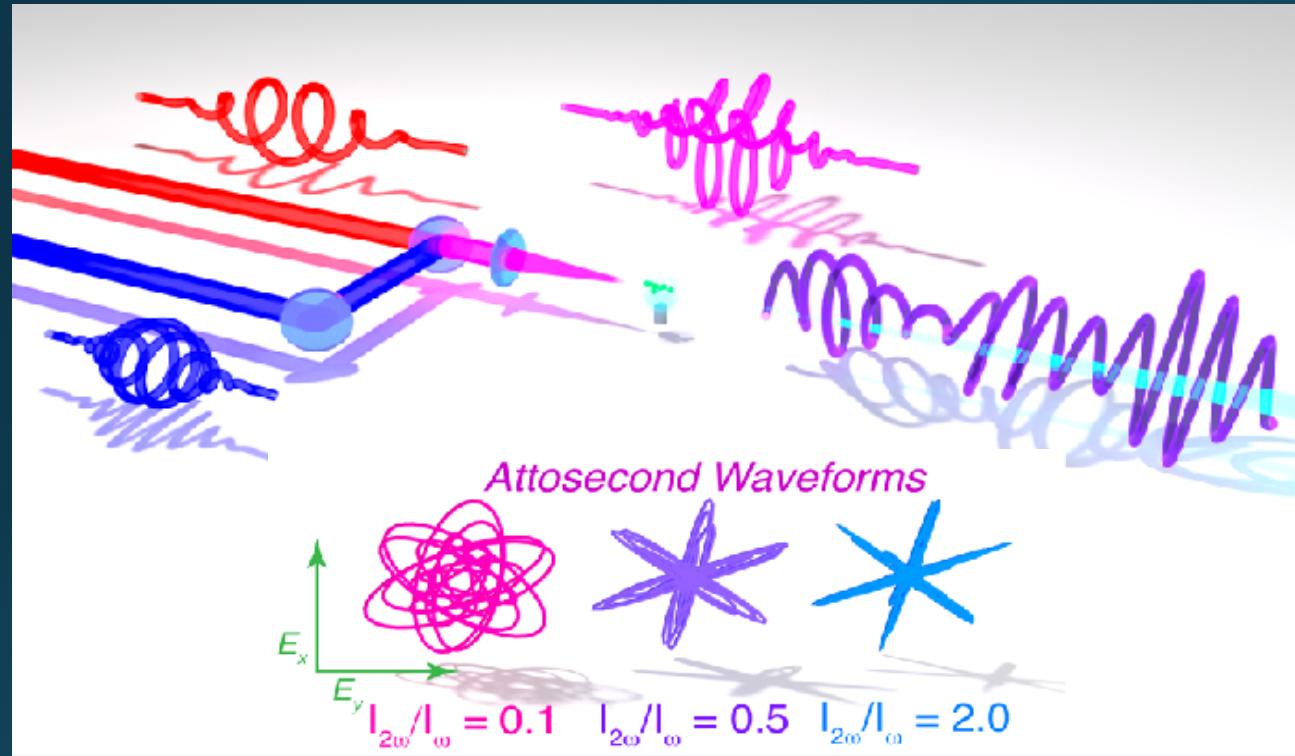
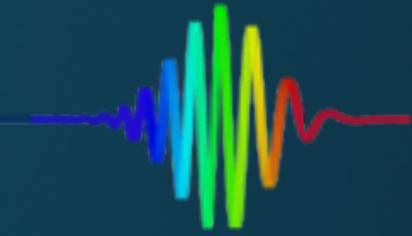


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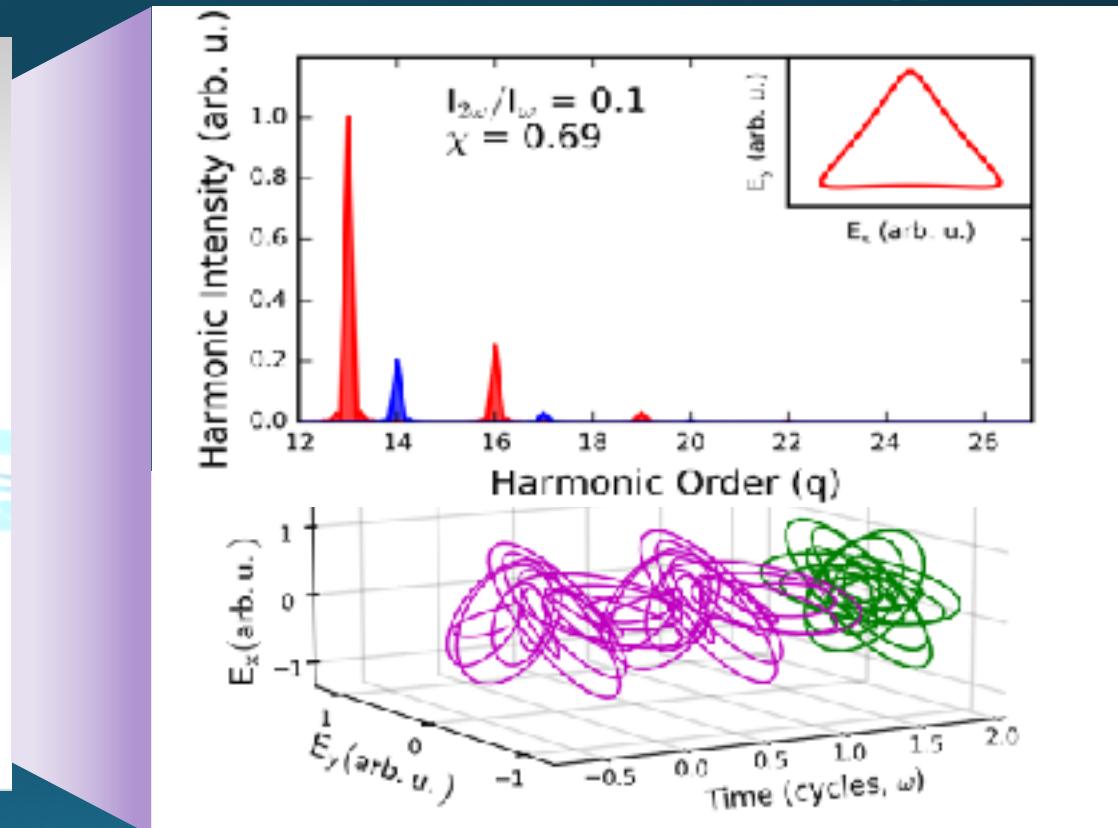
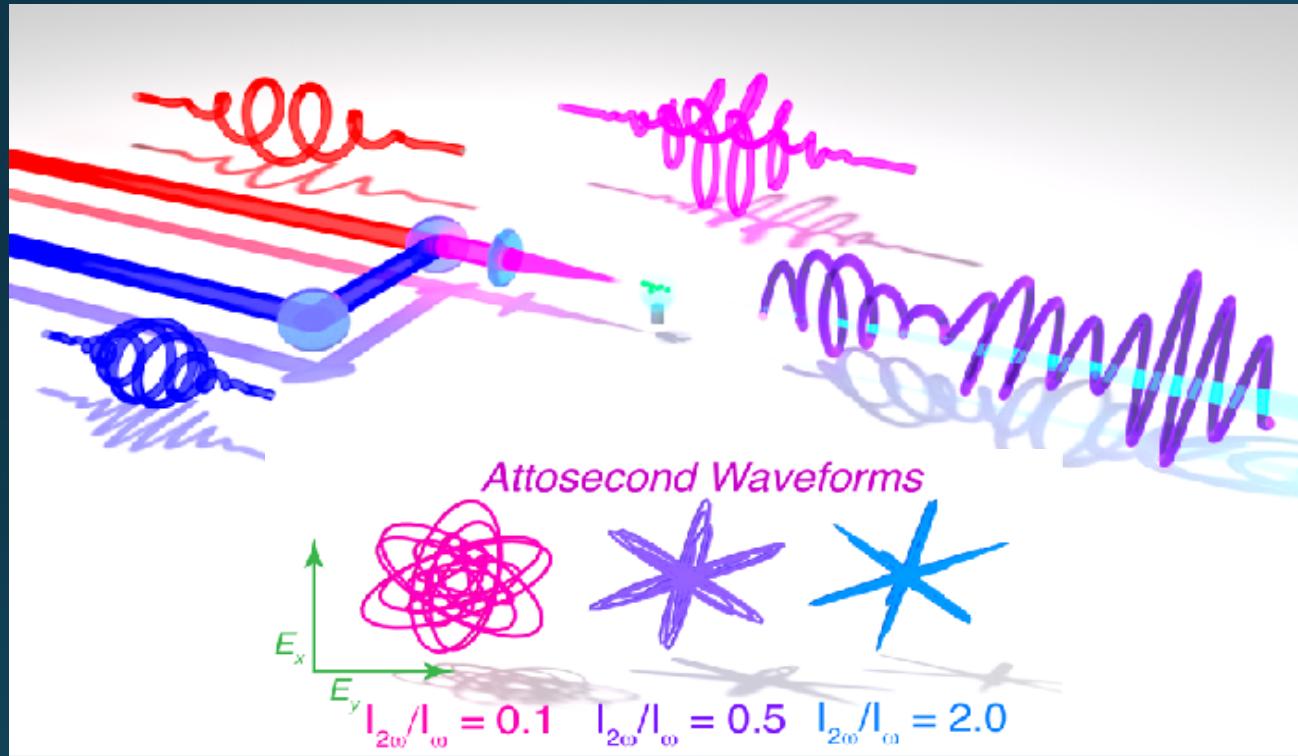
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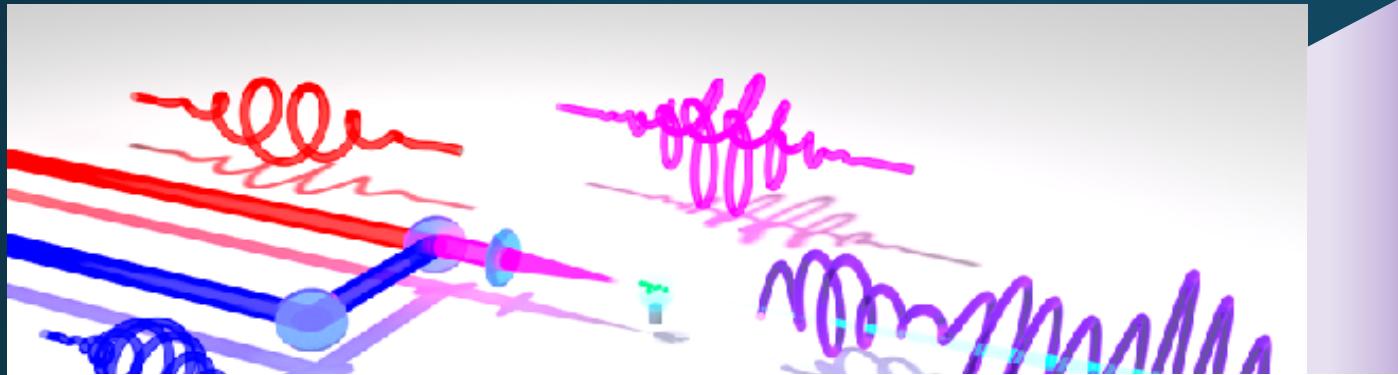
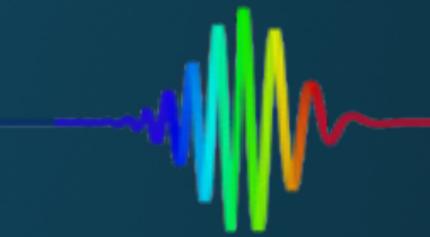


Dorney et. al., Phys. Rev. Lett., 118, 2017



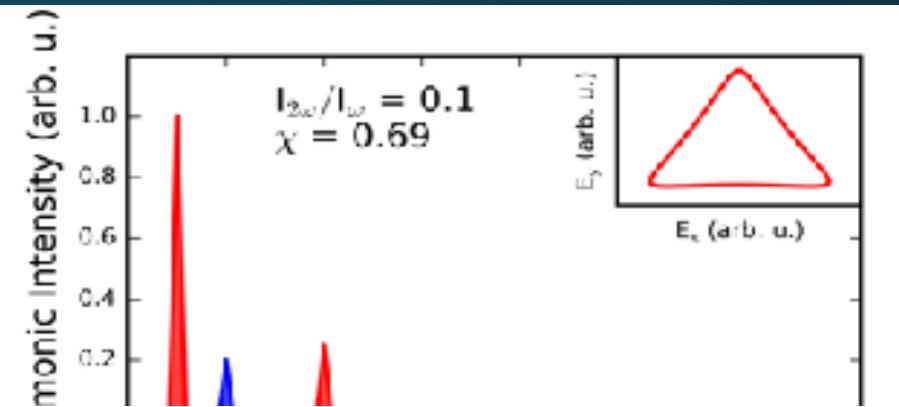
Production of Elliptically Polarized Attosecond Waveforms: Custom Attosecond Pulses for Chiral Spectroscopies





PRL 119, 063201 (2017)

PHYSICAL REVIEW LETTERS

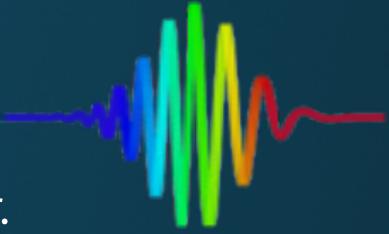
week ending
11 AUGUST 2017

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

Kevin M. Domey,^{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)!

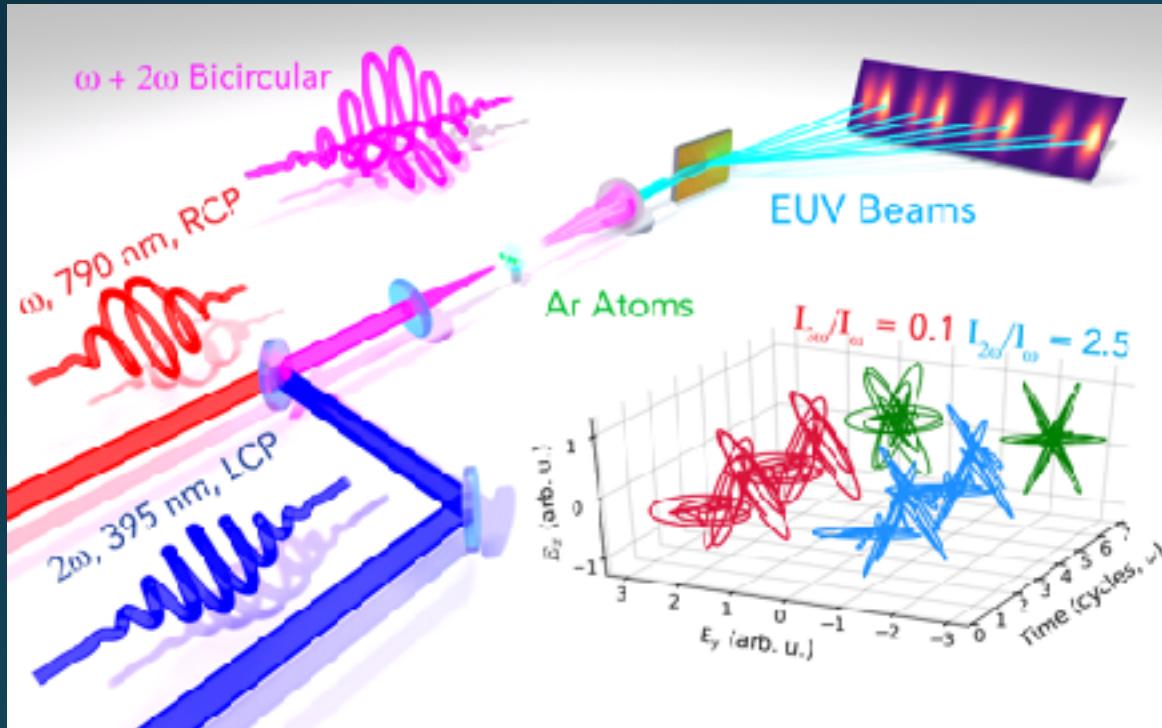


➤ CPHHG vs BHHS; two sides of the same coin, yet often far removed from each other.

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CPHHG: EUV Light Pulses for Spectroscopy



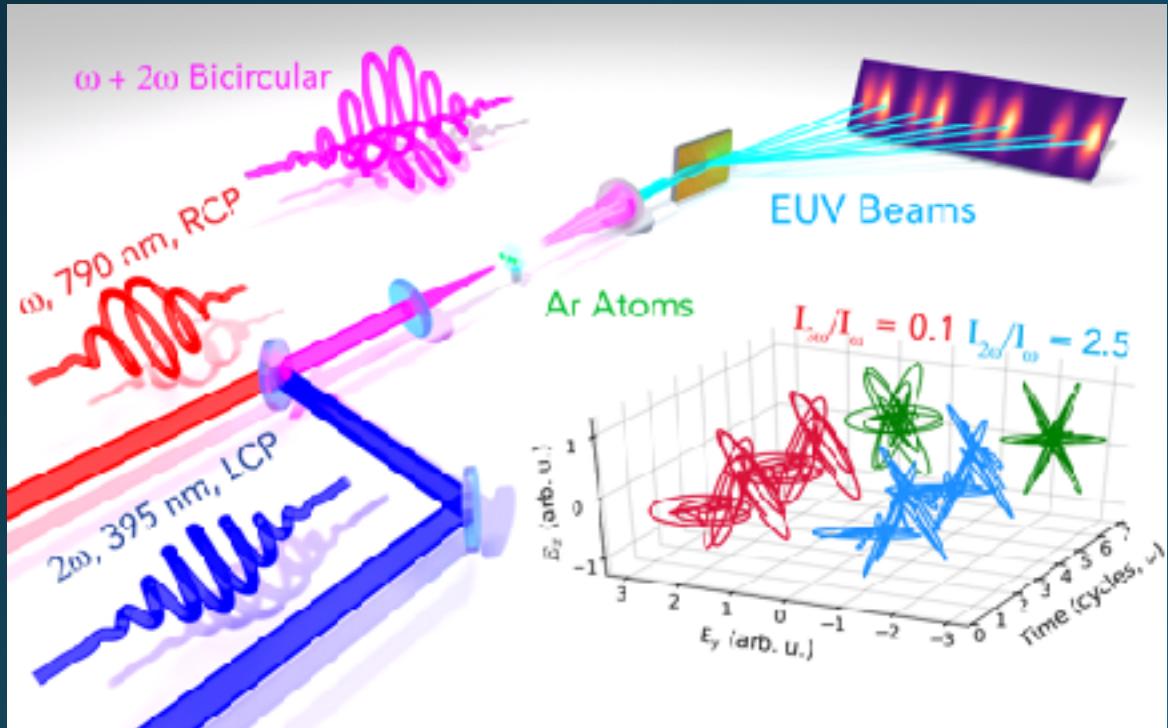
Baykusheva, PRL, 116, 2016

Dorney, PRL 119, 2017

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

CPHHG: EUV Light Pulses for Spectroscopy



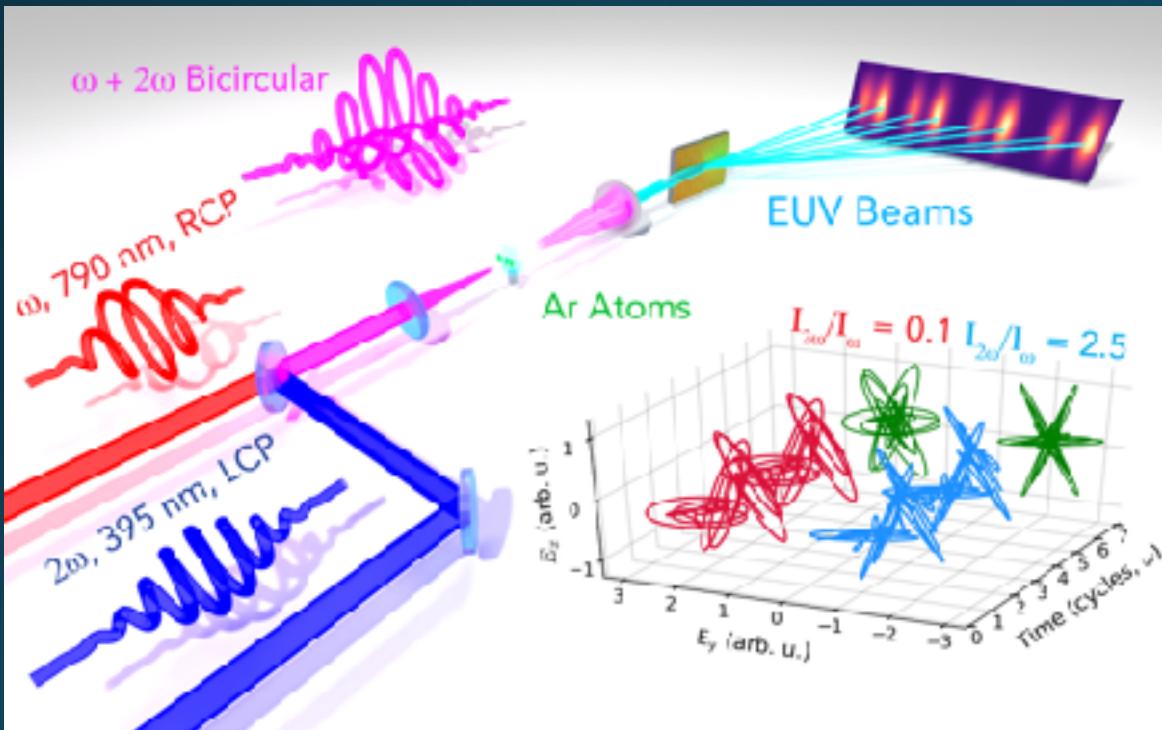
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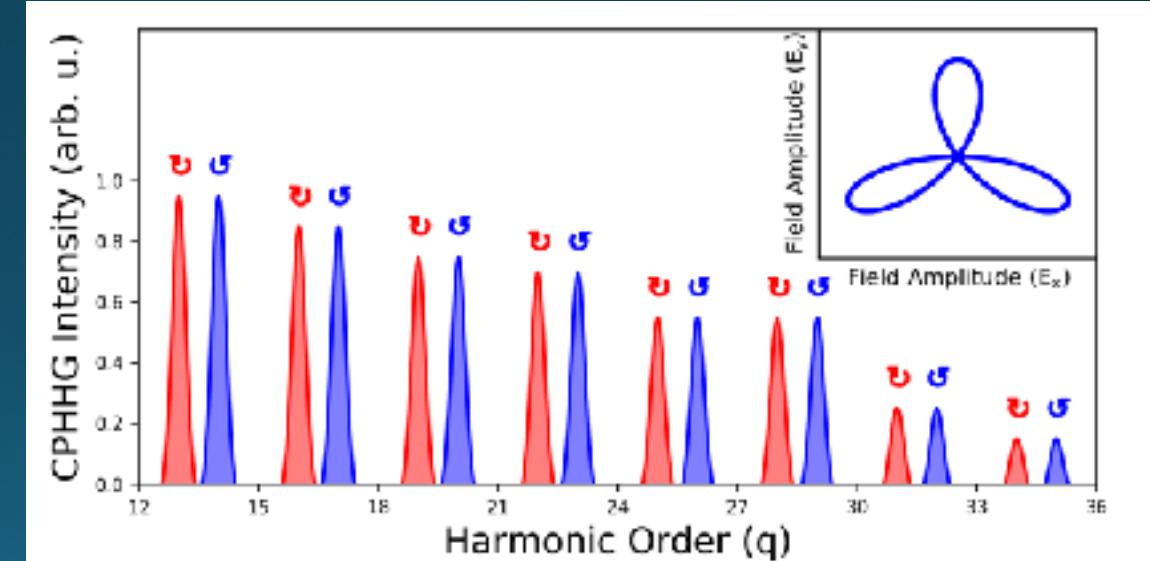


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Baykusheva, PRL, 116, 2016

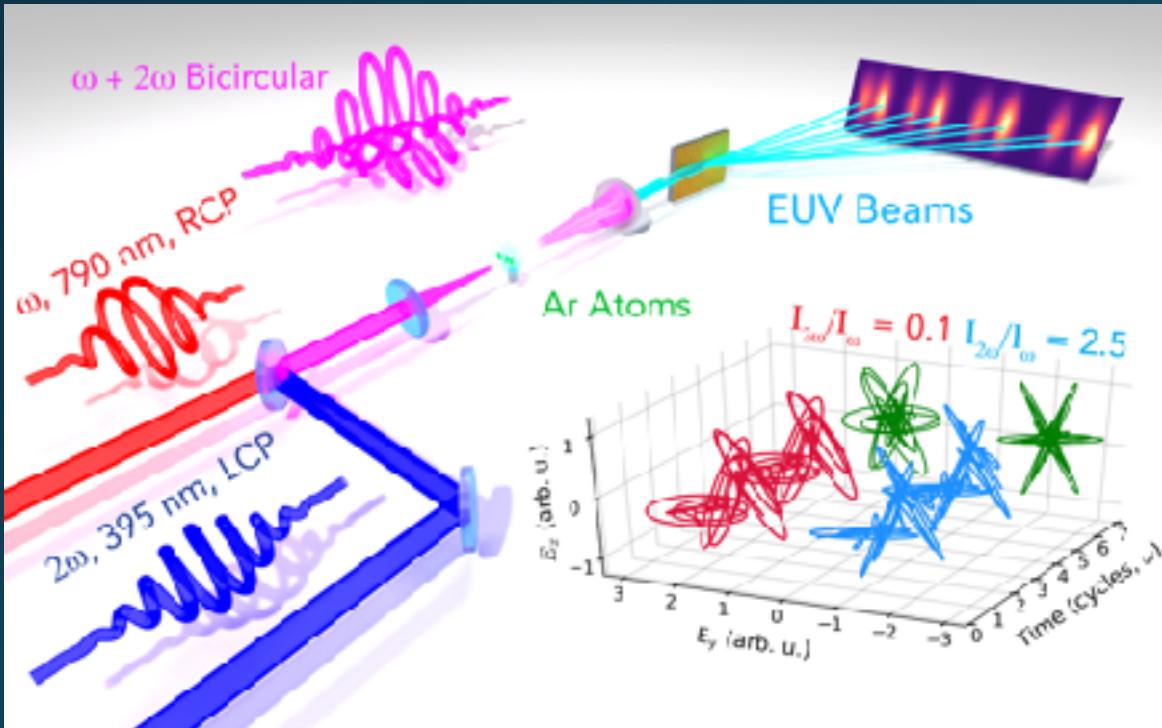
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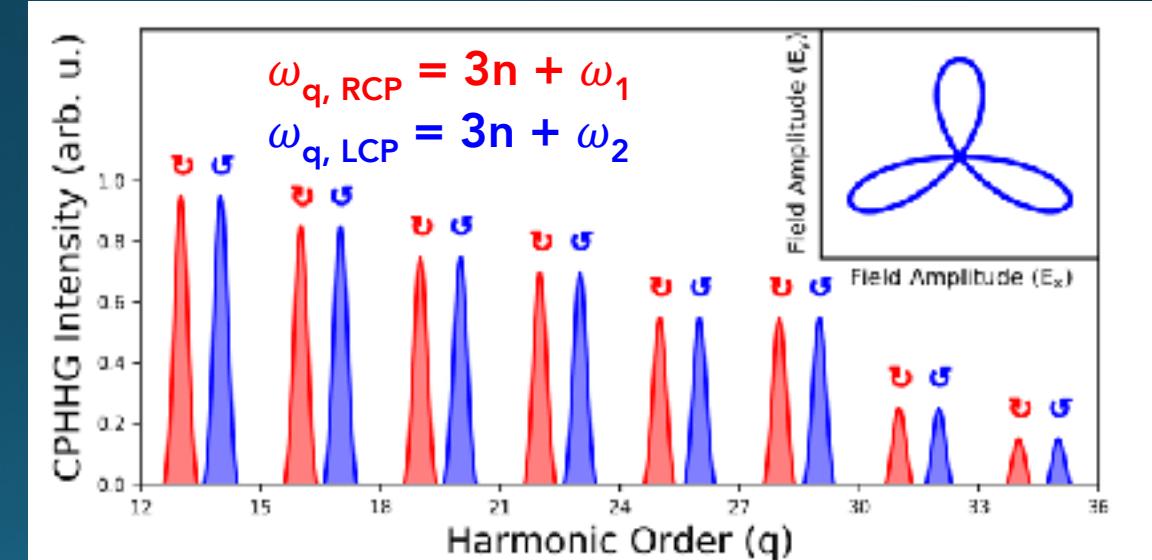


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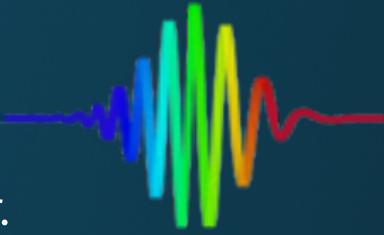
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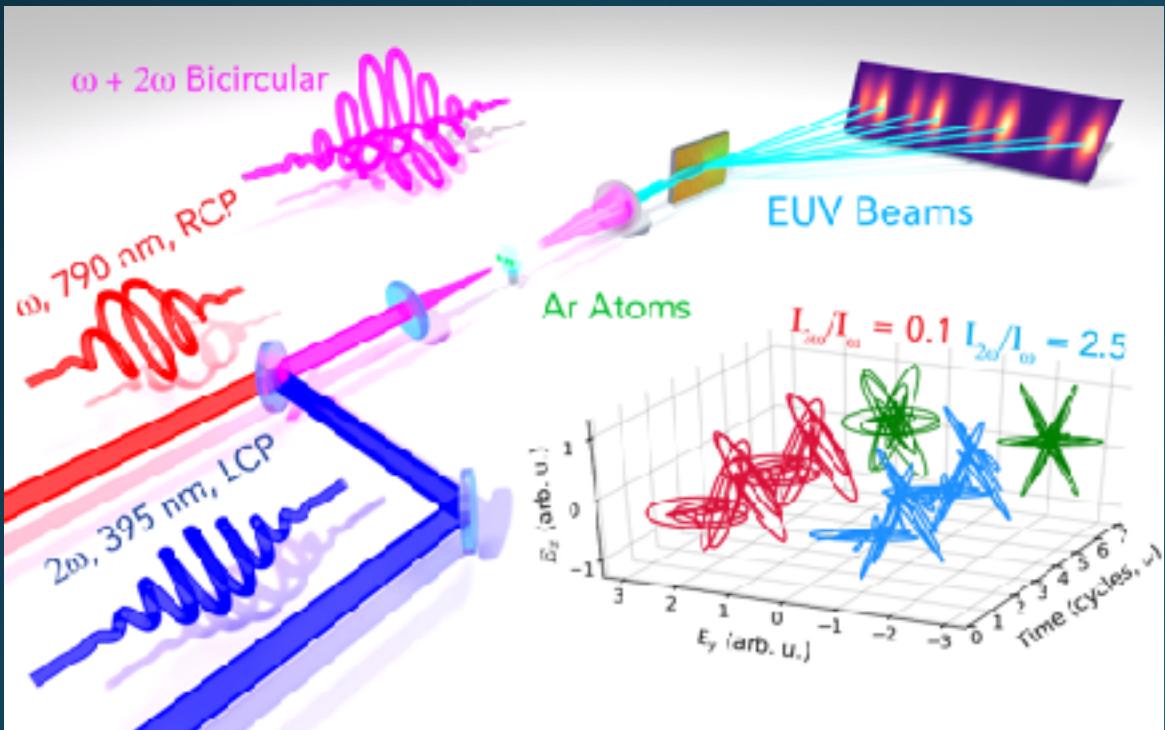


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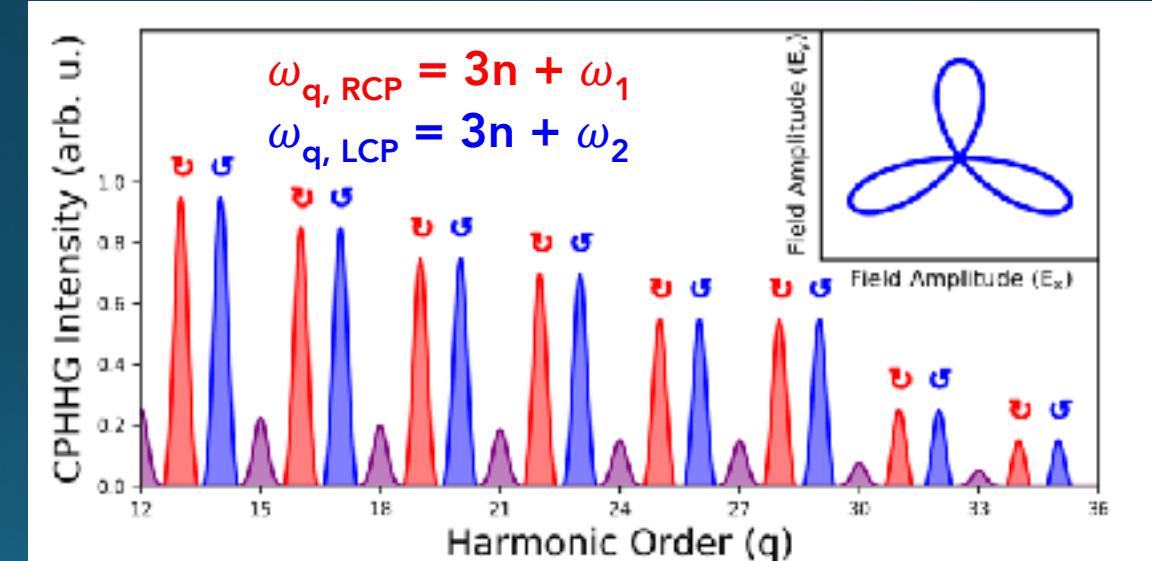


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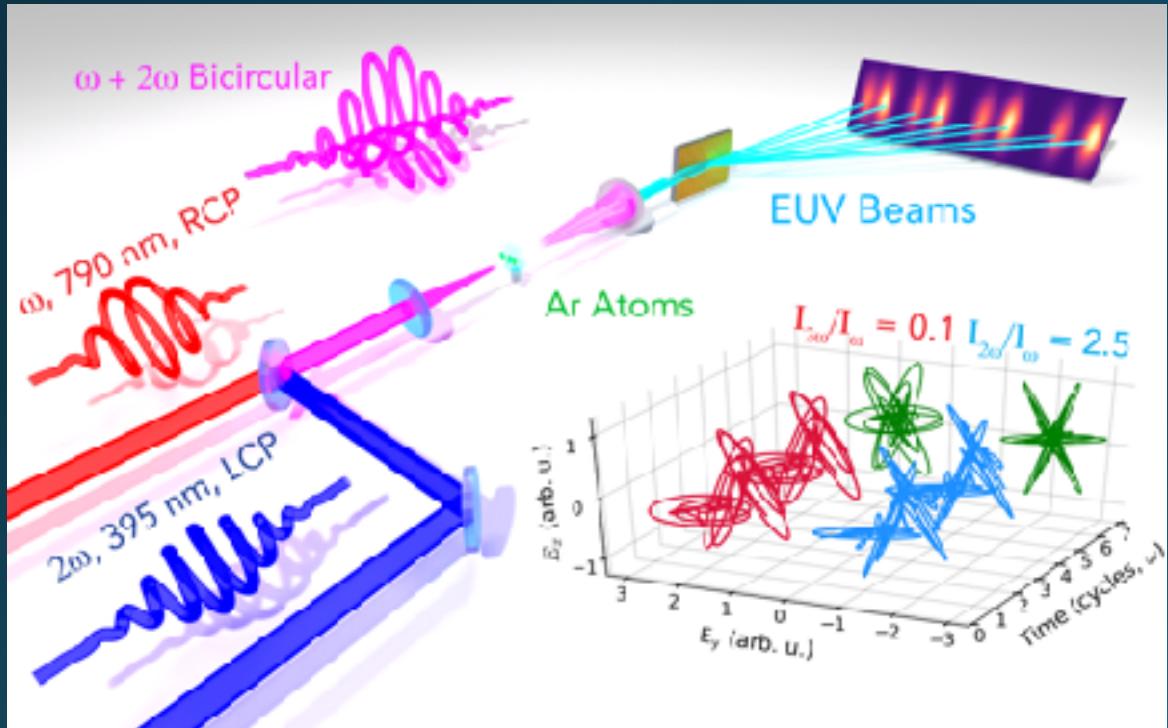
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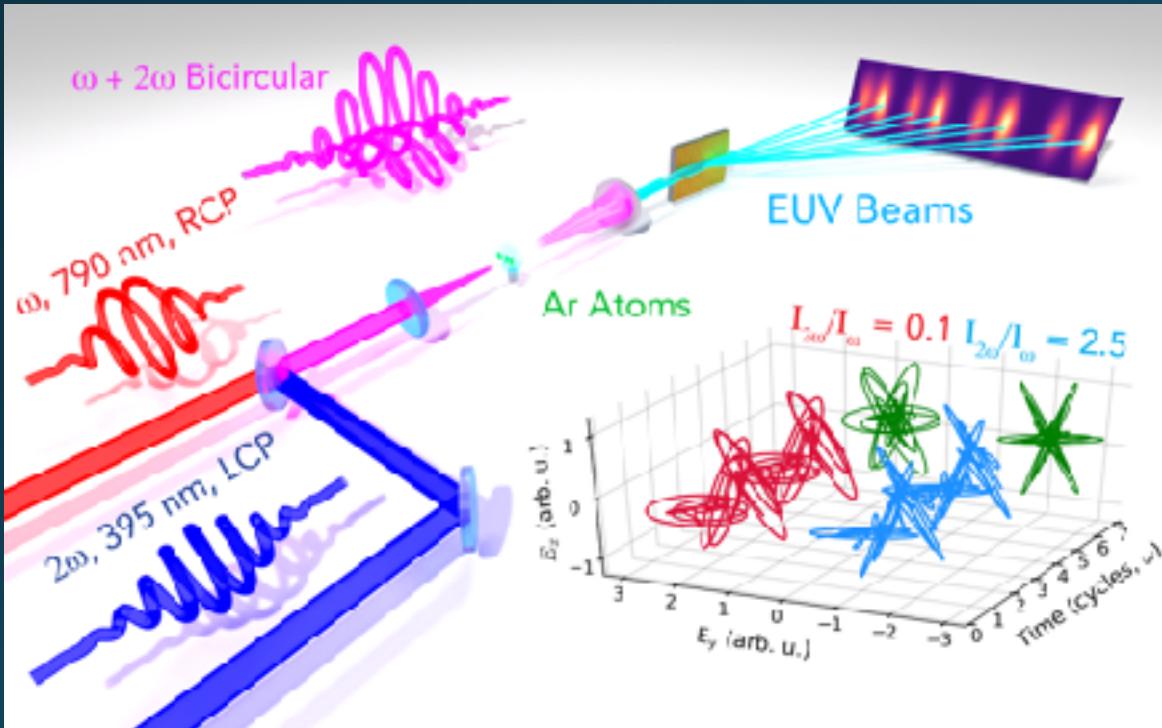
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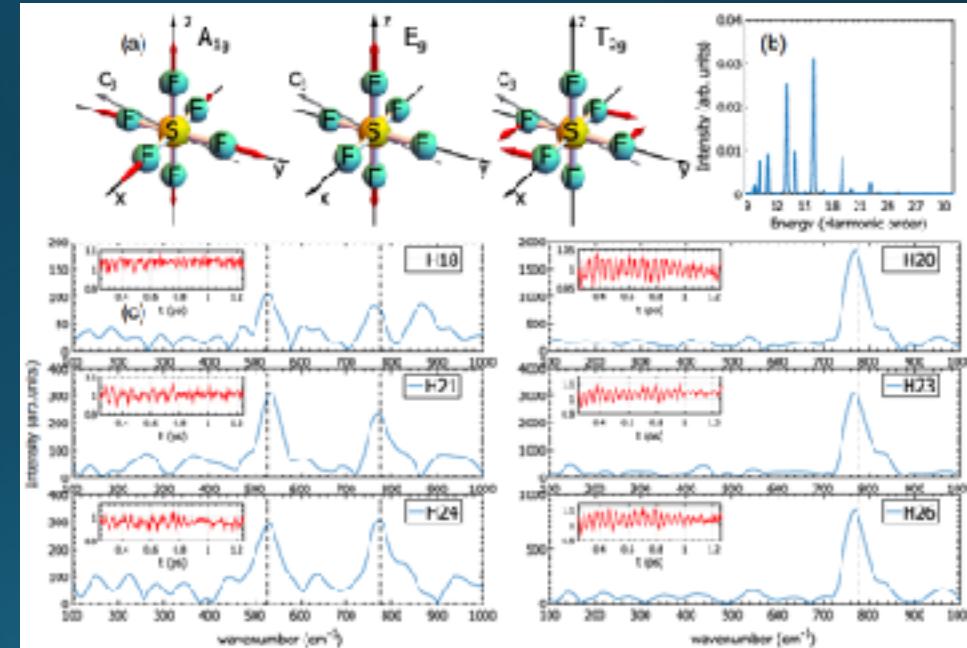


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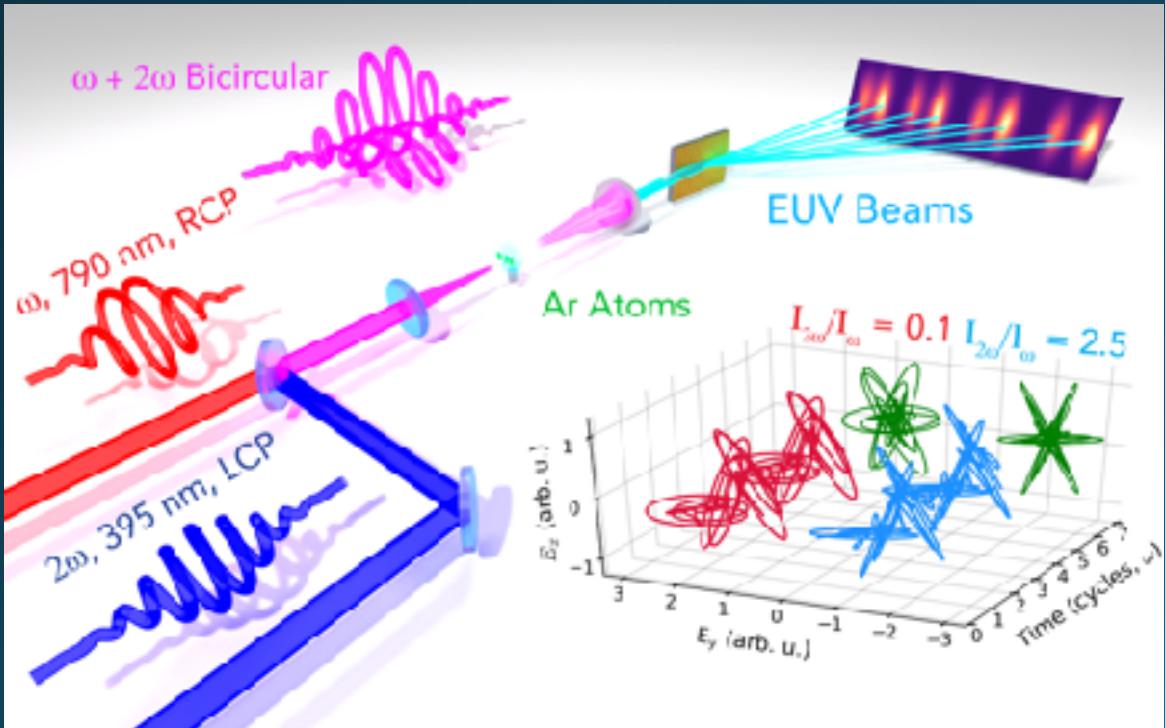


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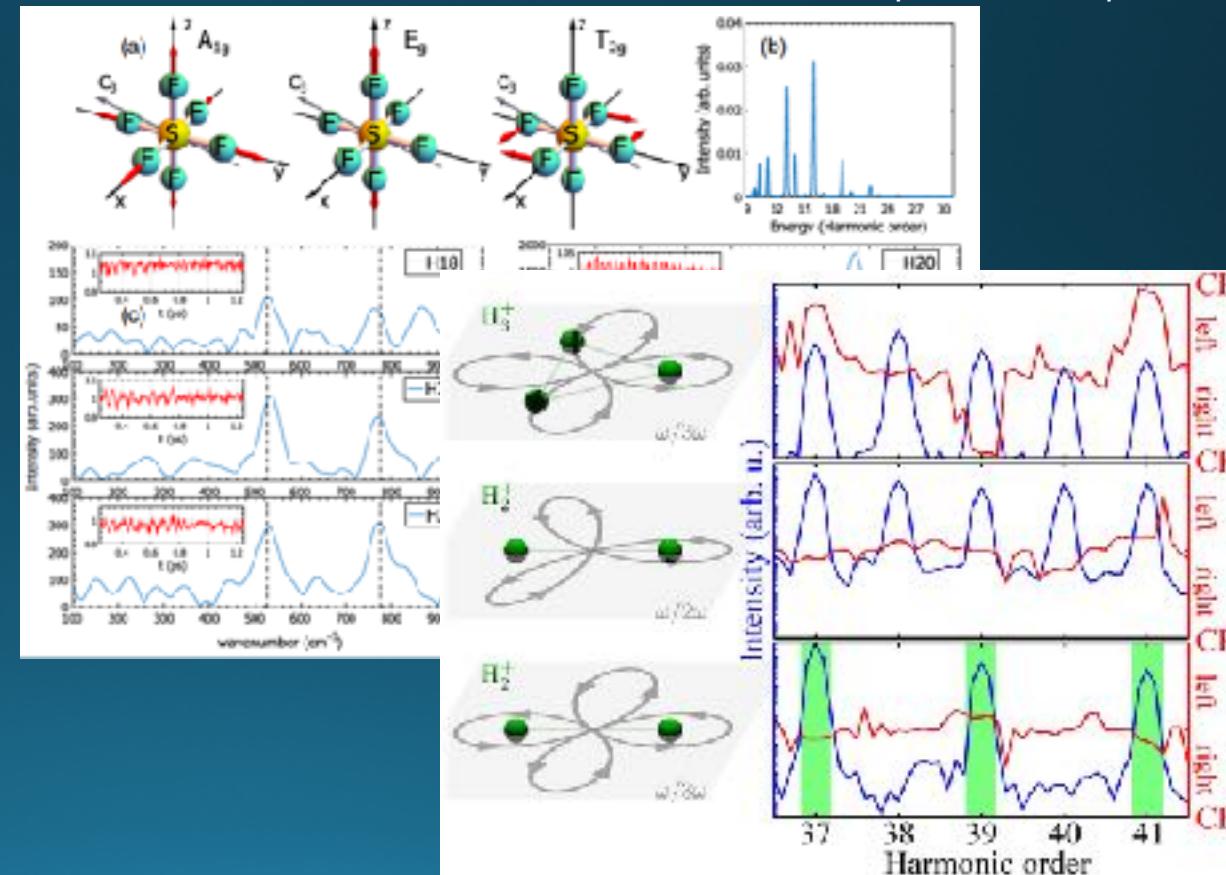
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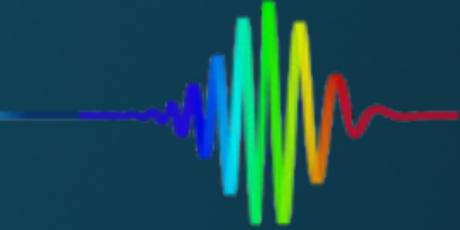
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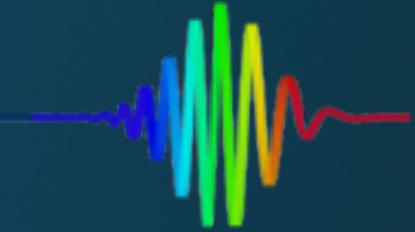
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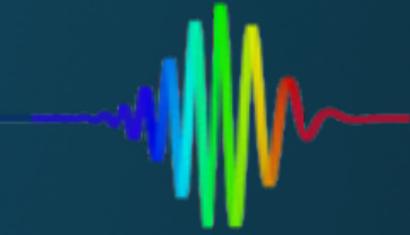


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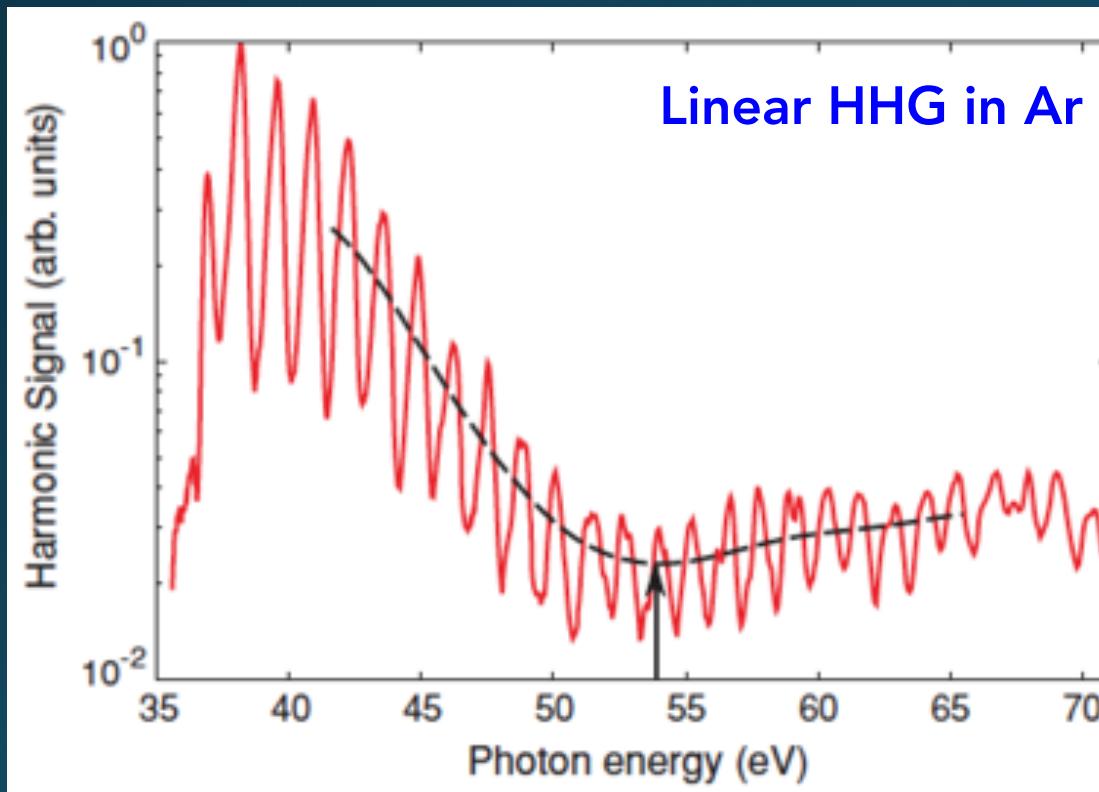


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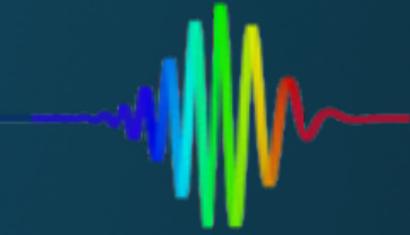
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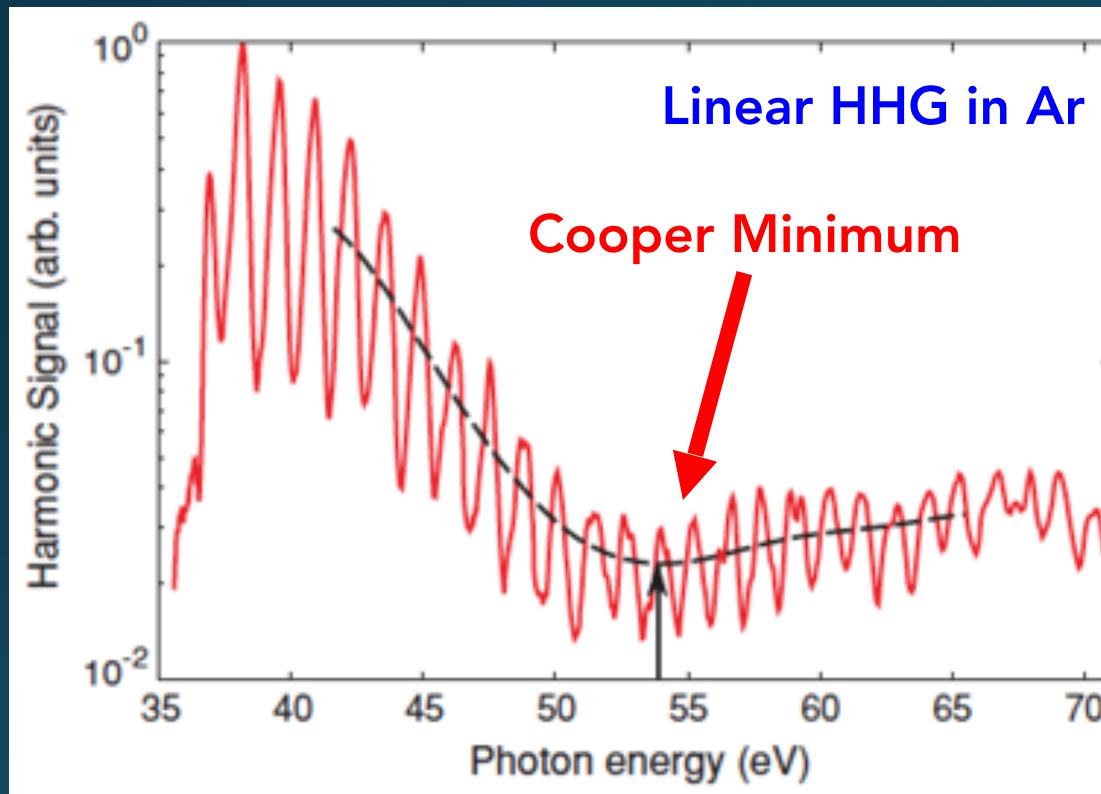
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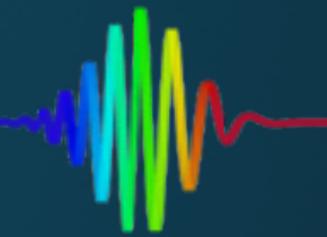
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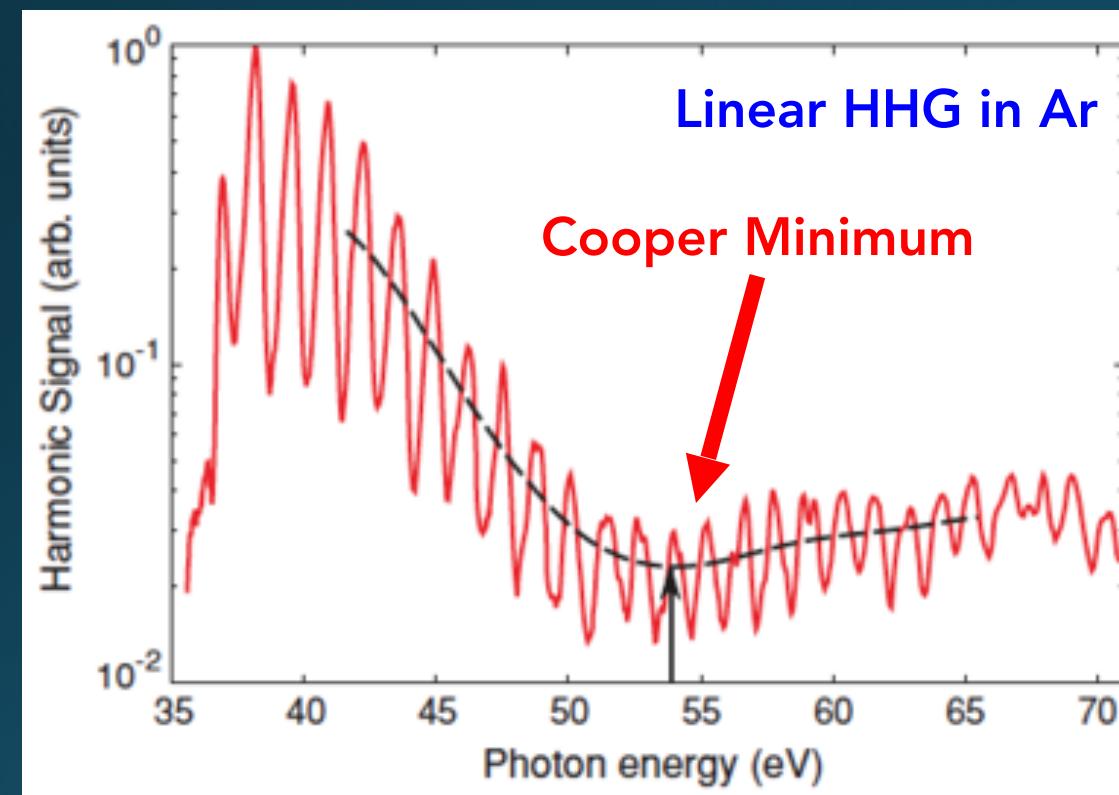


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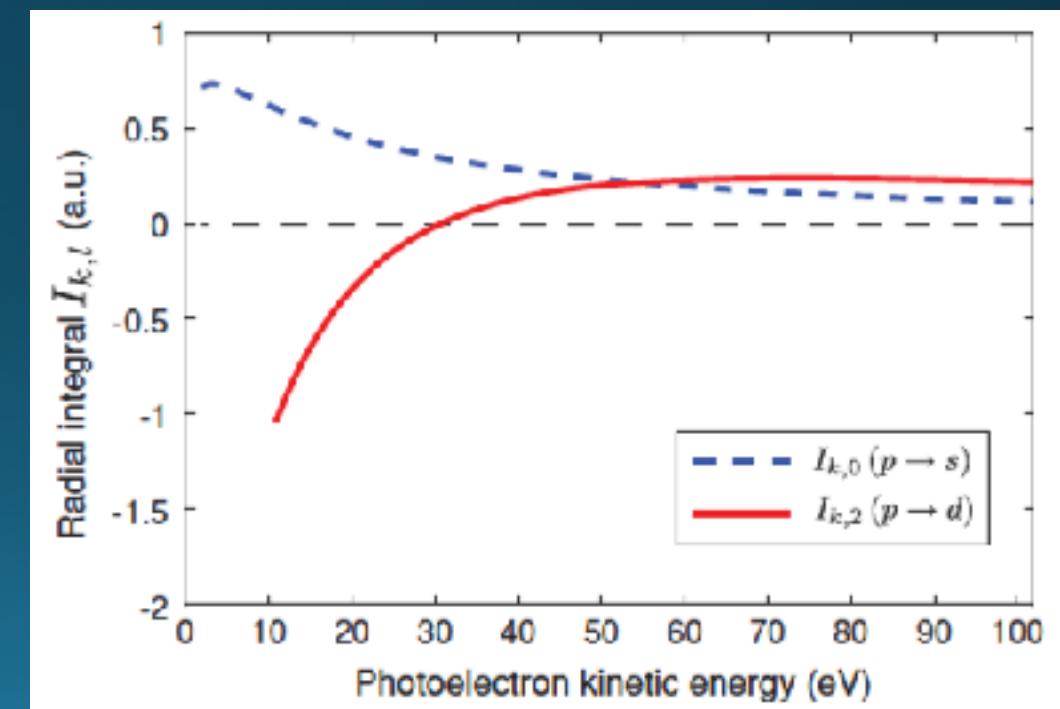
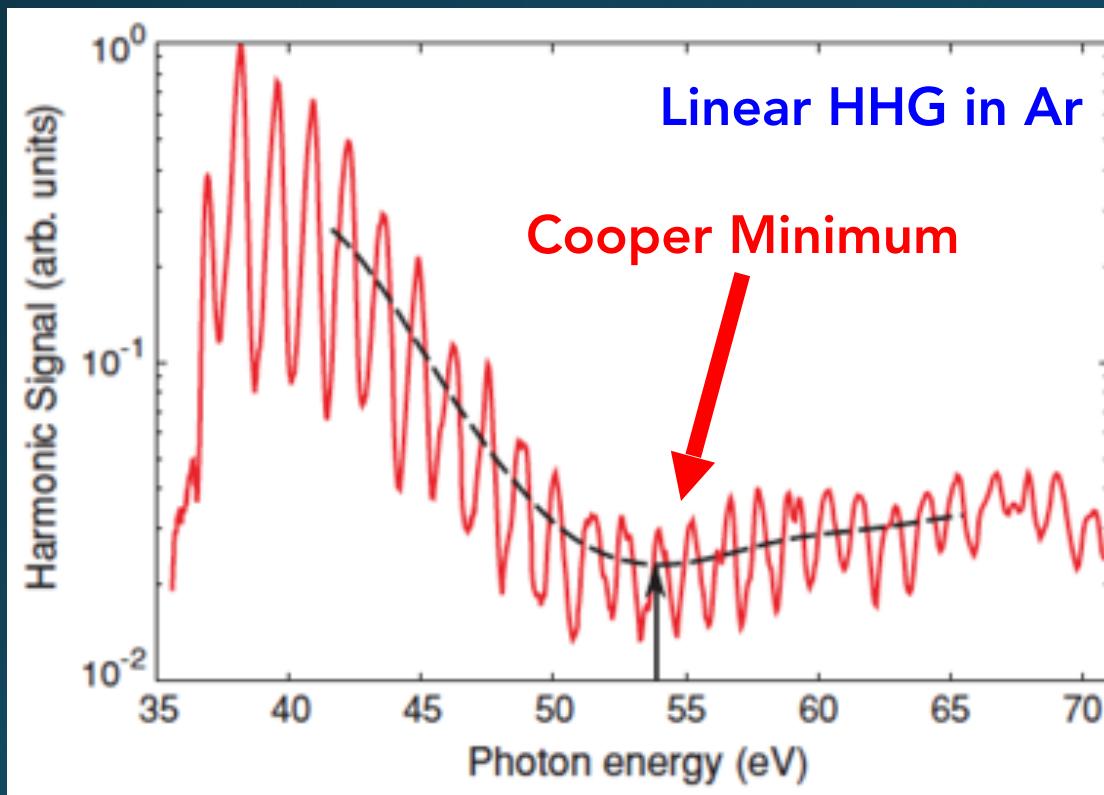


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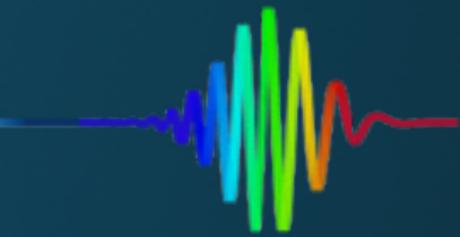
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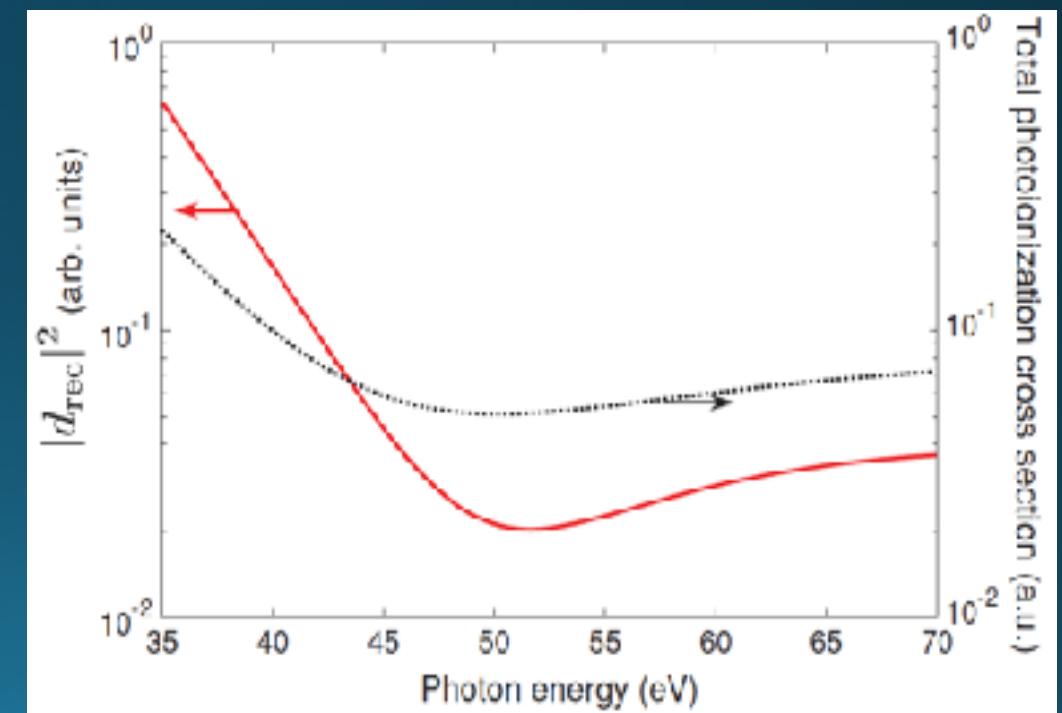
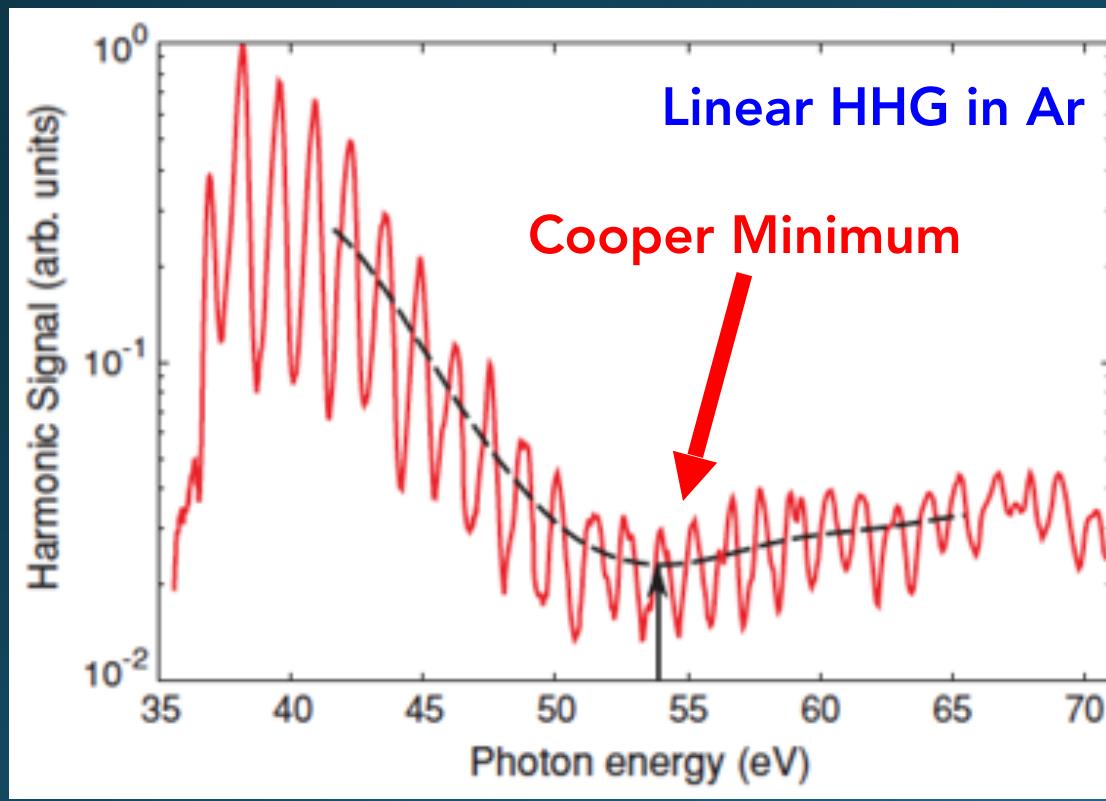


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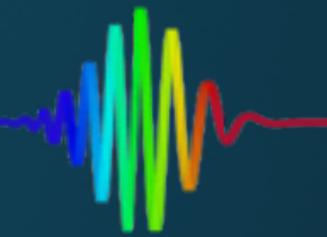


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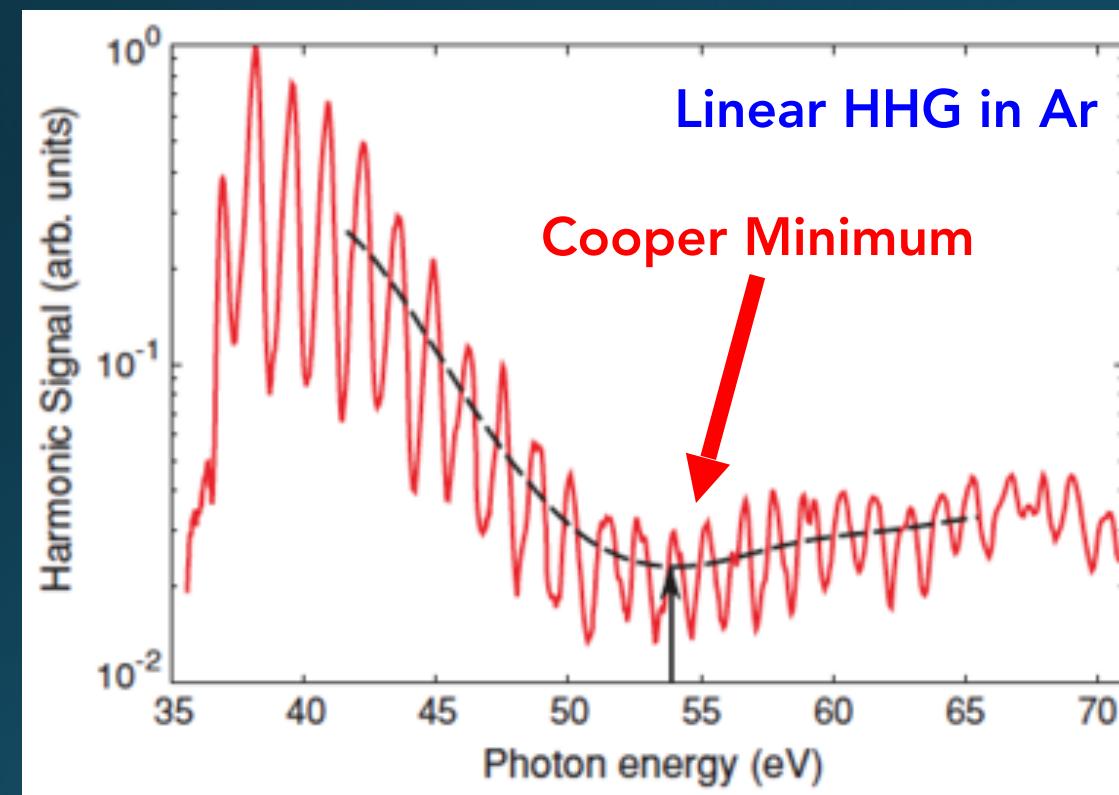


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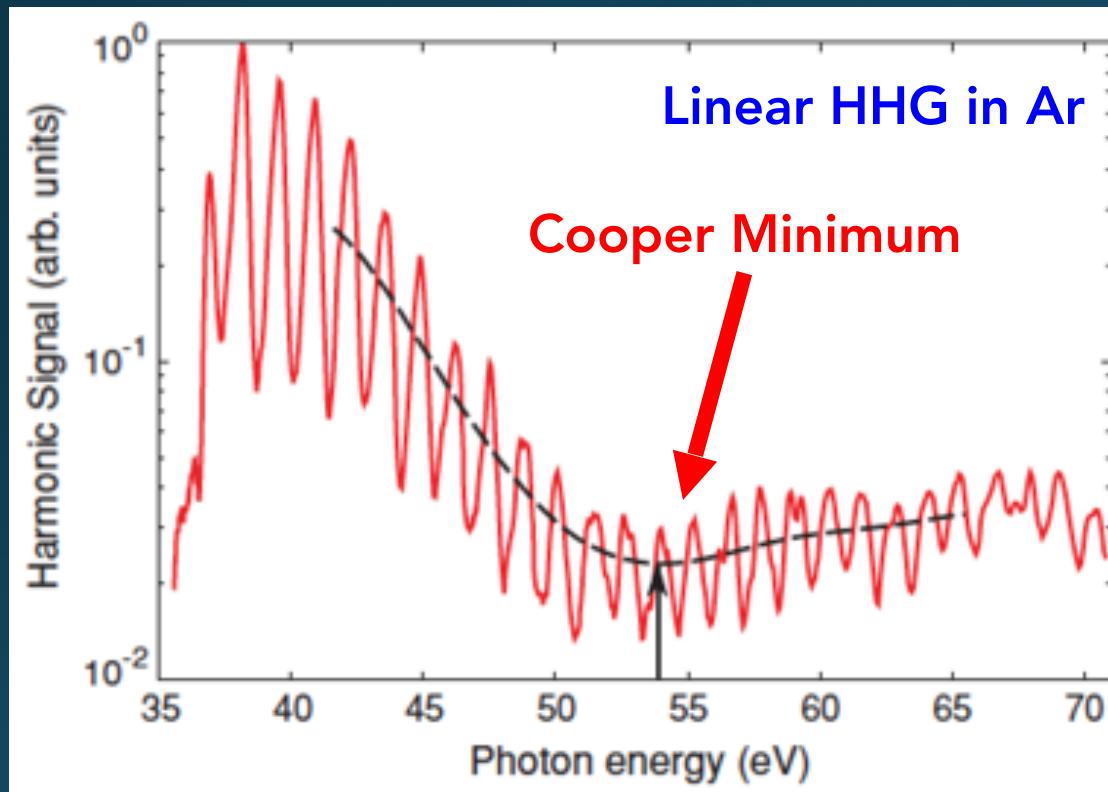


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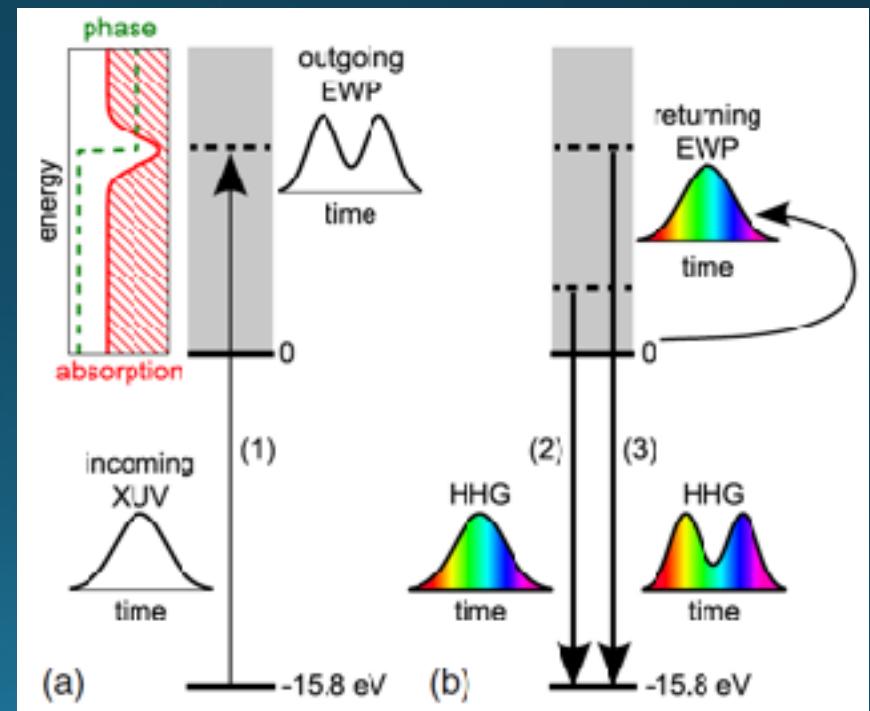
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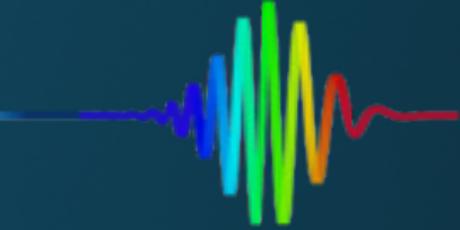
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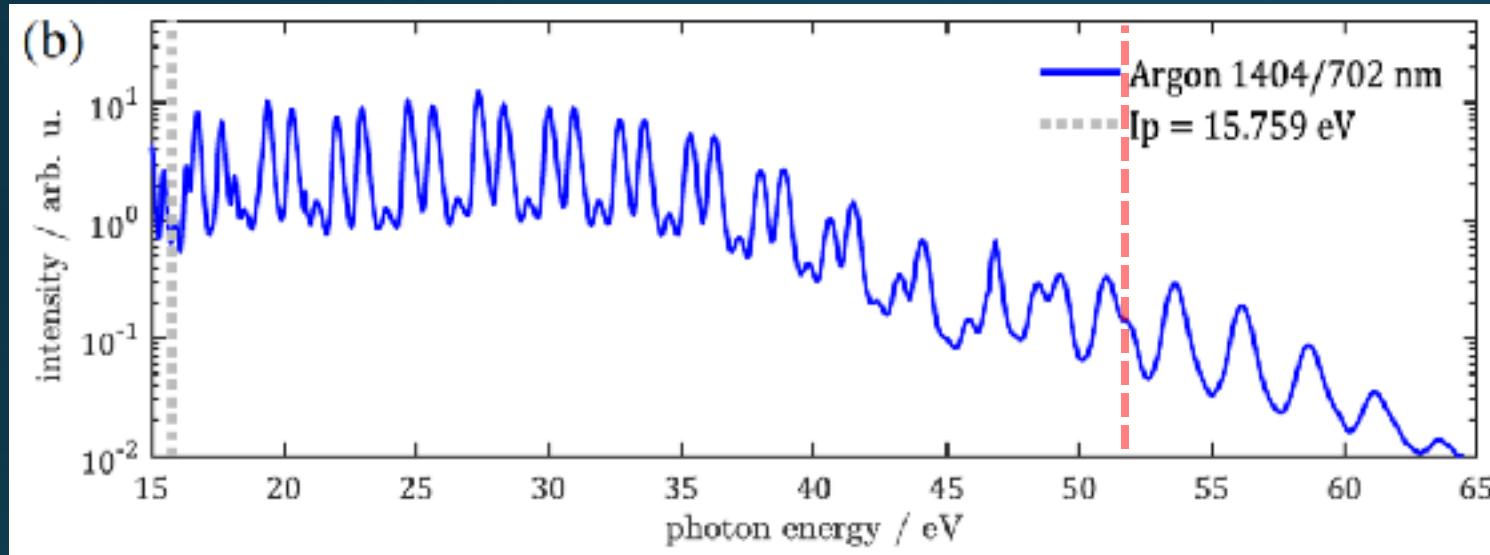
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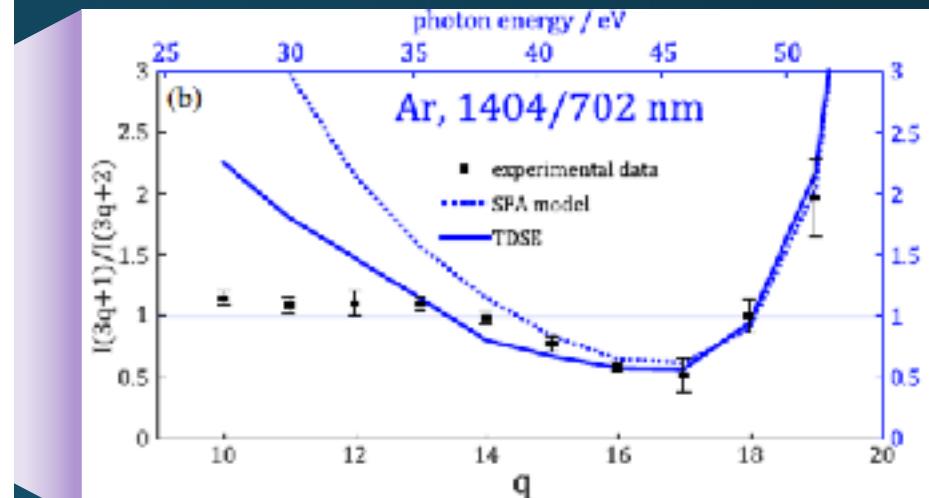
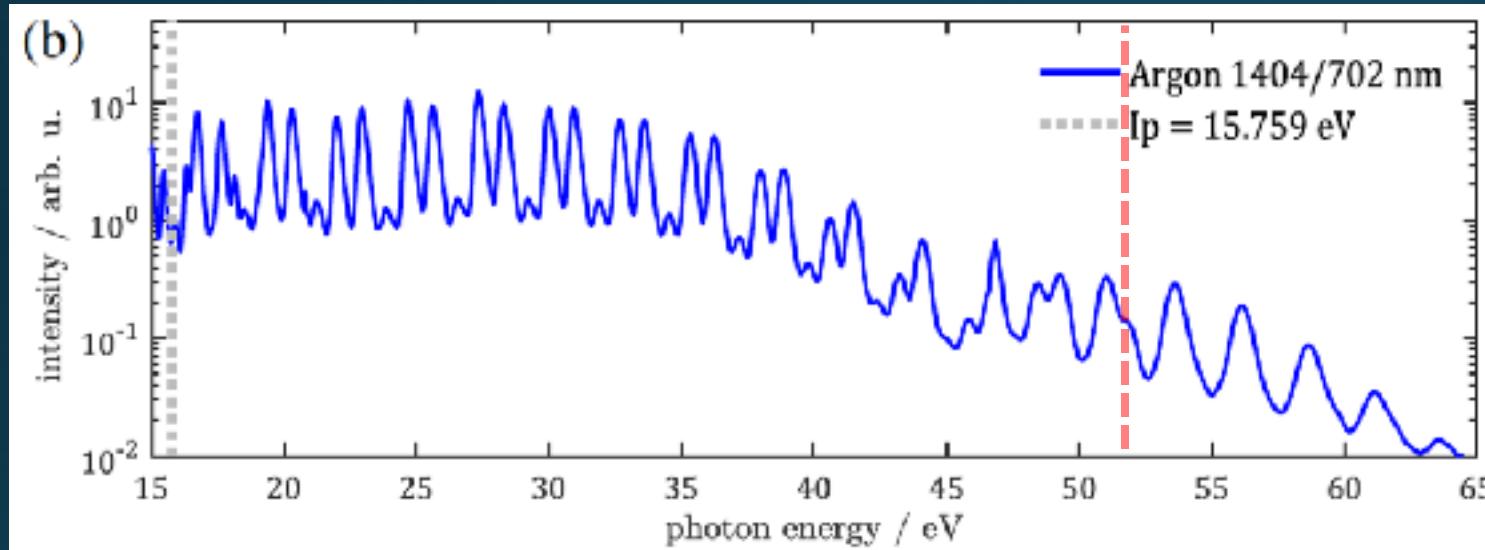


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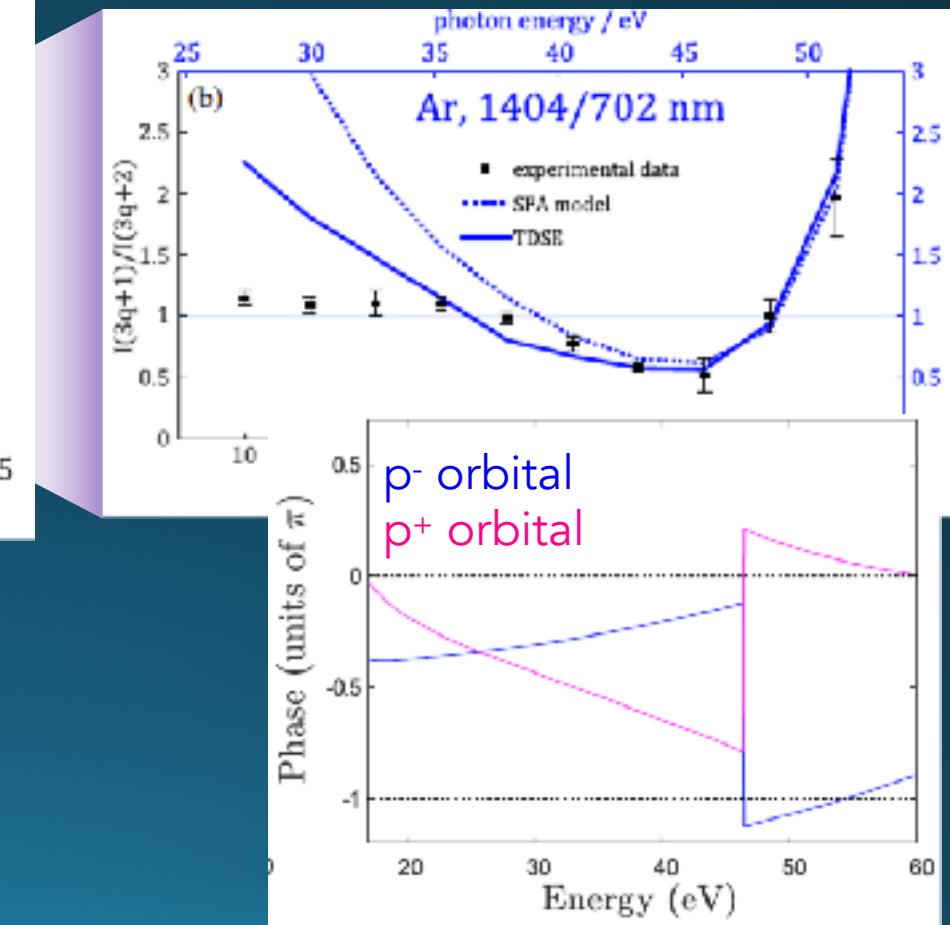
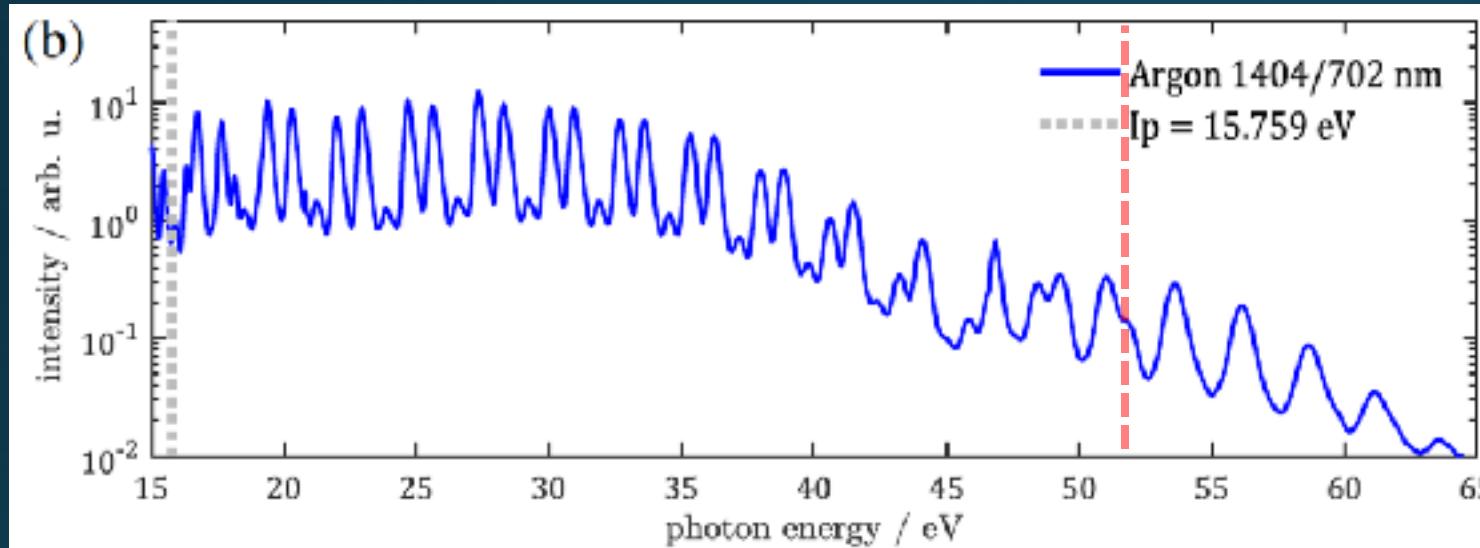
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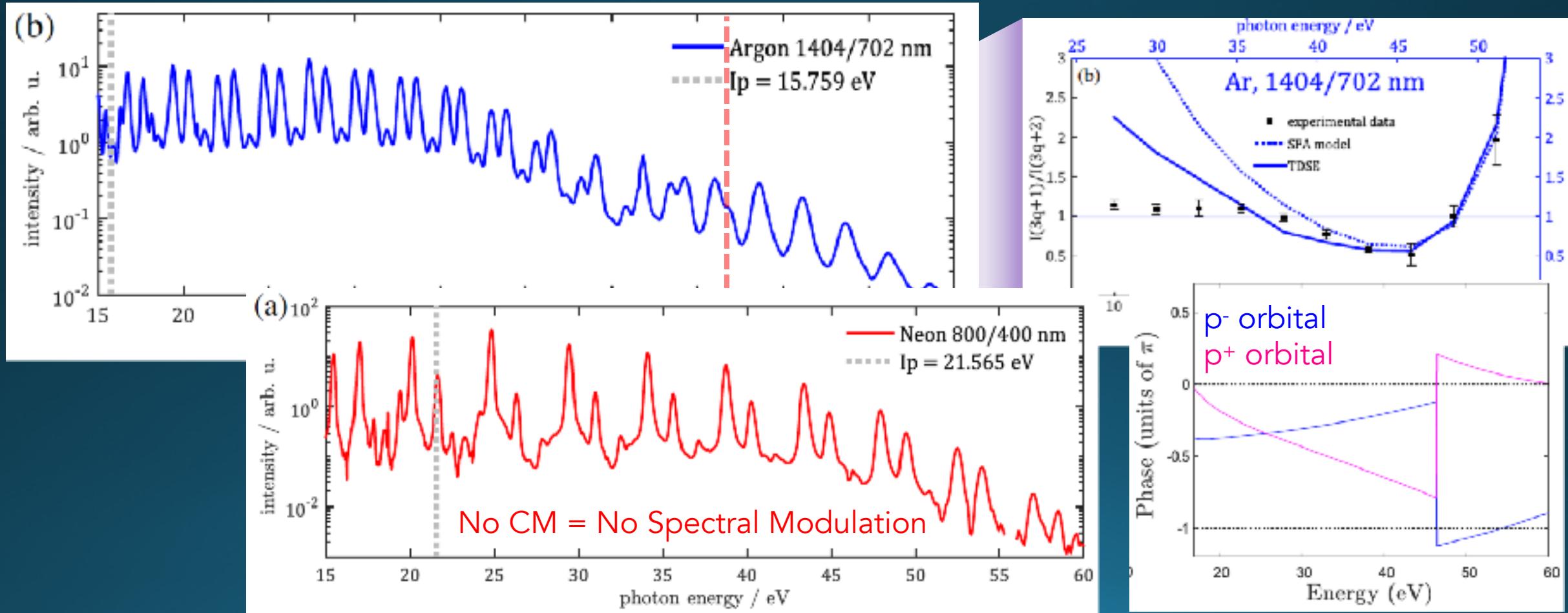
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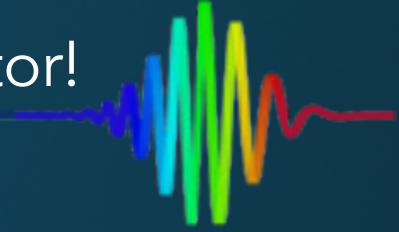
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Merging CPHHG and BHHS:
Exploiting Electronic Structure as an EUV Waveform Modulator!





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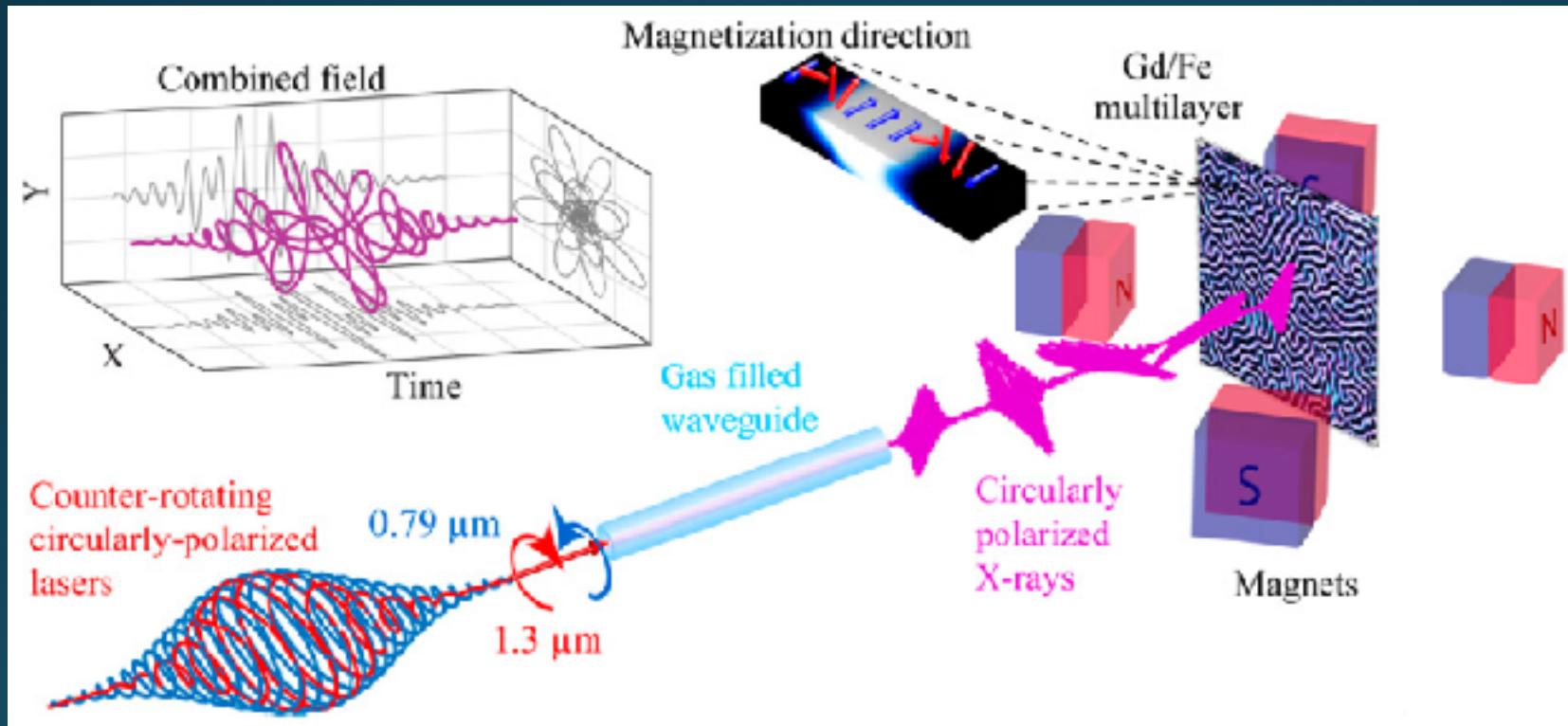


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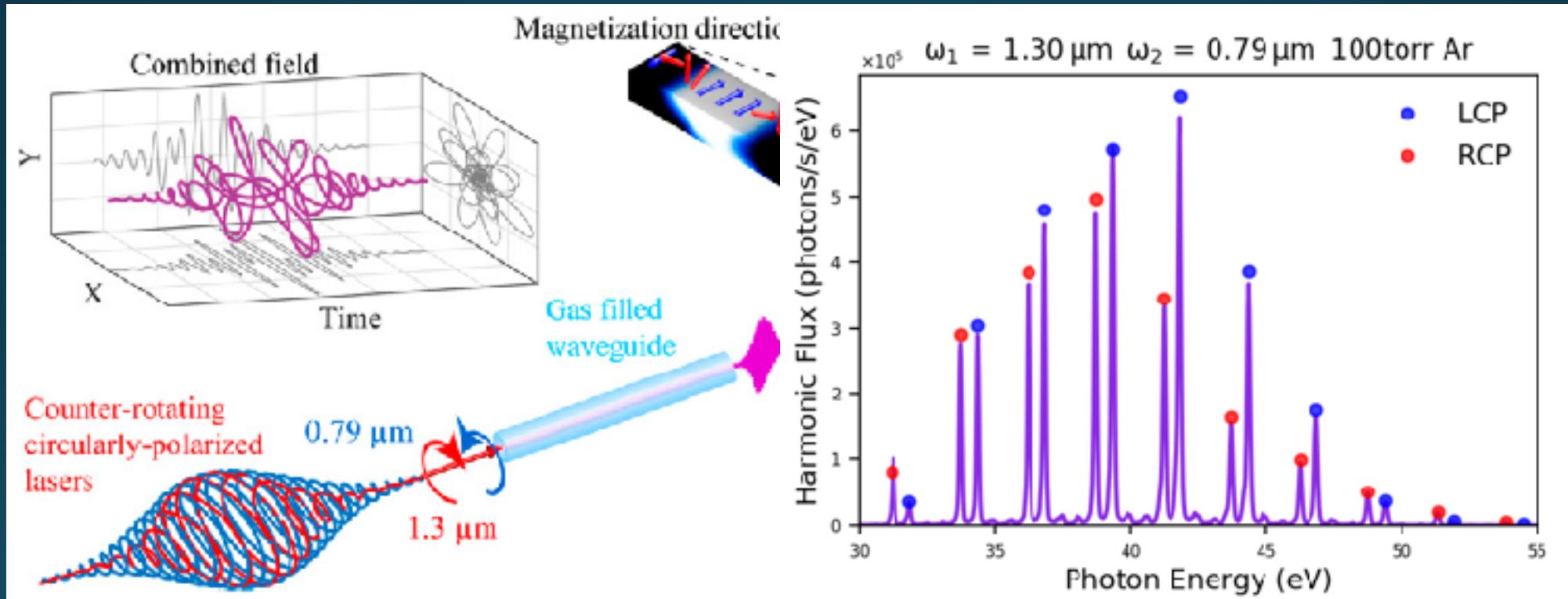
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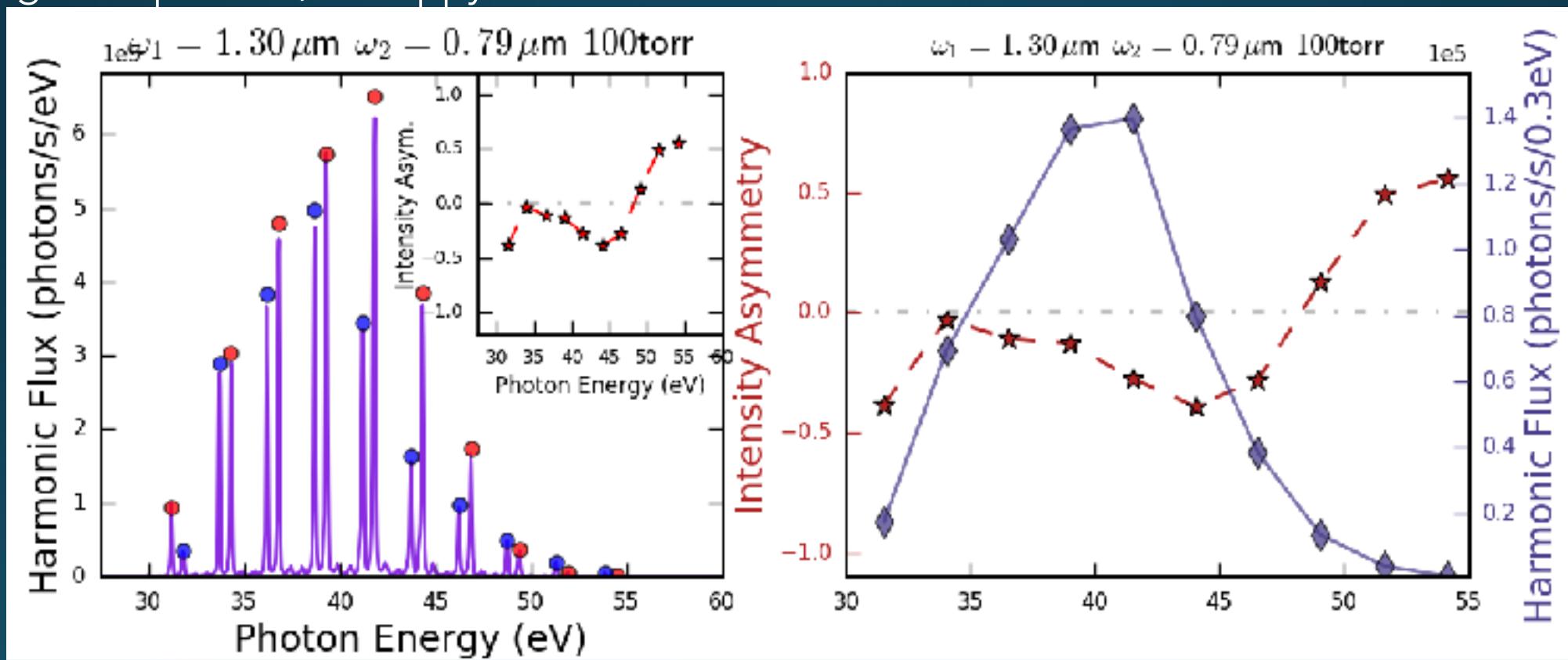
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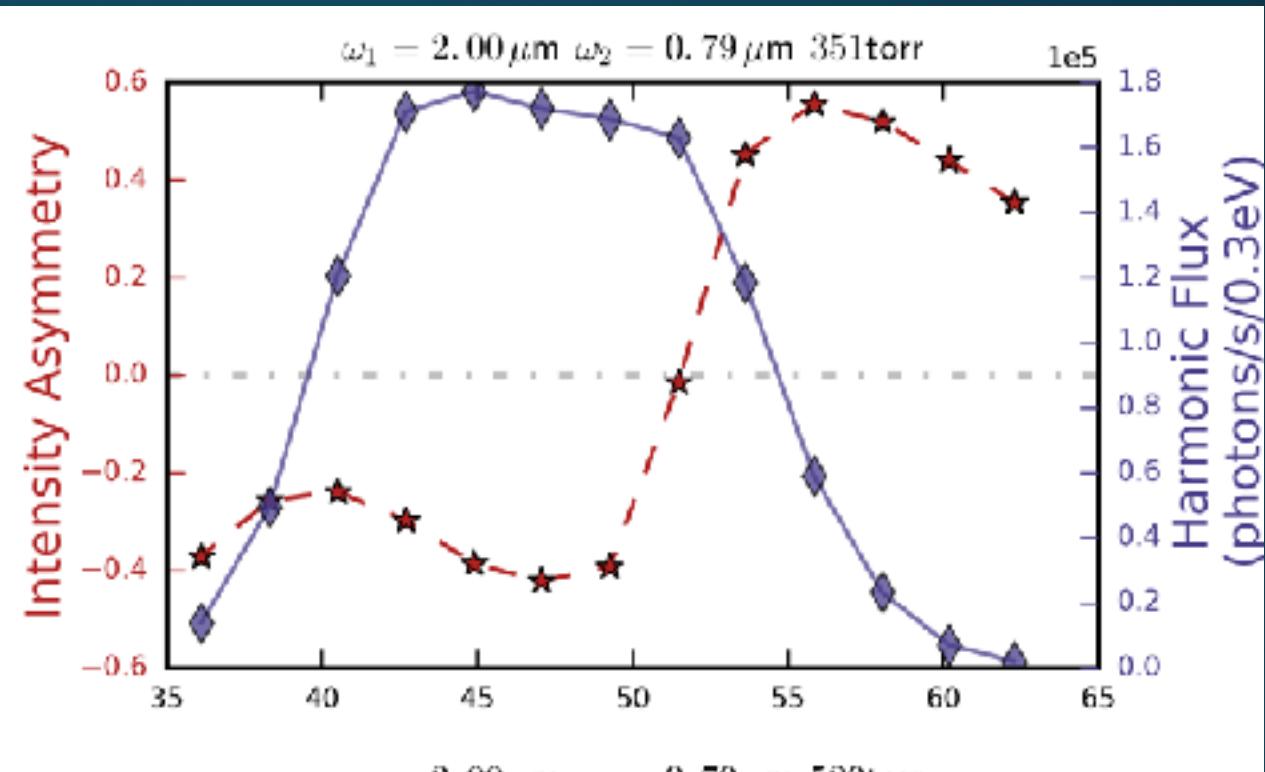
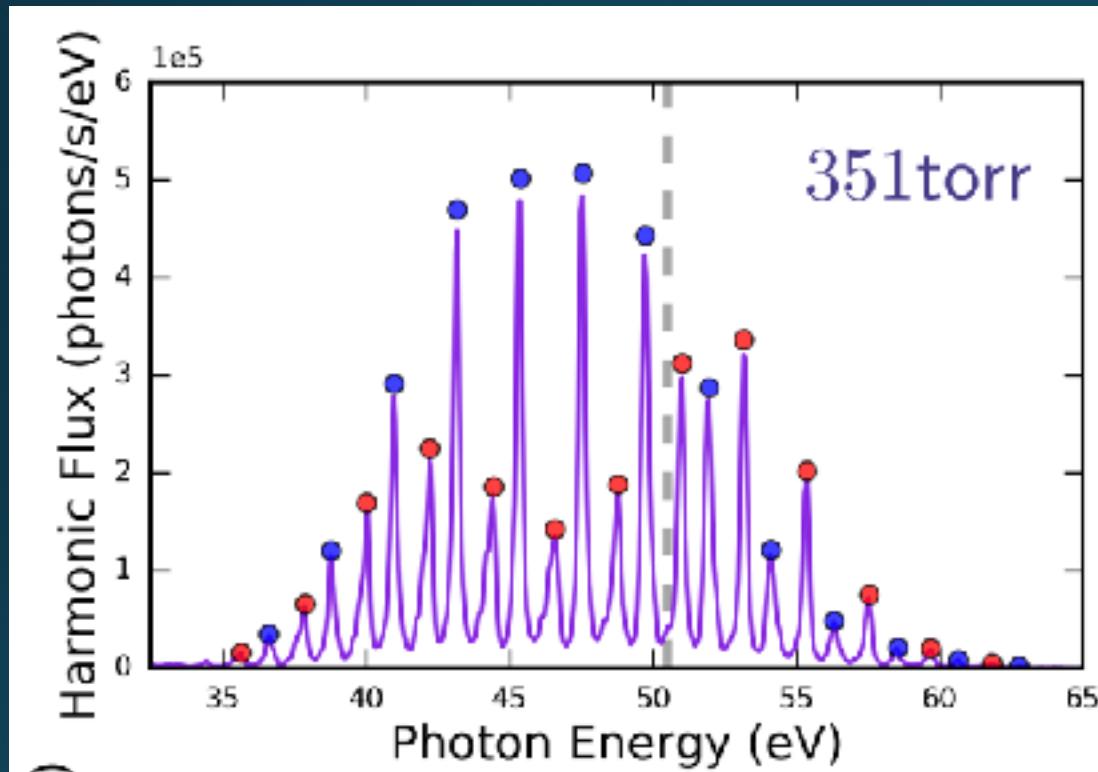
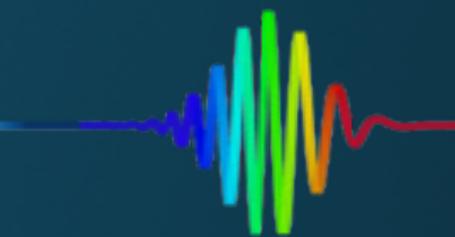


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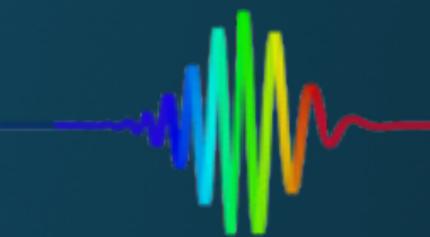


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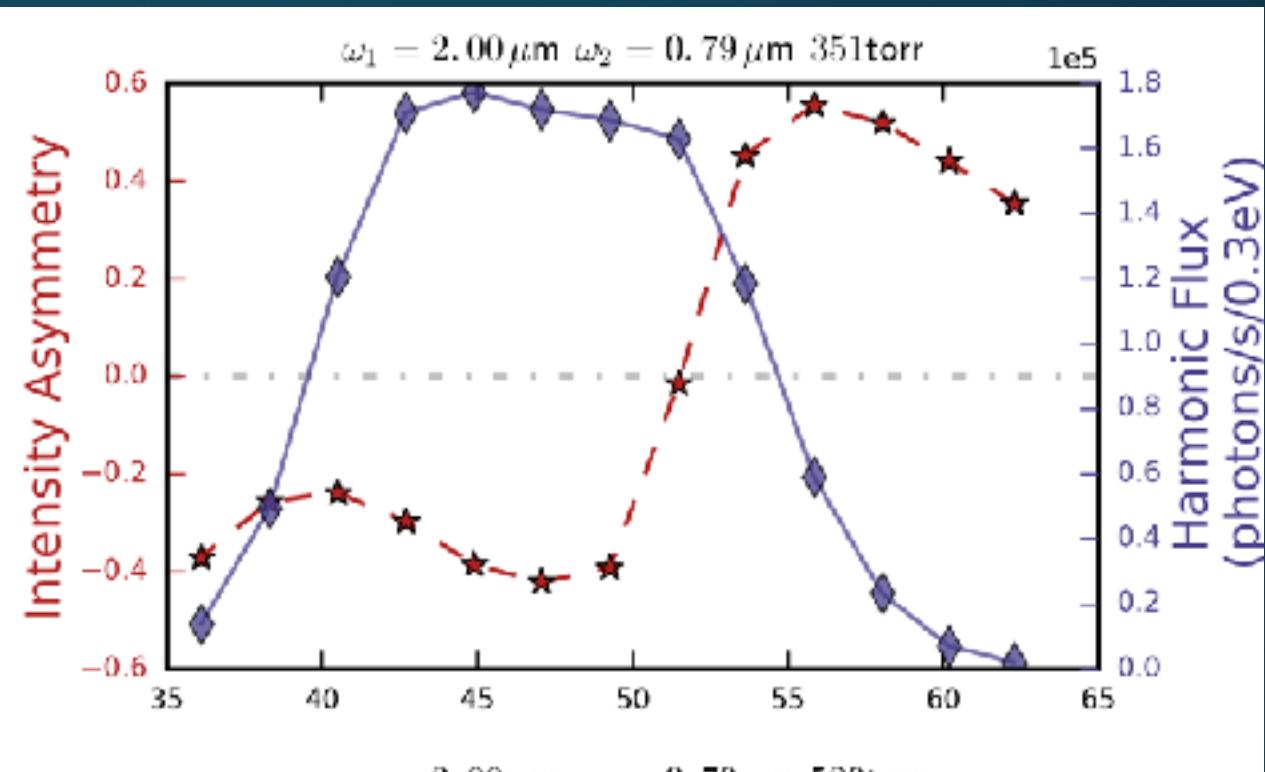
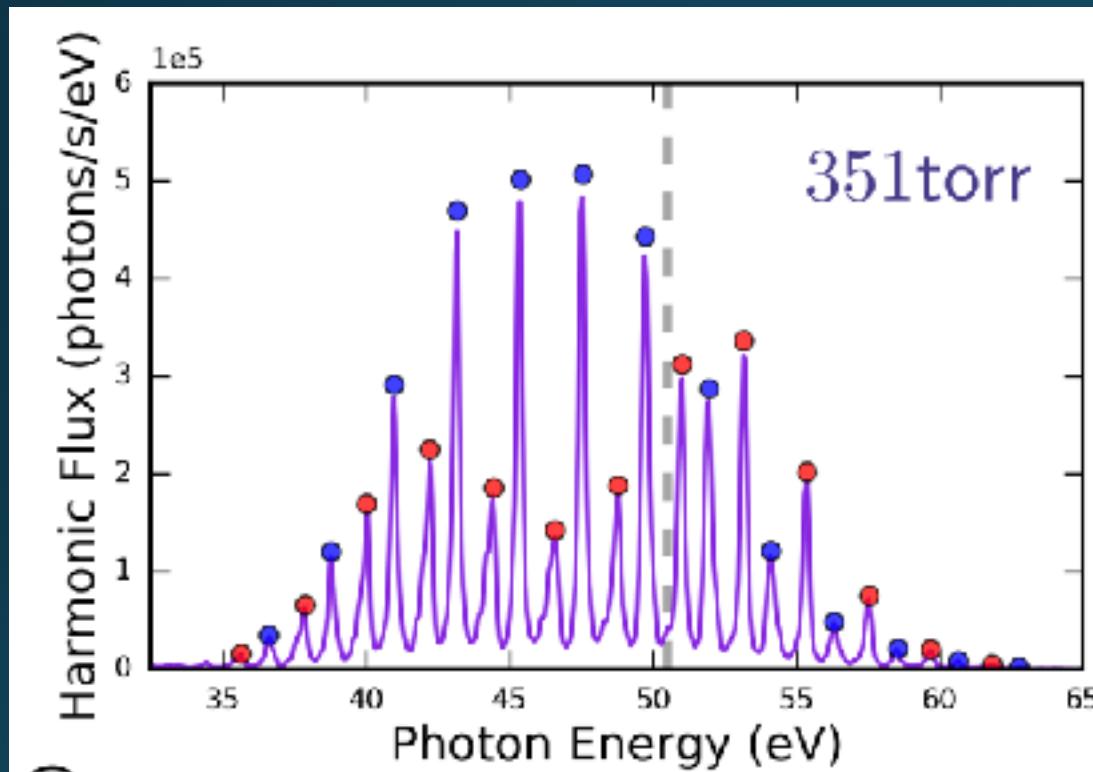




CPHHG + BHHS + IR Driving Fields: Bright, Spectrally Dense, Single-Helicity CPHHG Spectra!

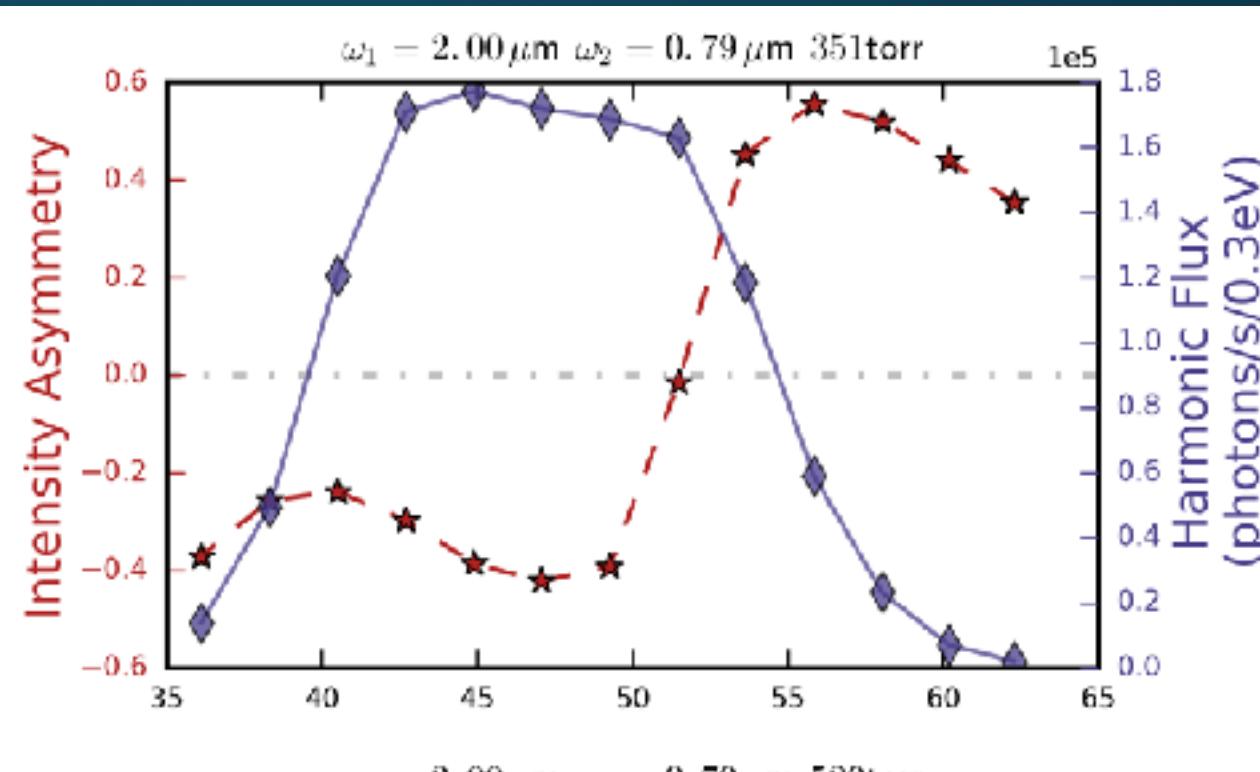
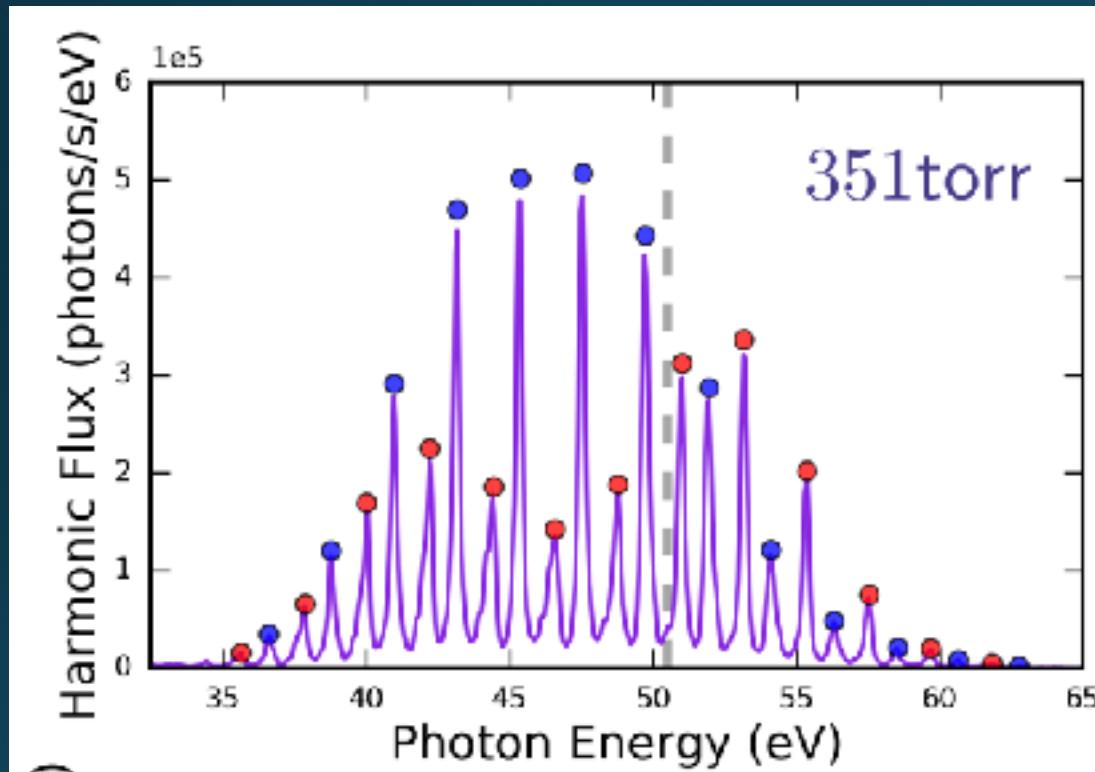


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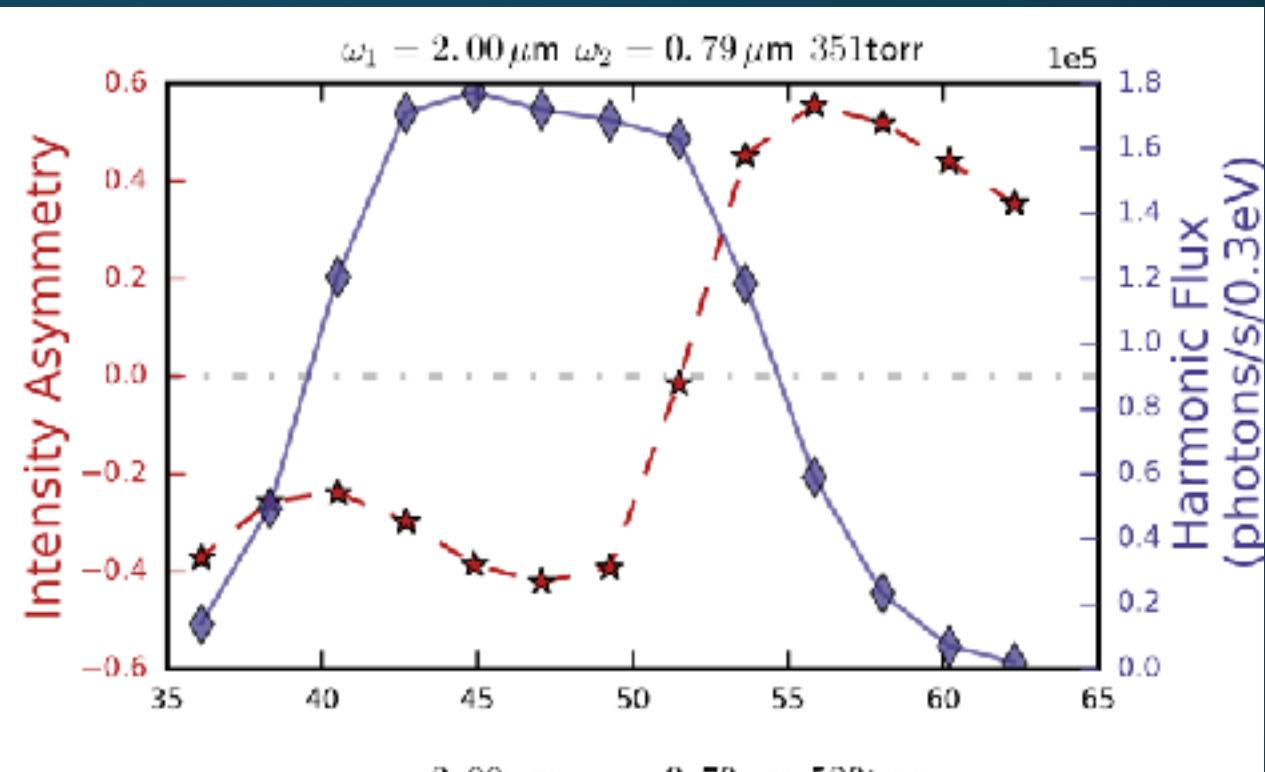
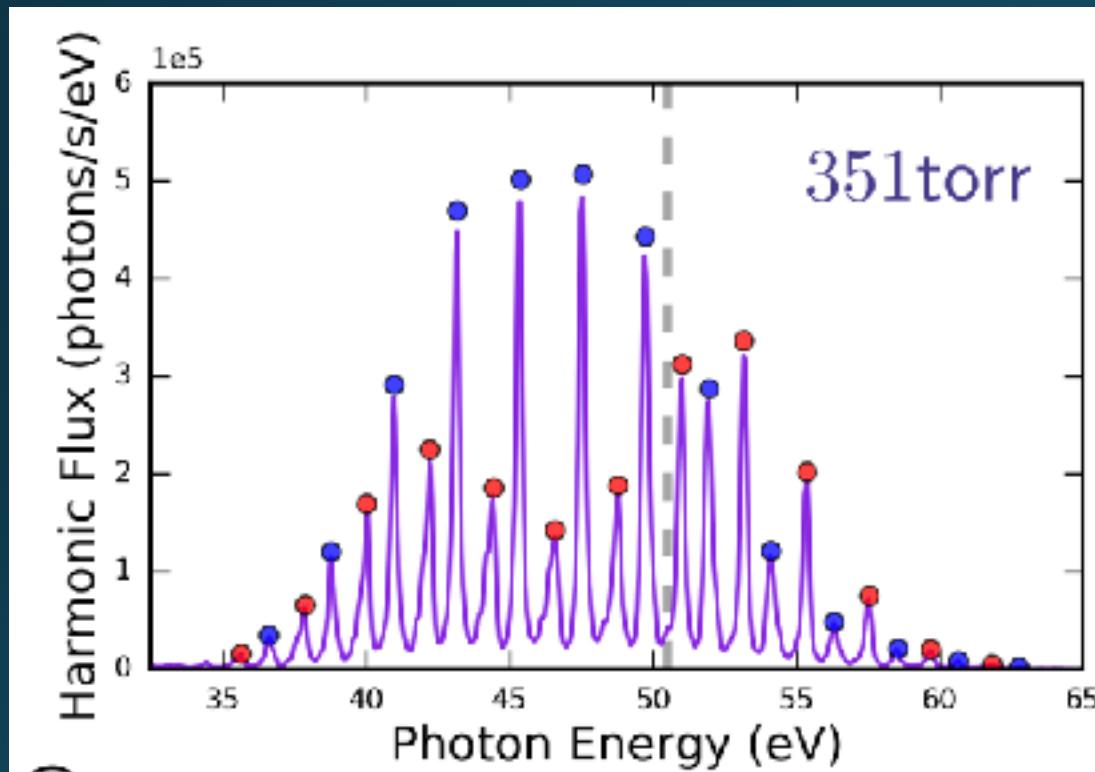


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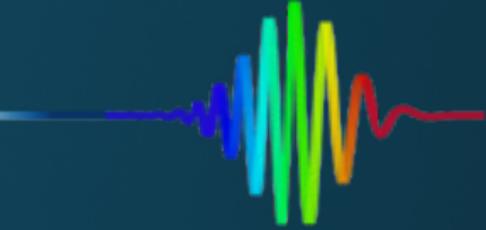


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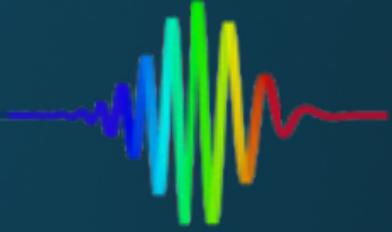
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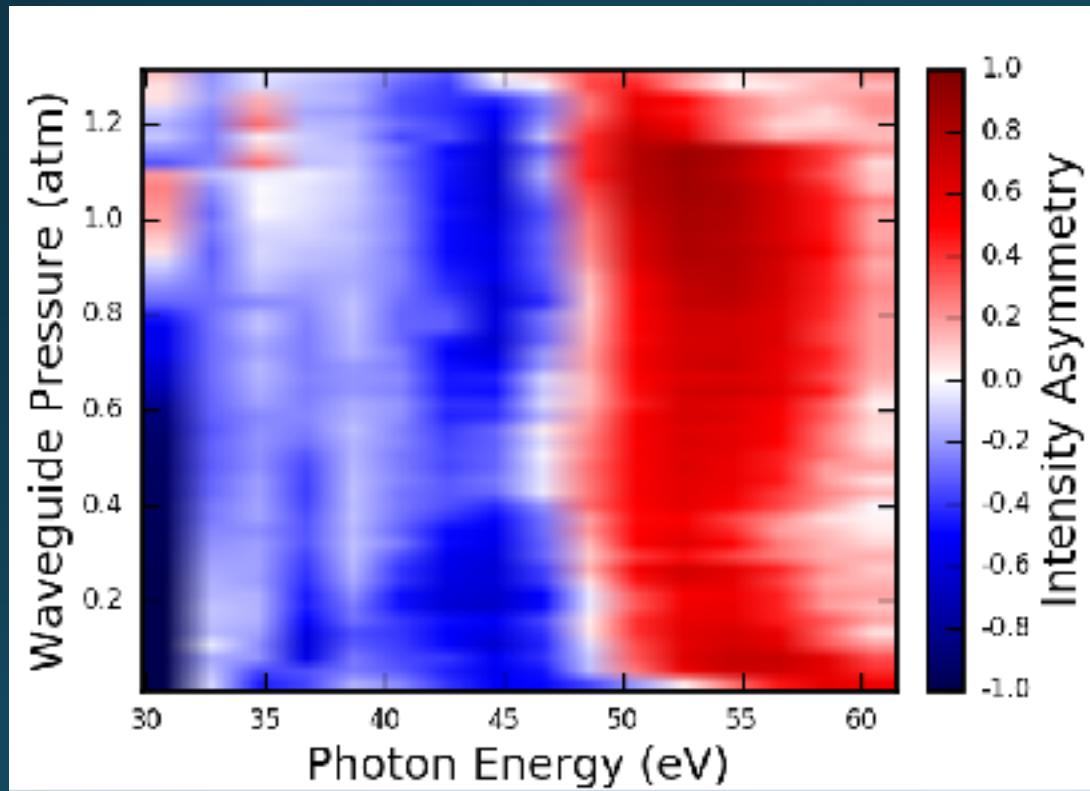
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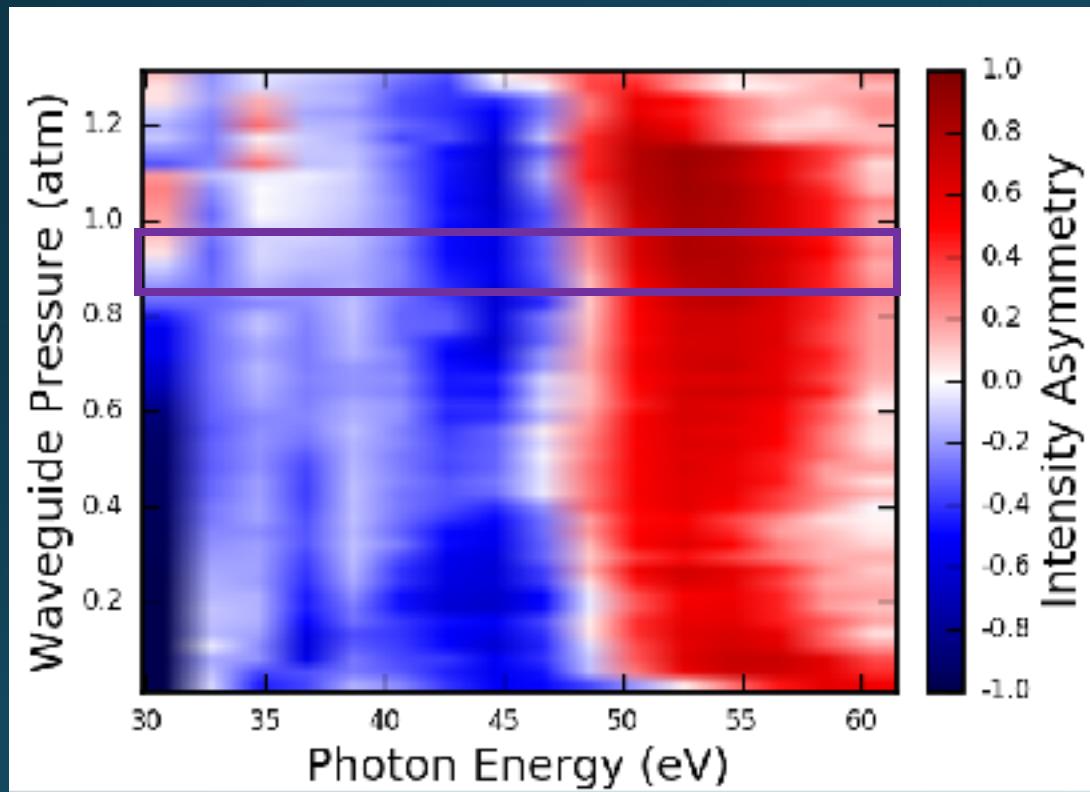
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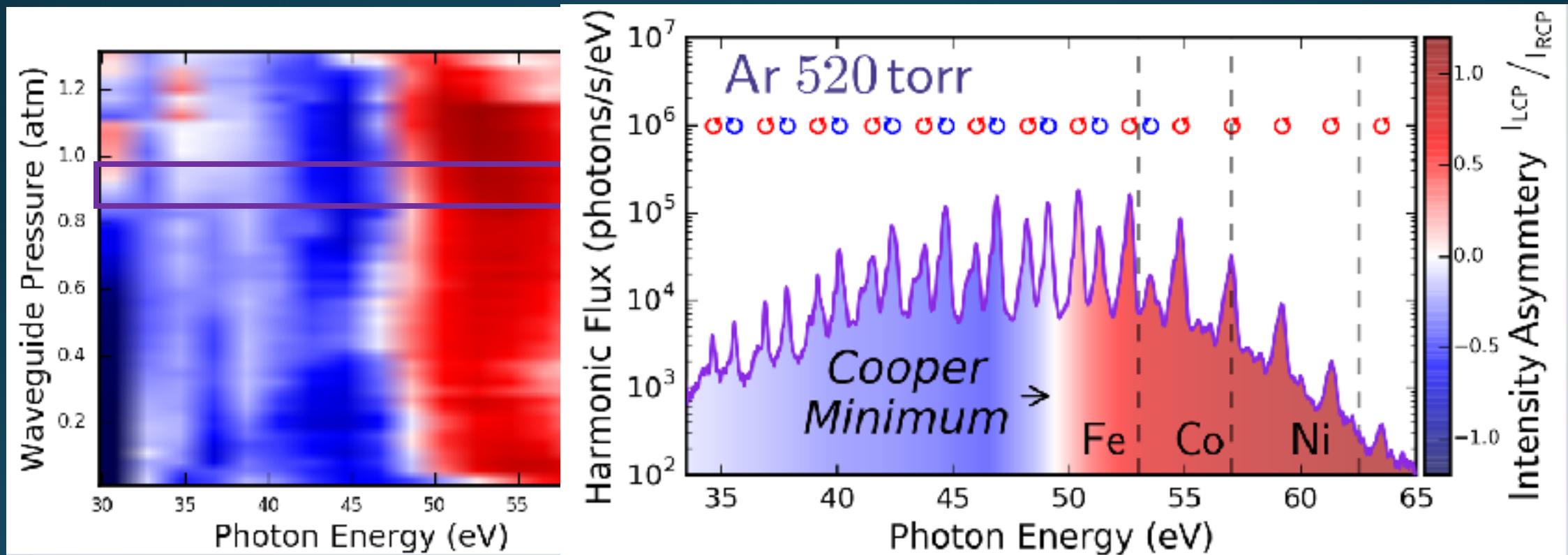
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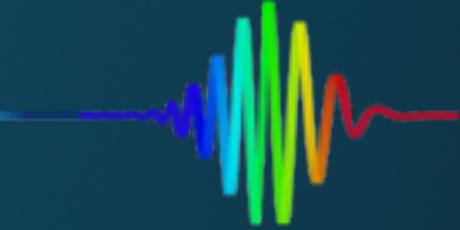
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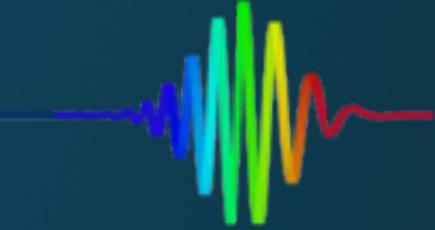
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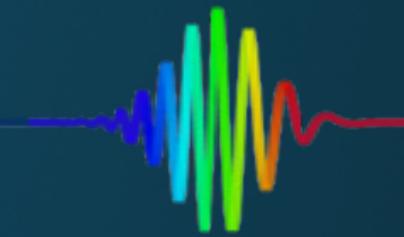




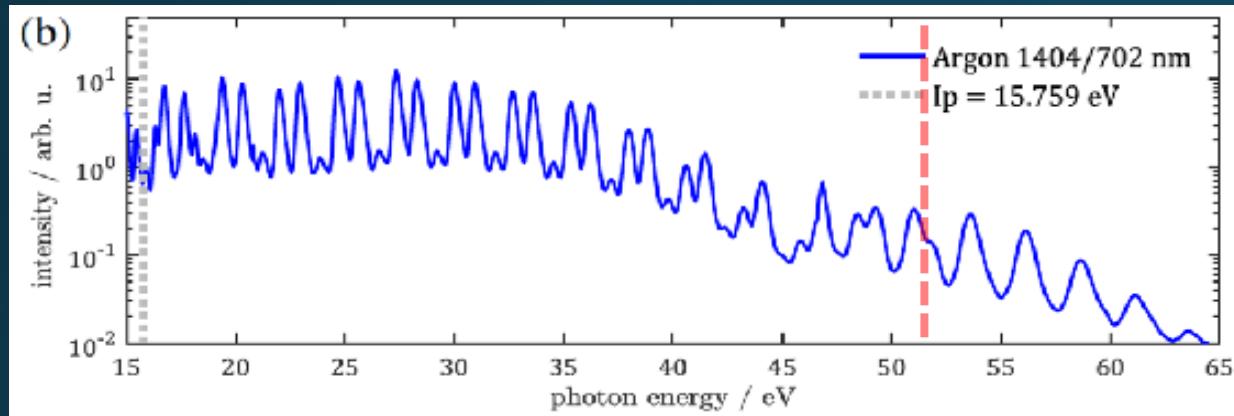
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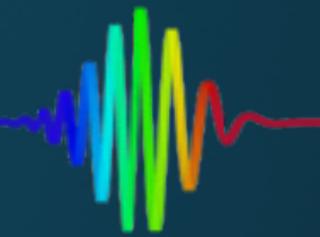
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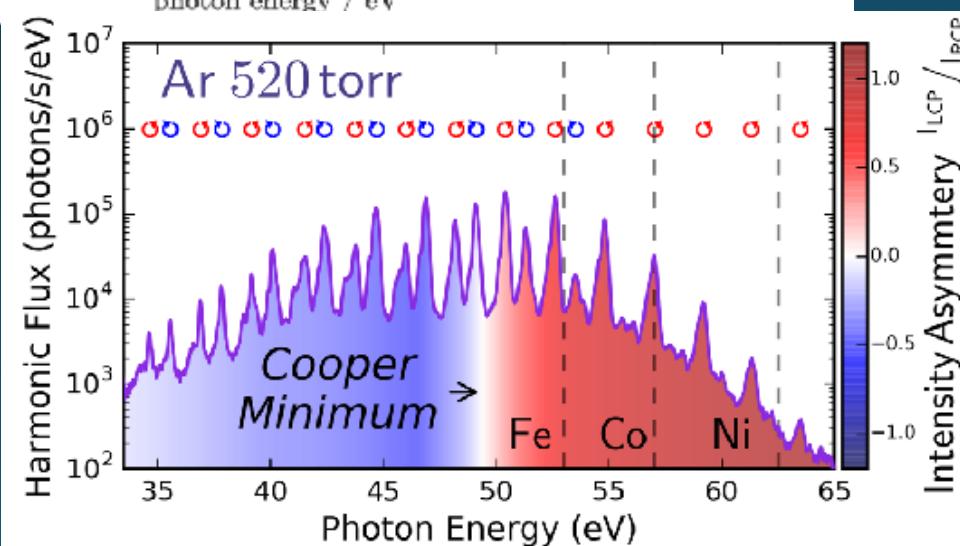
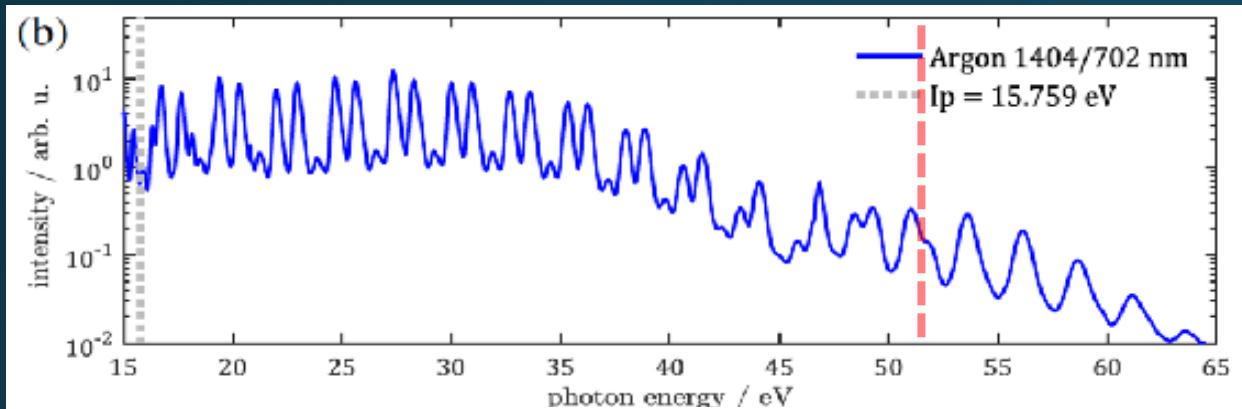


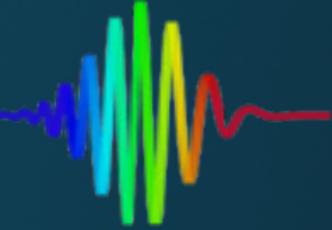
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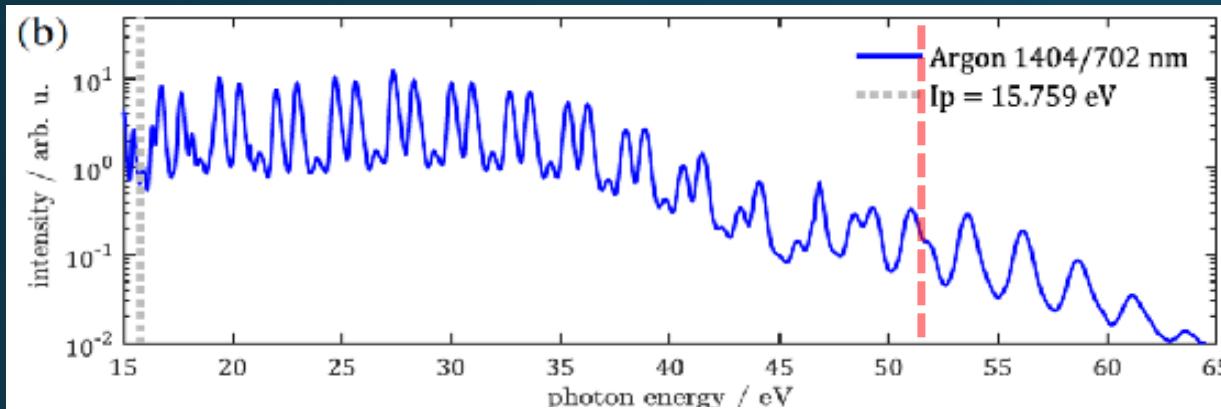


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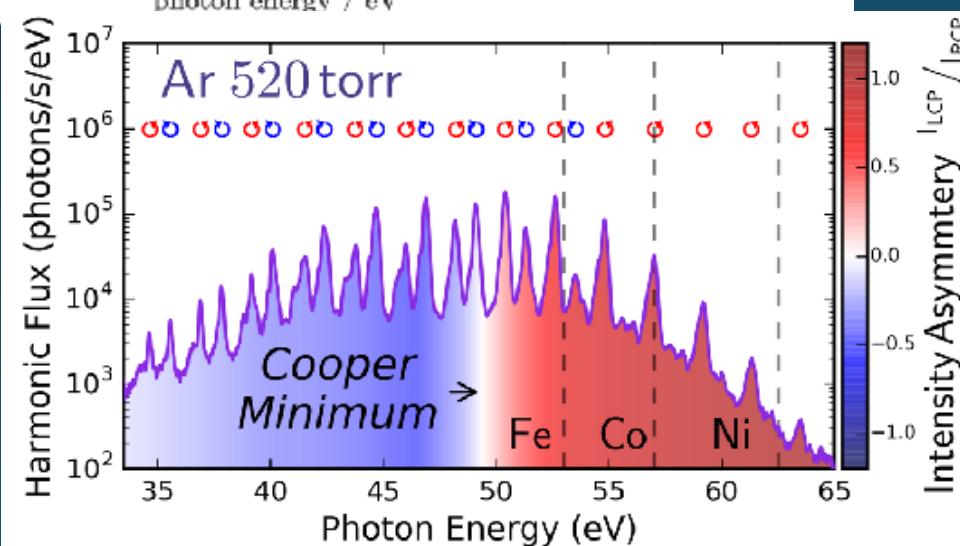




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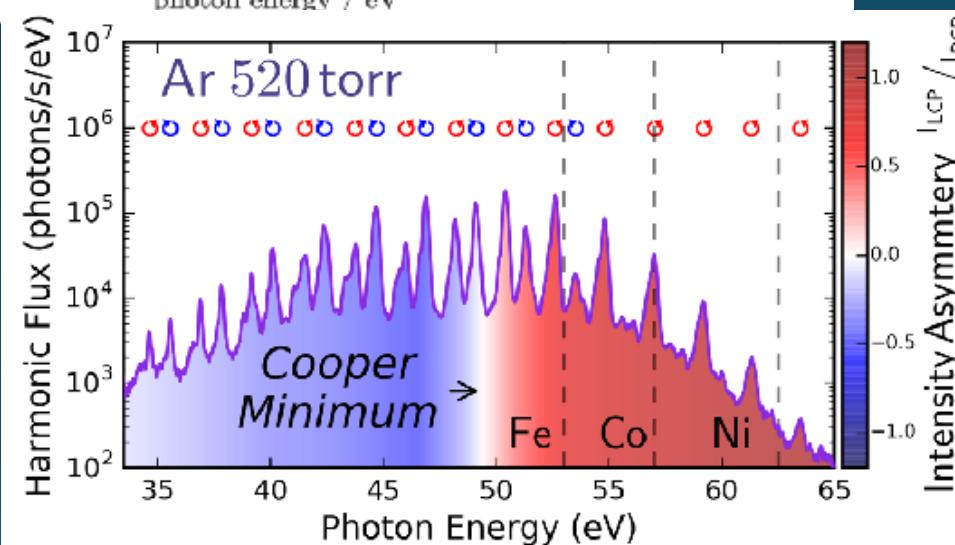
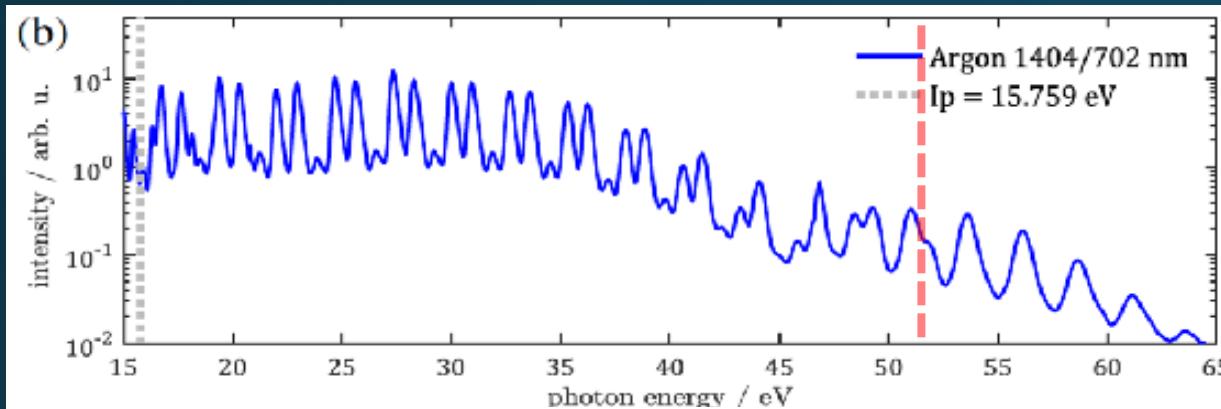


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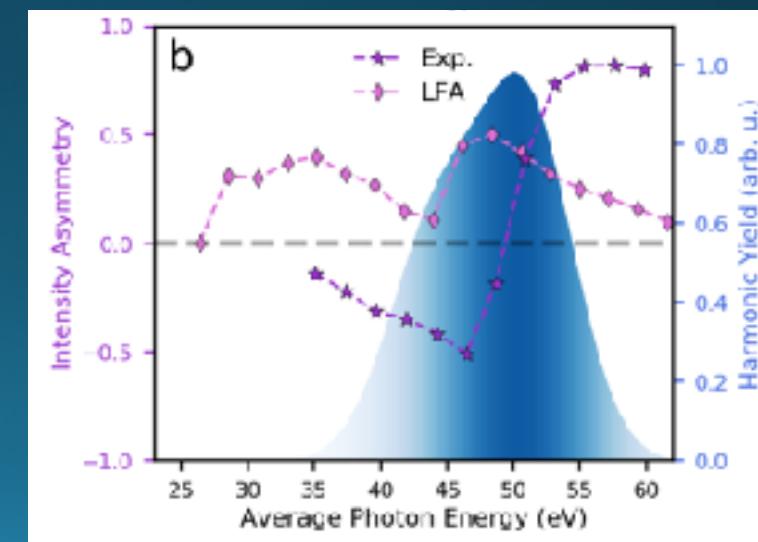




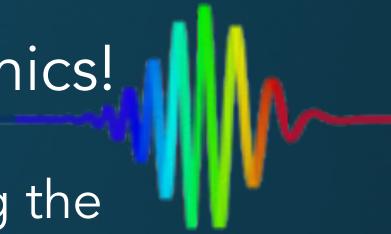
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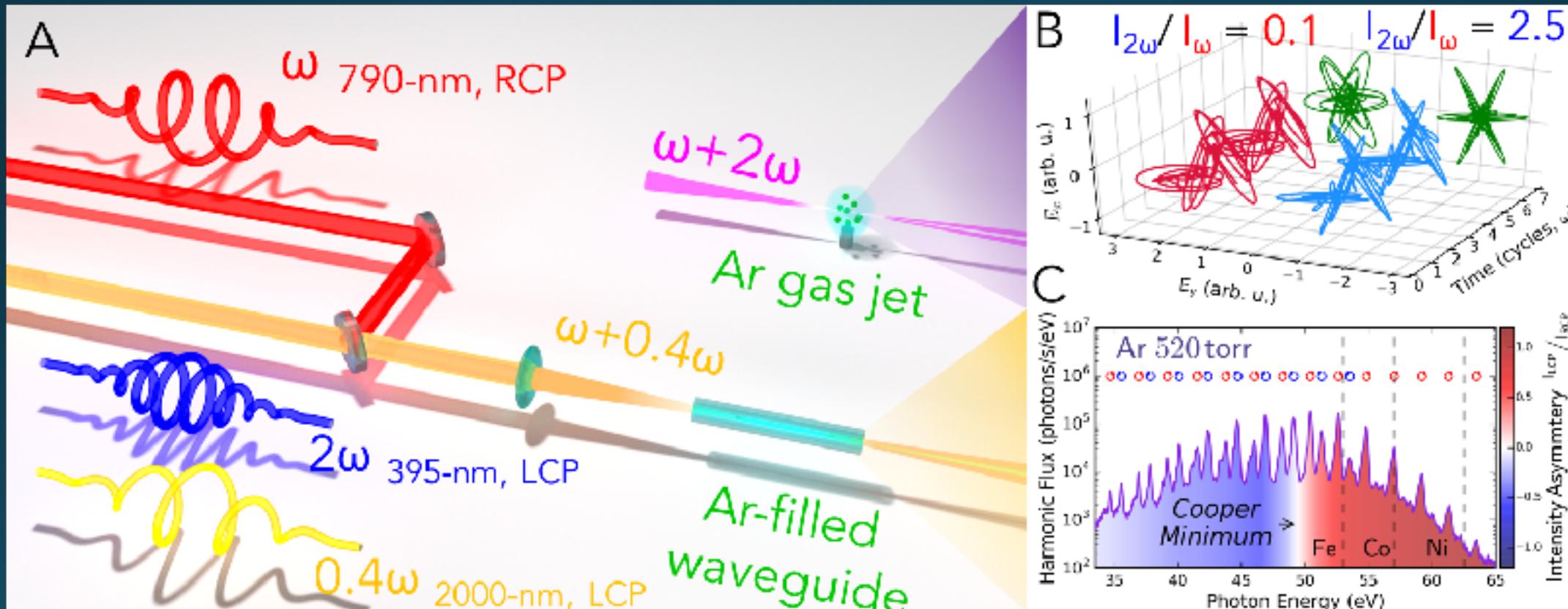
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Bicircular HHG and HHS: A Versatile Light Source for Chiral Spectroscopies and Dynamics!

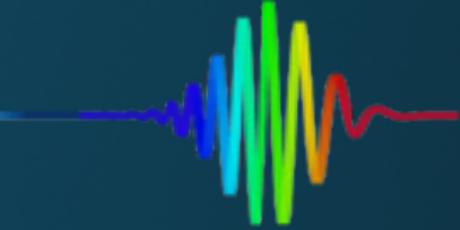


- Straightforward, versatile control of the attosecond polarization, all while preserving the spectral circularity of the harmonics!

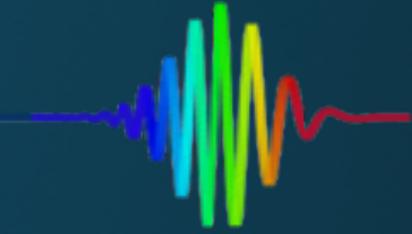




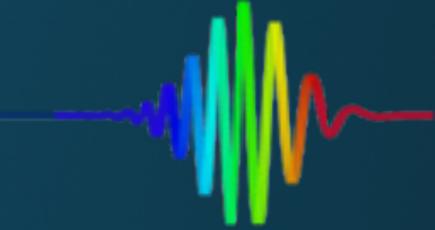
CPHHG from Linearly Polarized HHG Drivers: A New (or Lack of) Twist in Thinking



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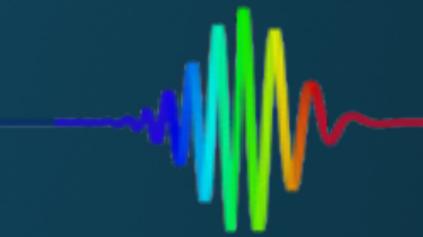


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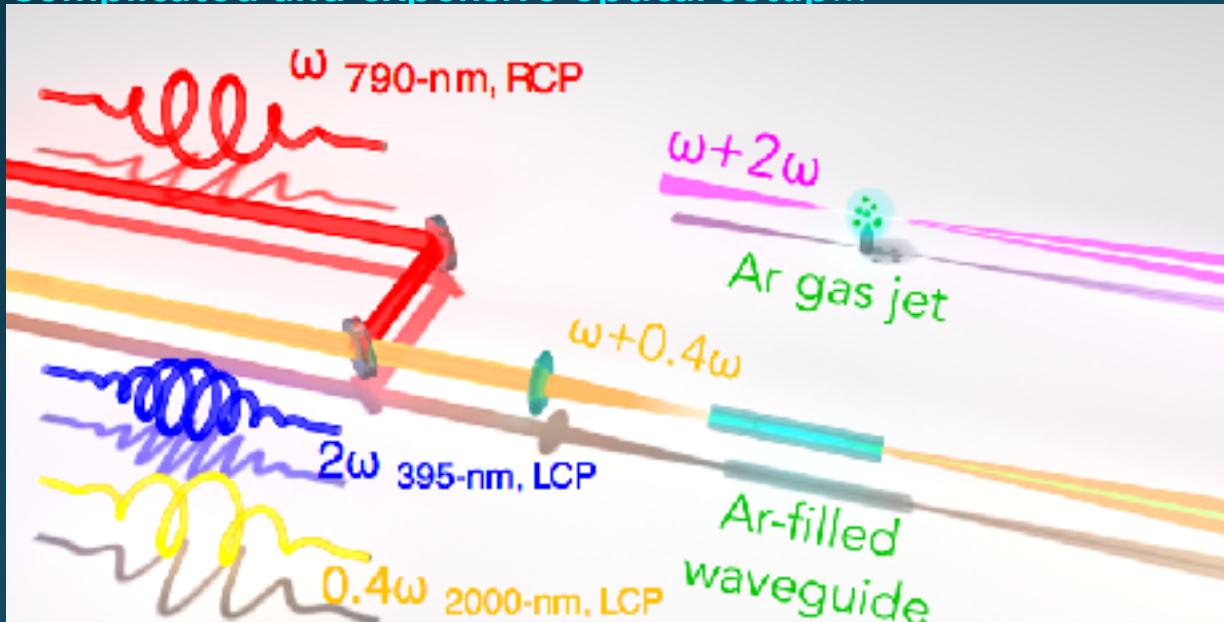


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CPHHG from Linearly Polarized HHG Drivers: A New (or Lack of) Twist in Thinking

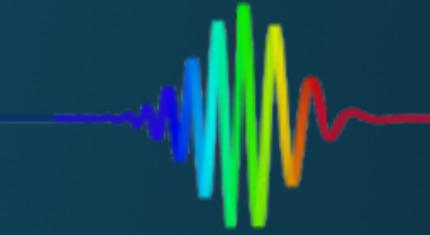


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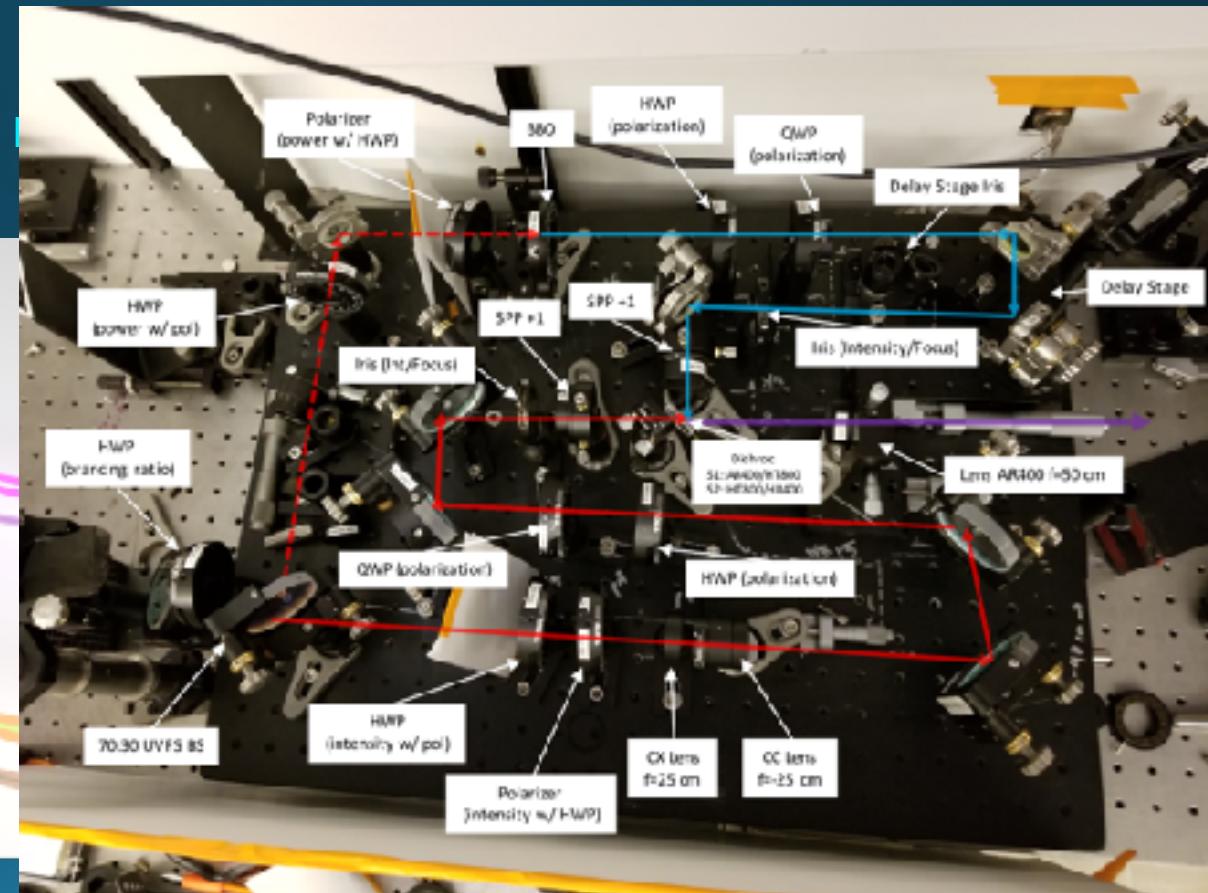
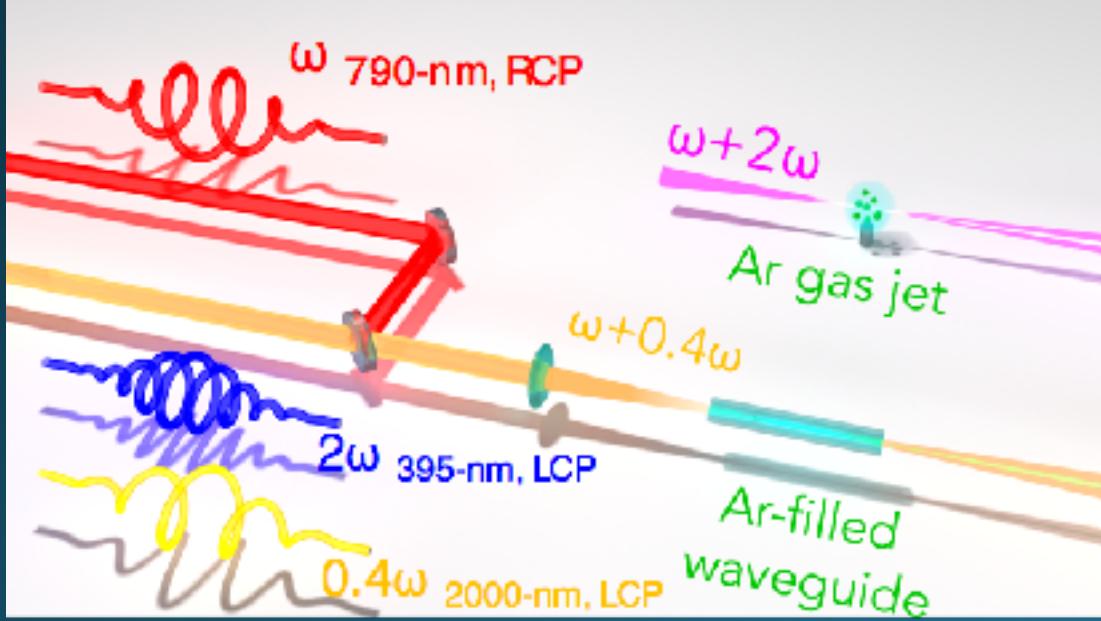


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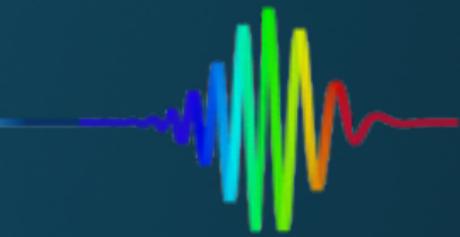


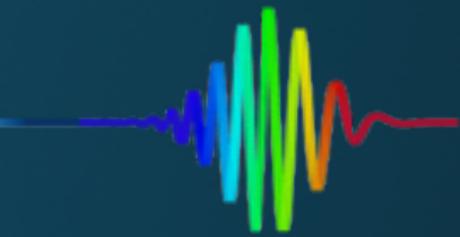
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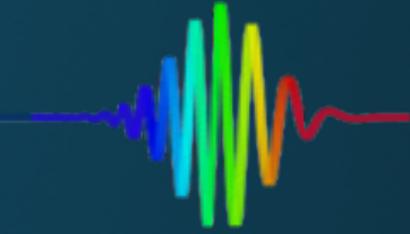


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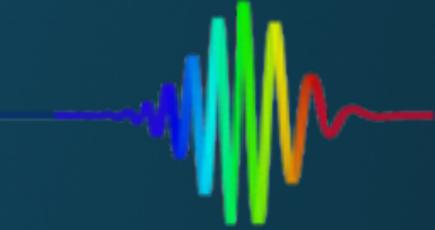


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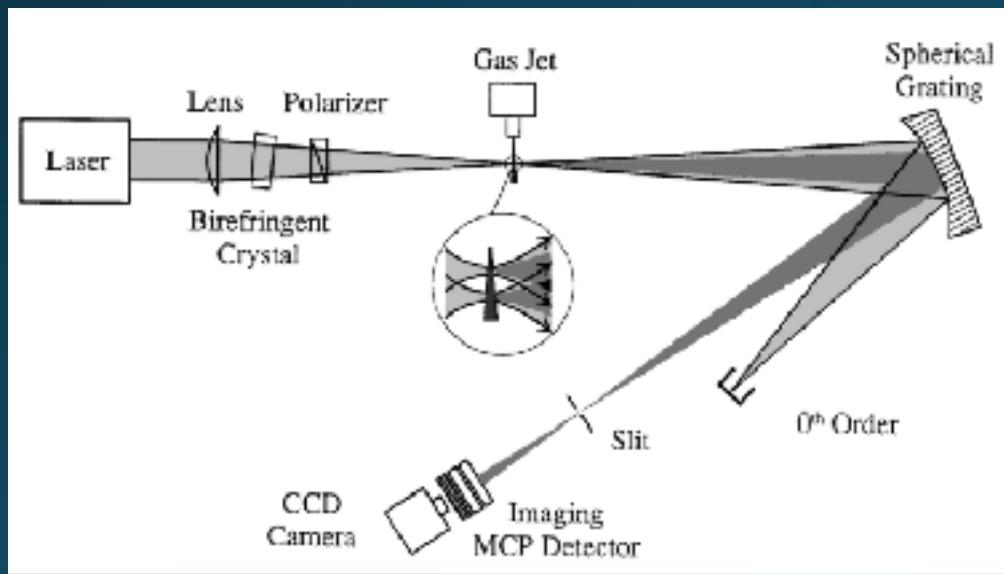


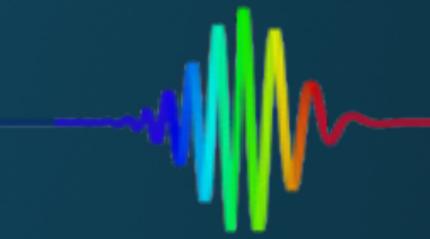
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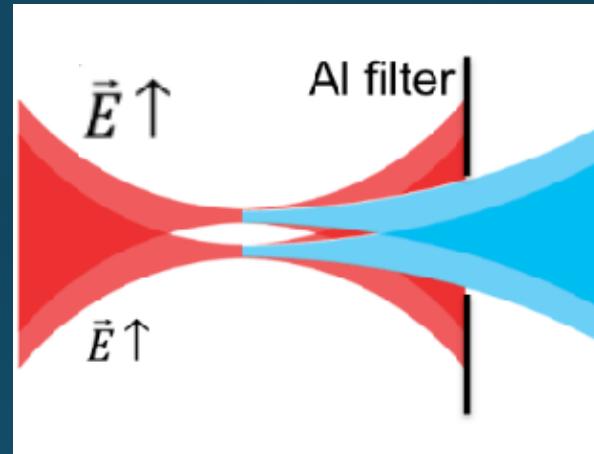
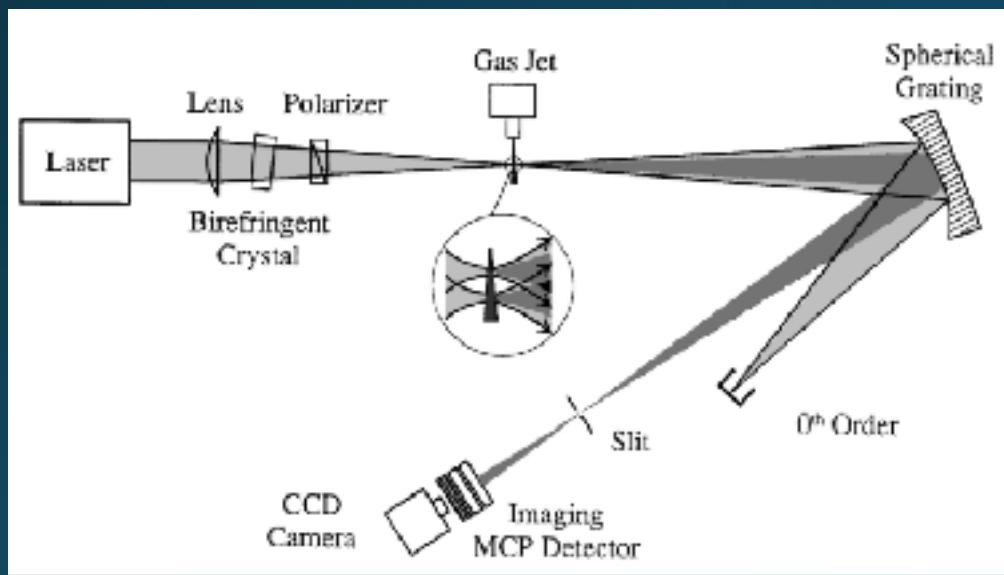


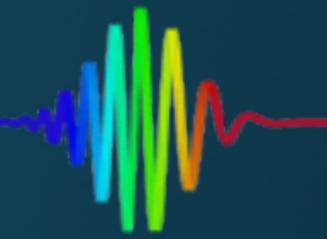
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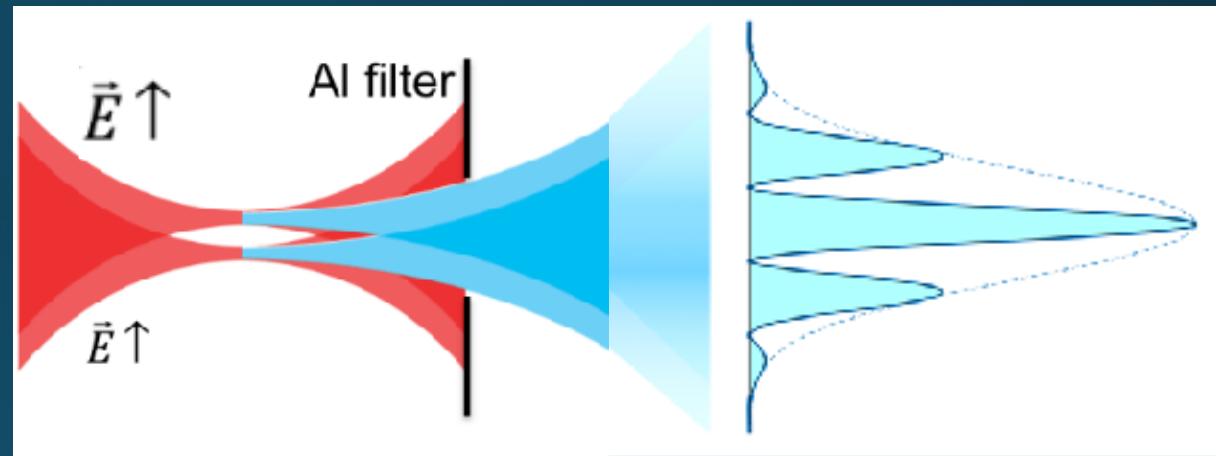
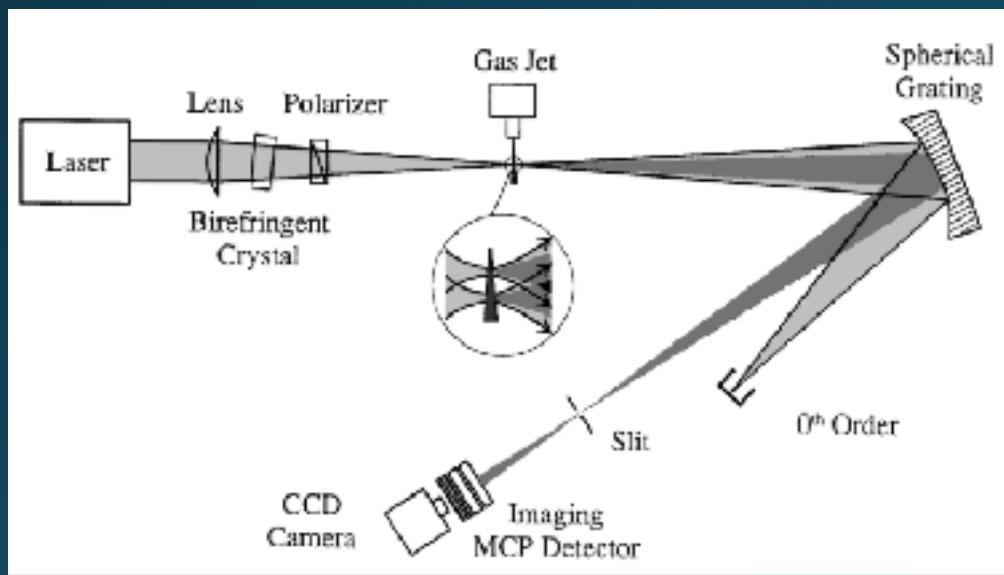
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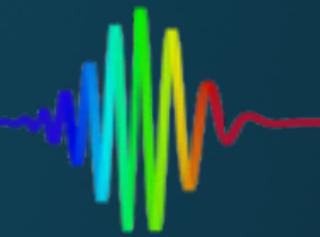
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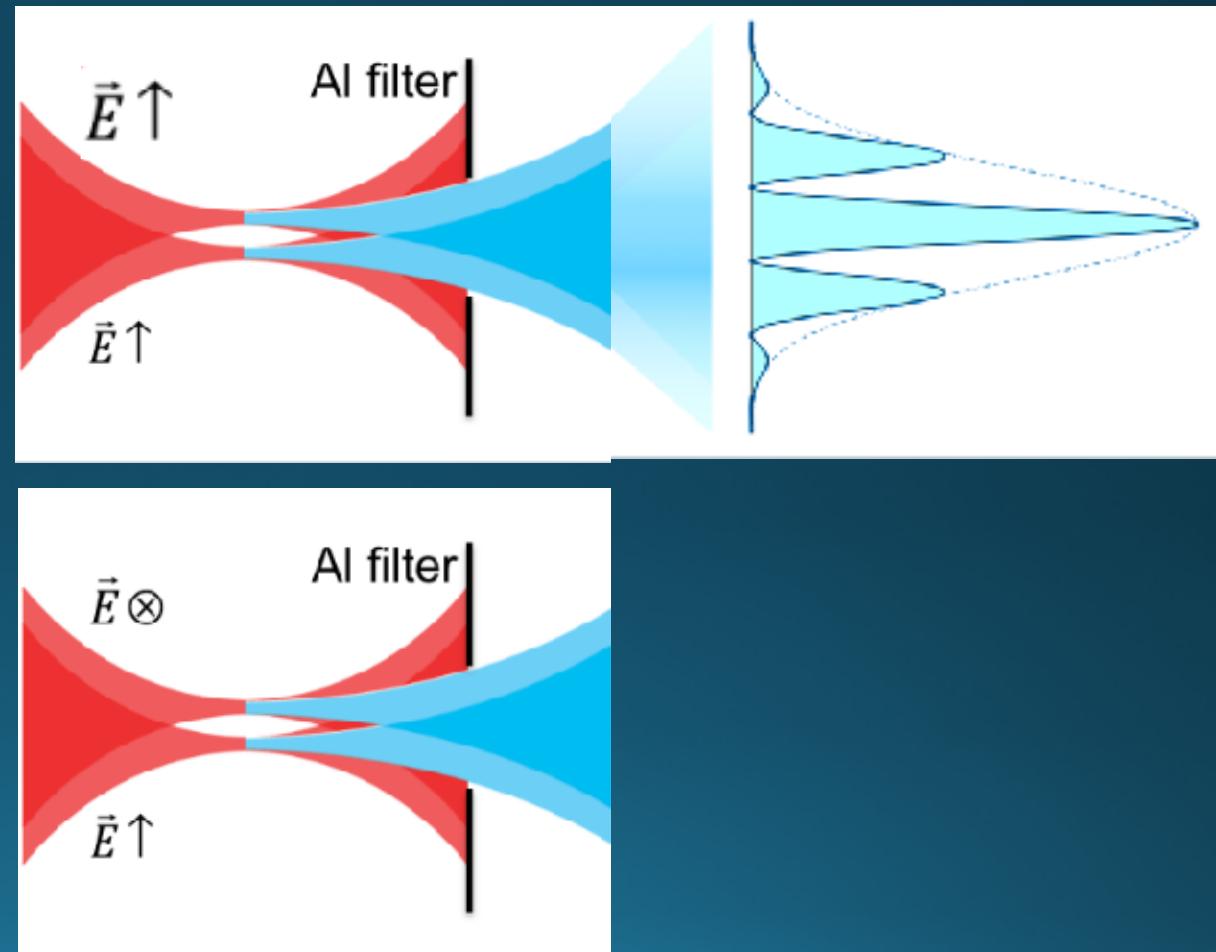
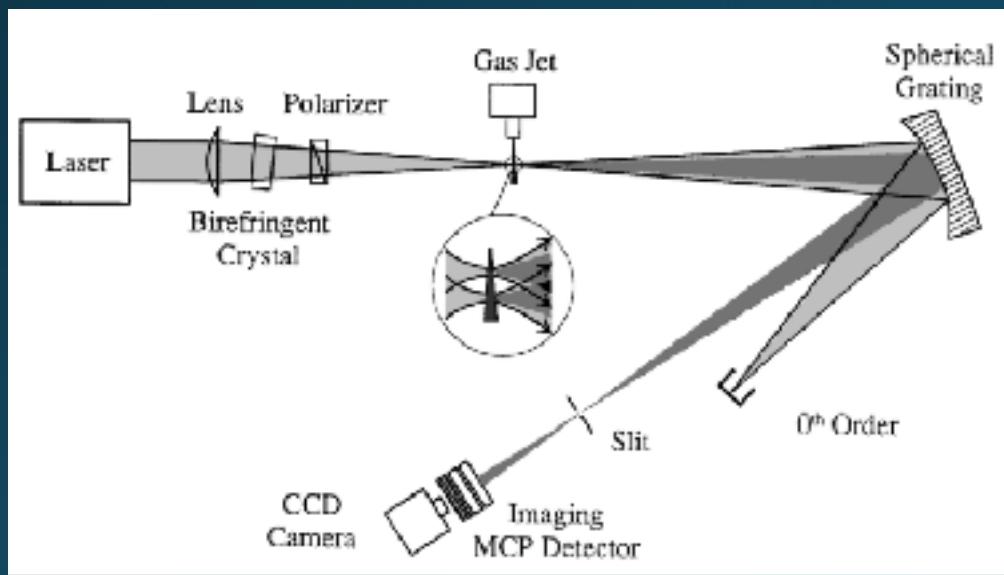
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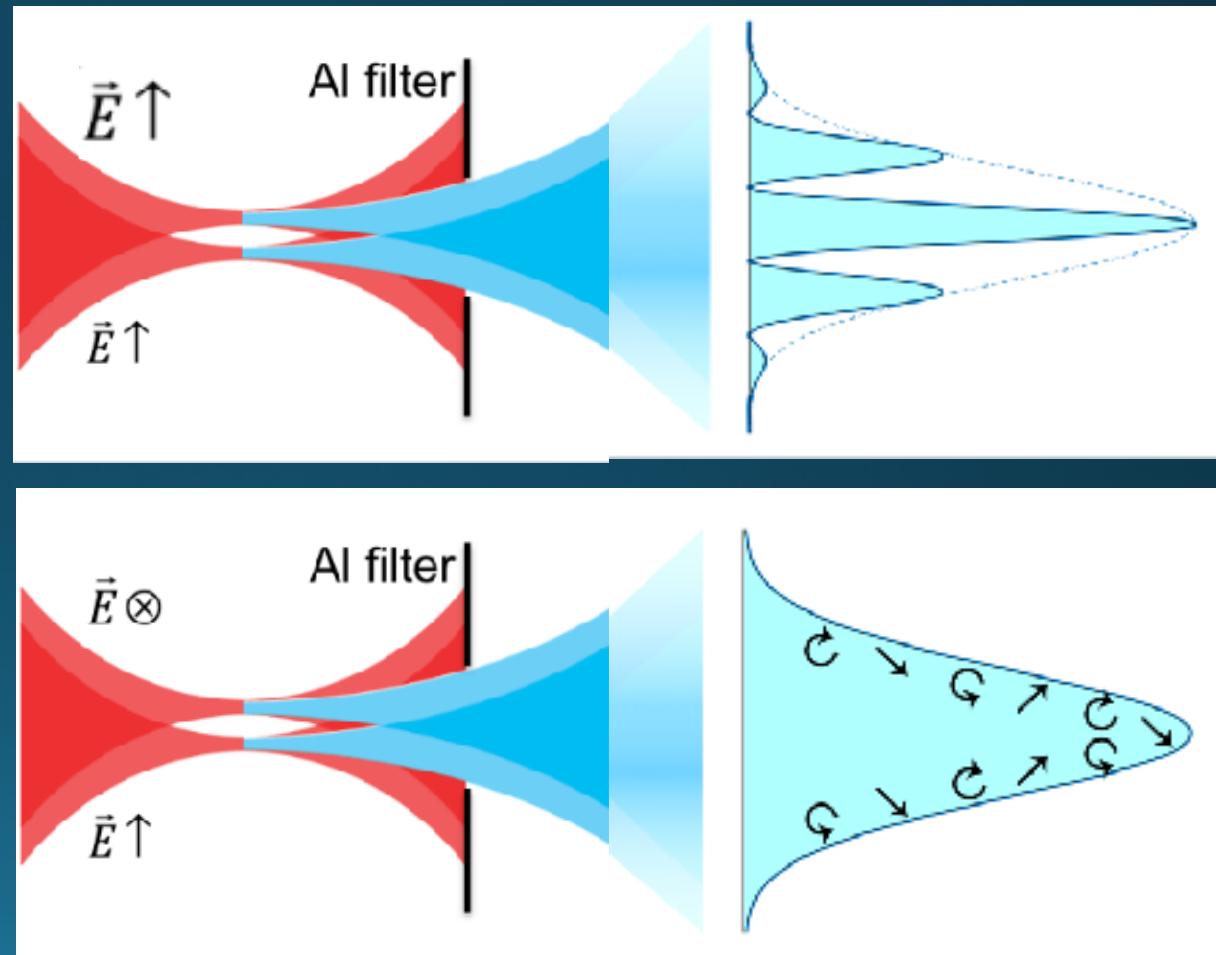
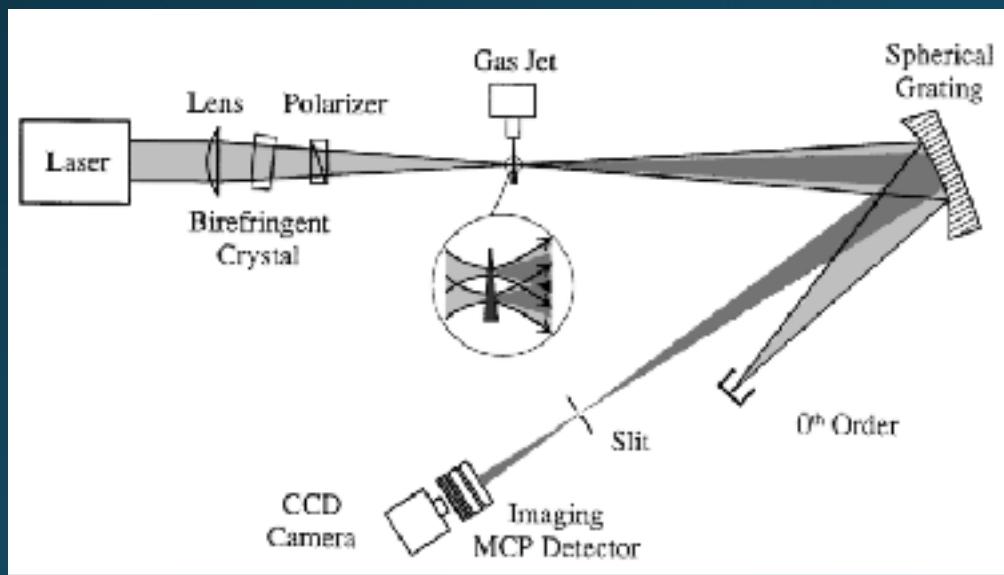
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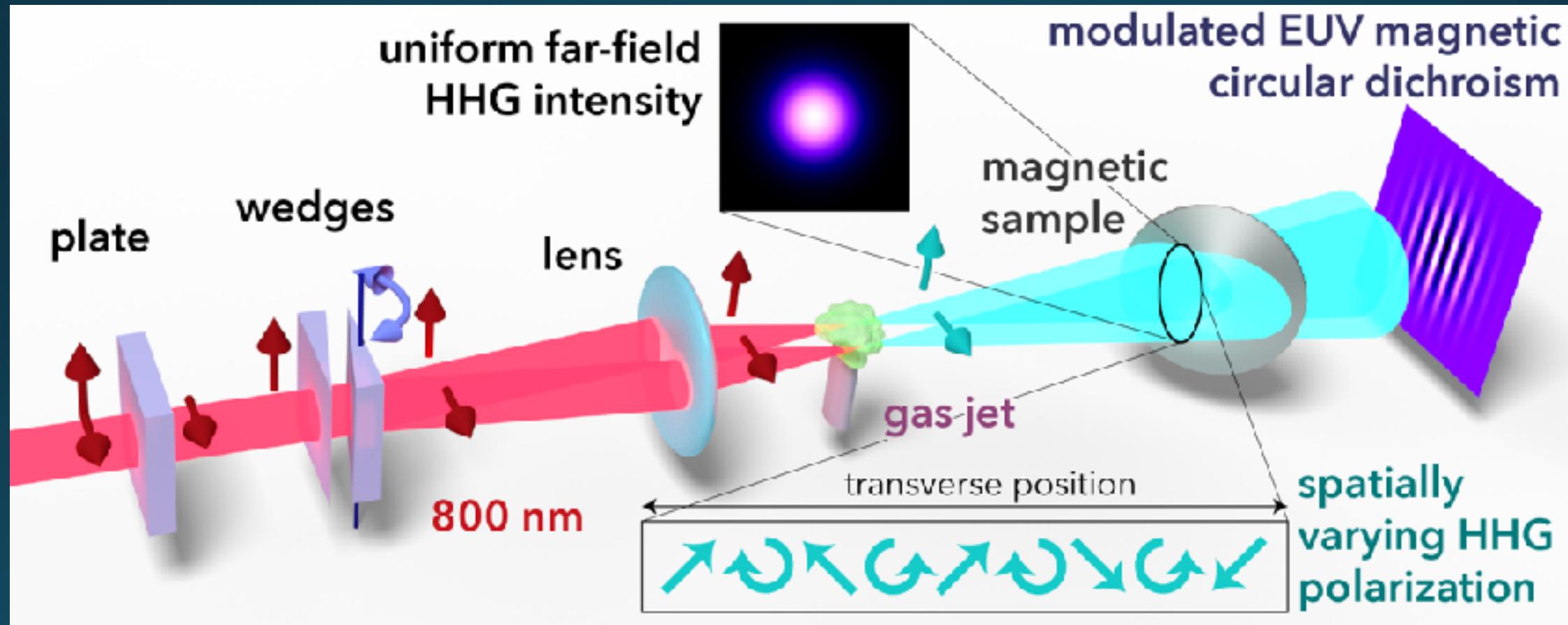
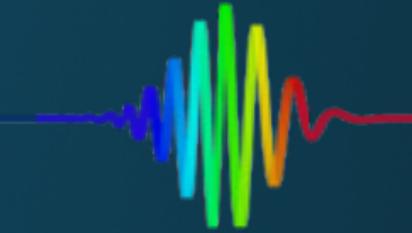


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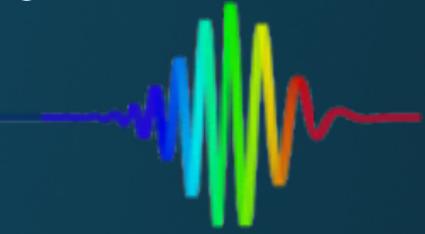
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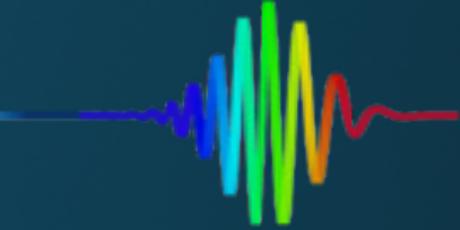
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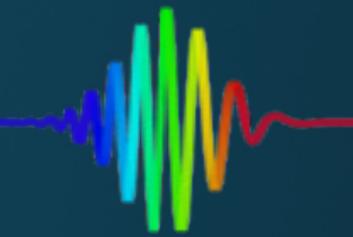




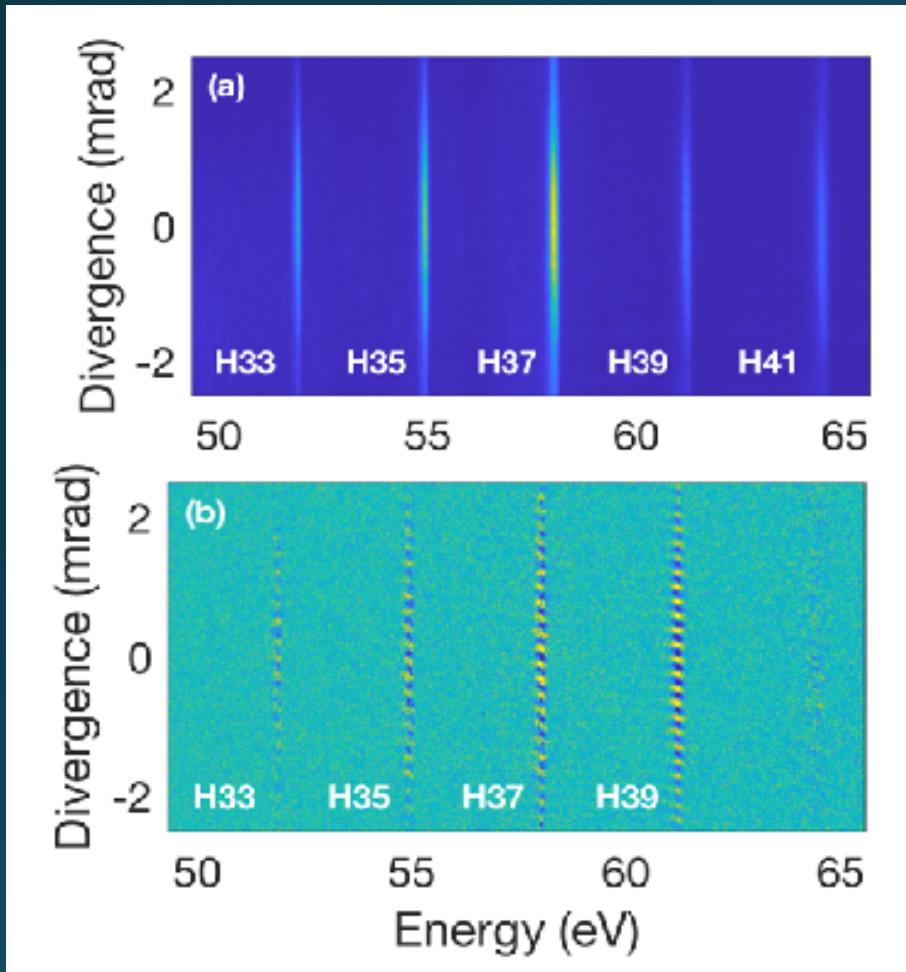
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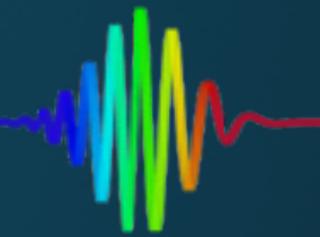
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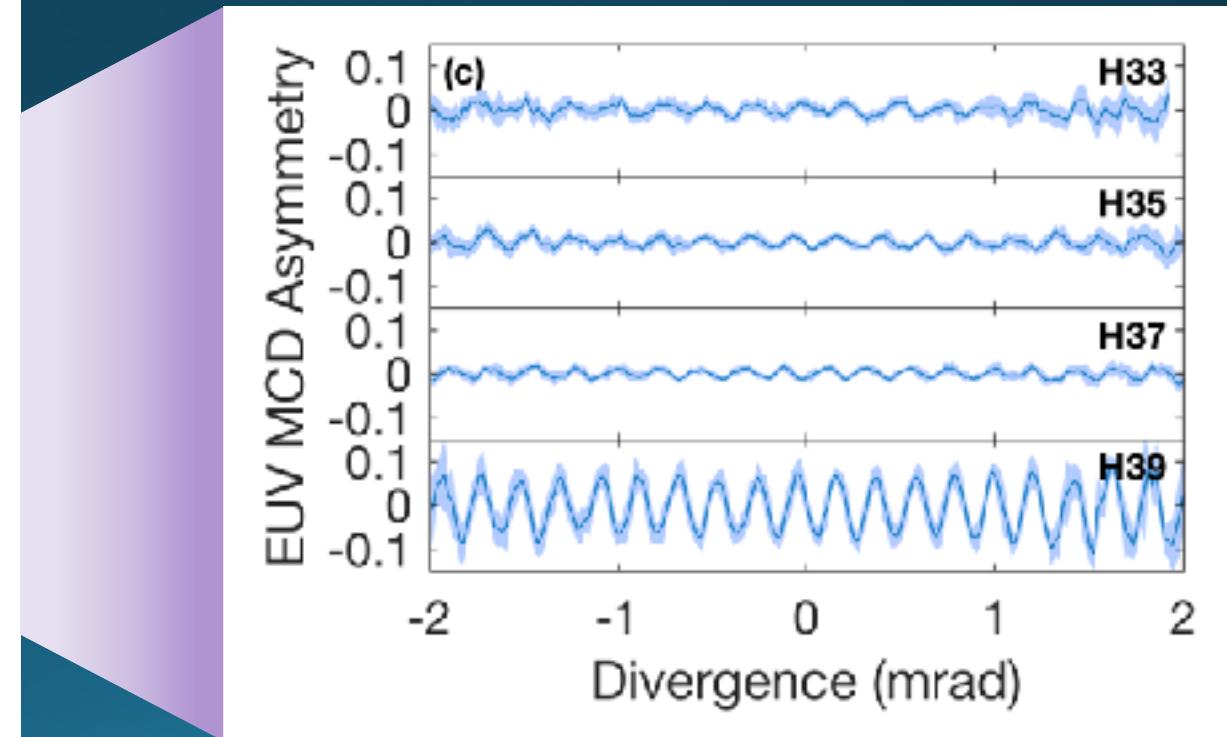
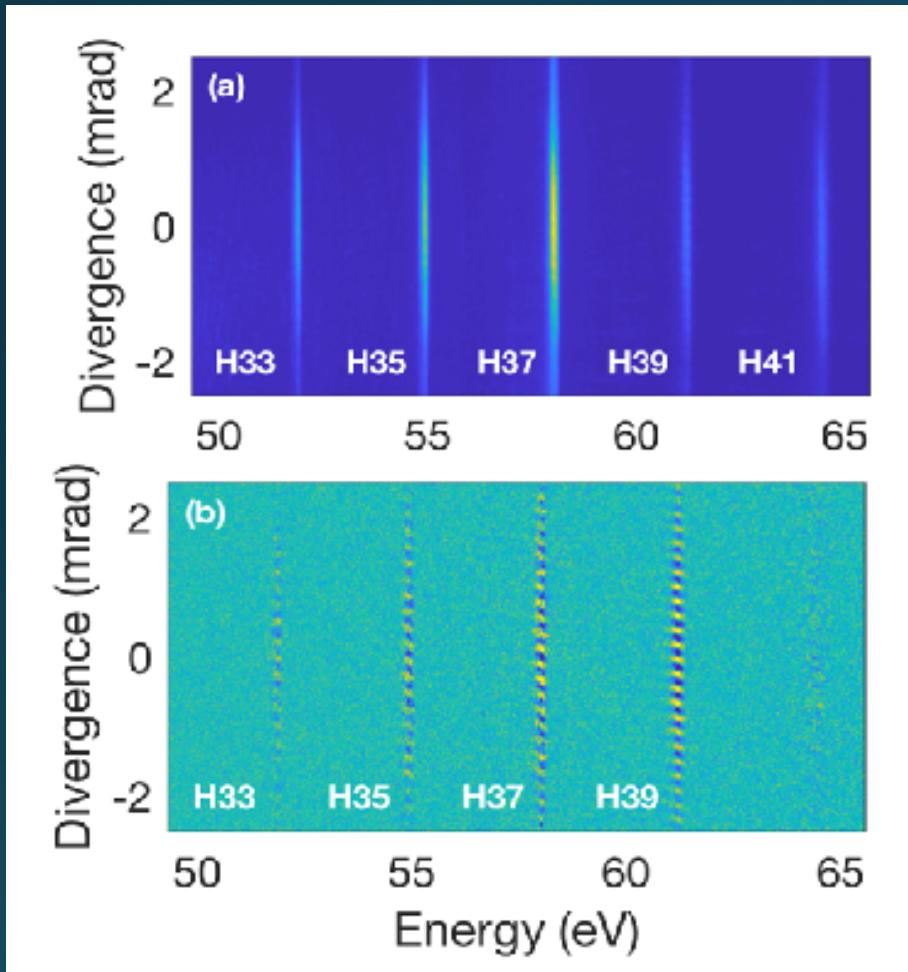


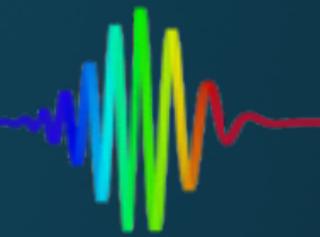
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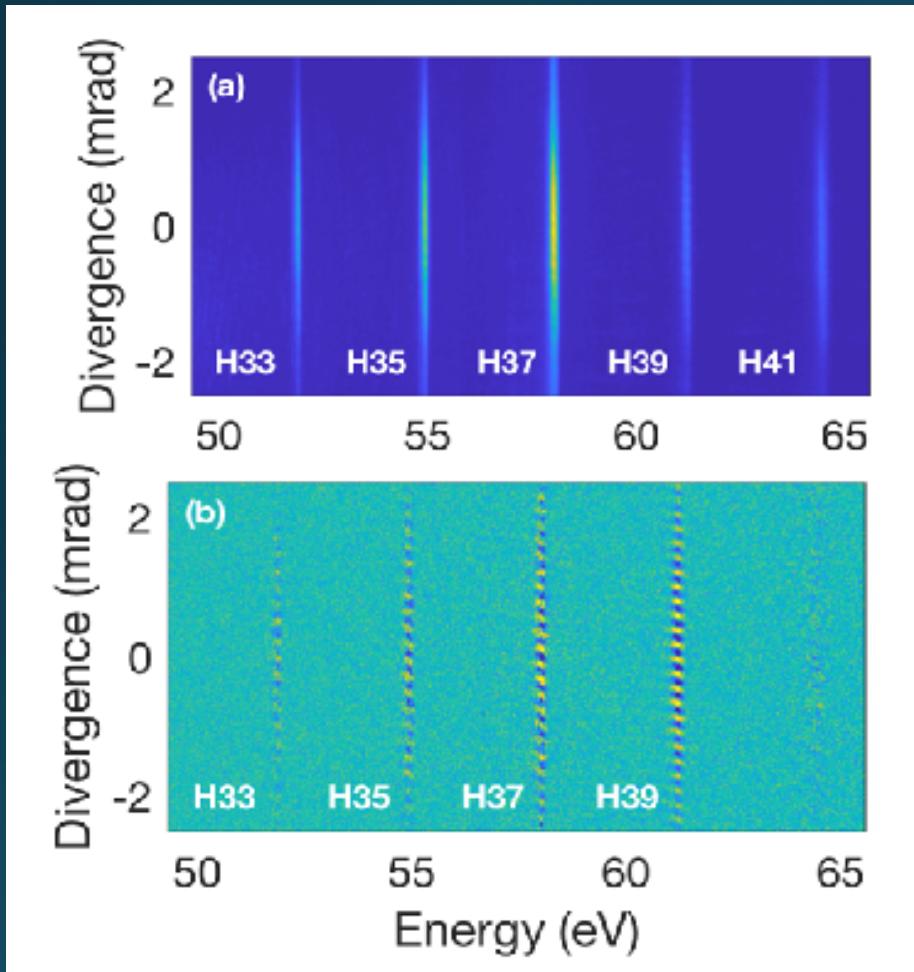


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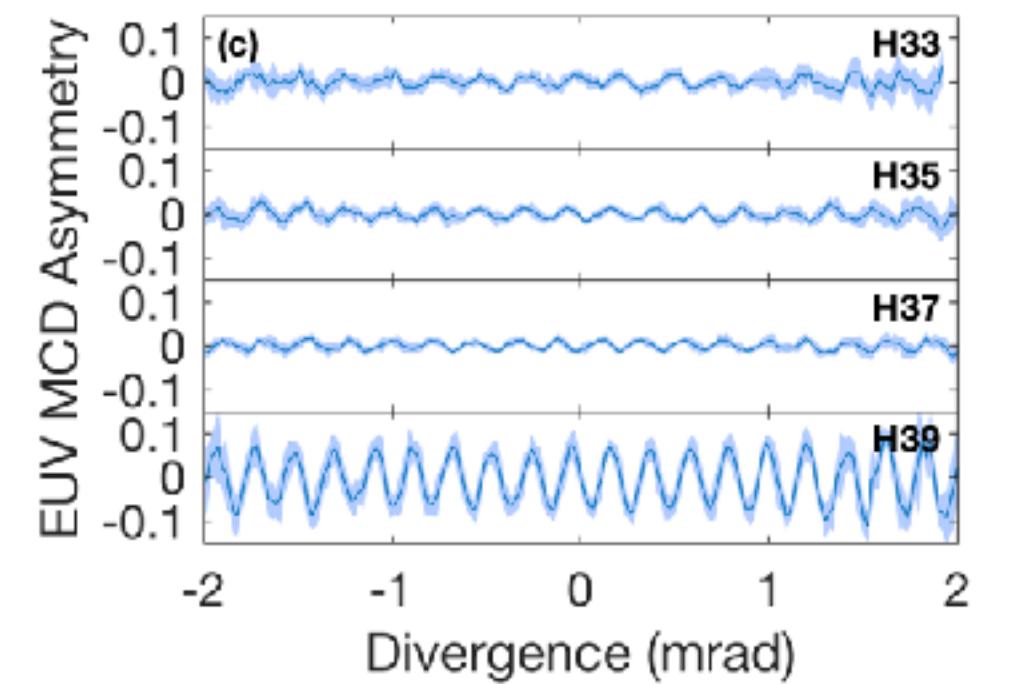


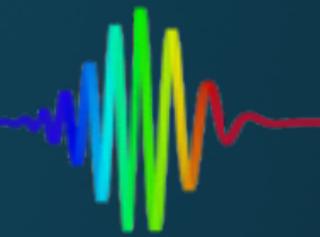


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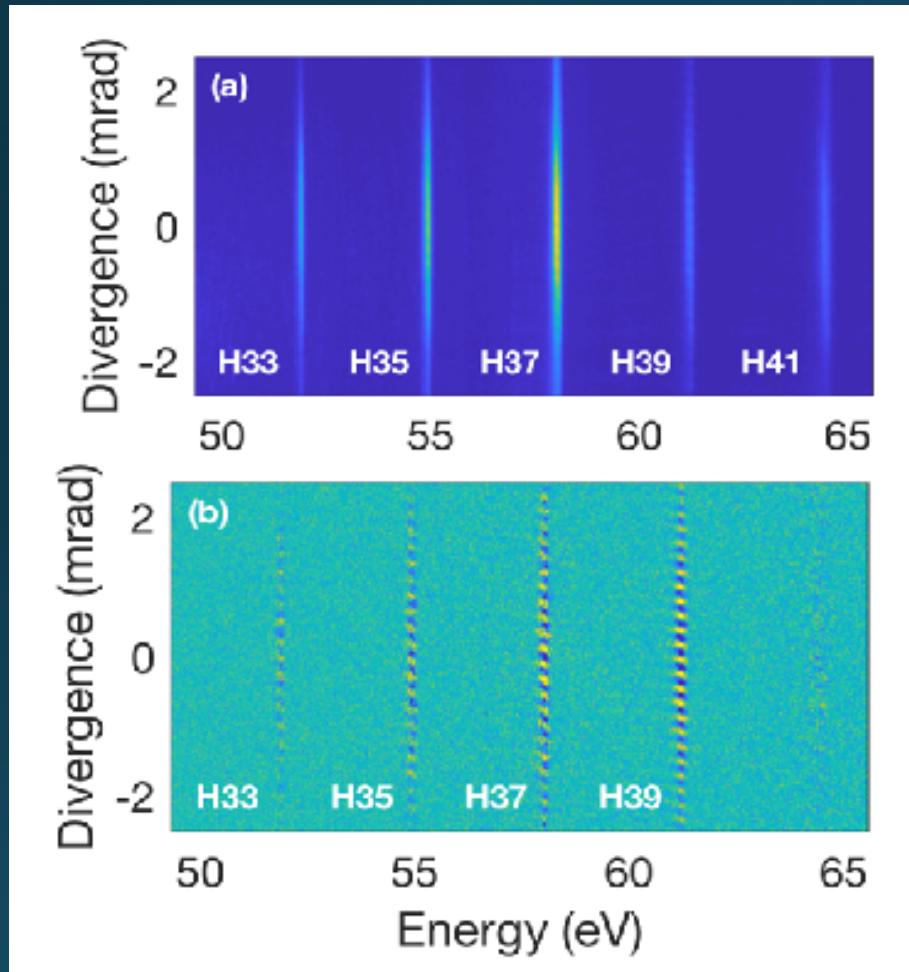


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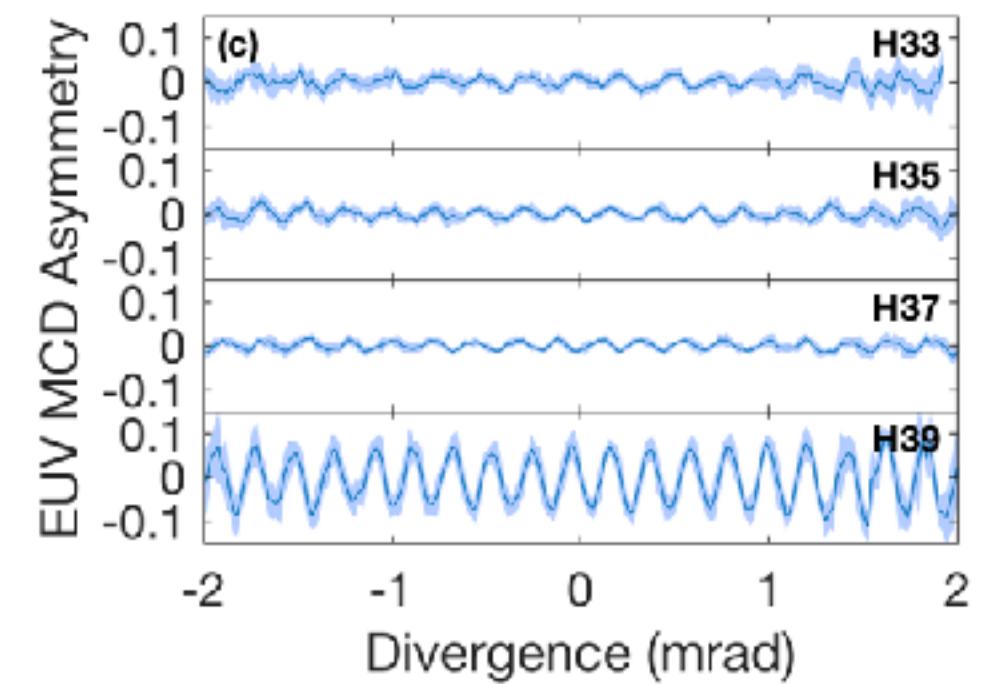




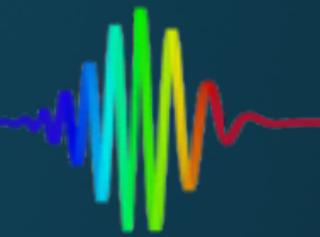
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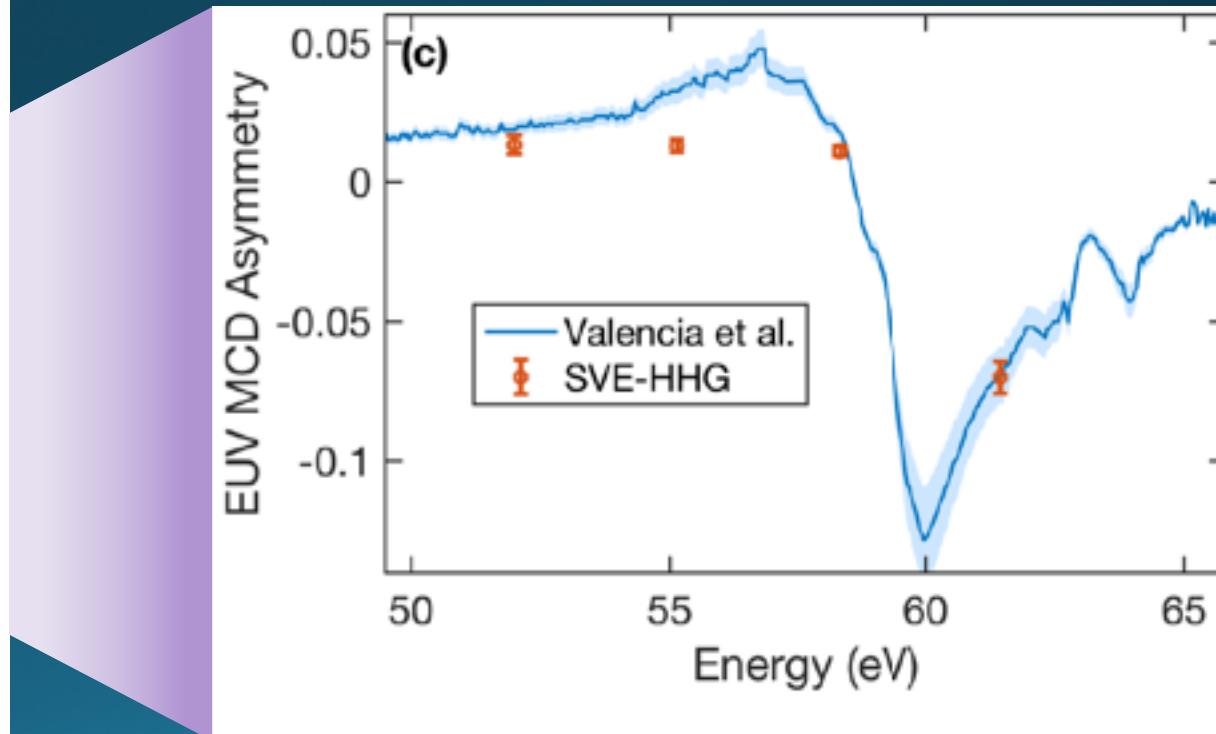
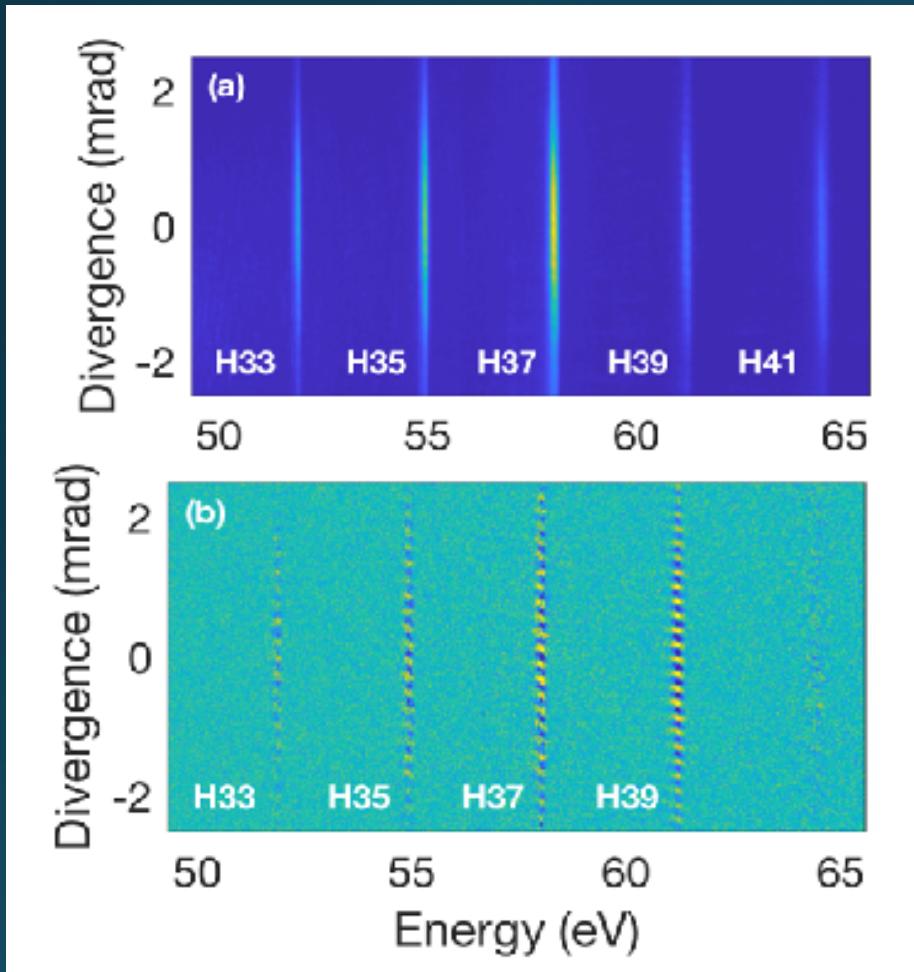
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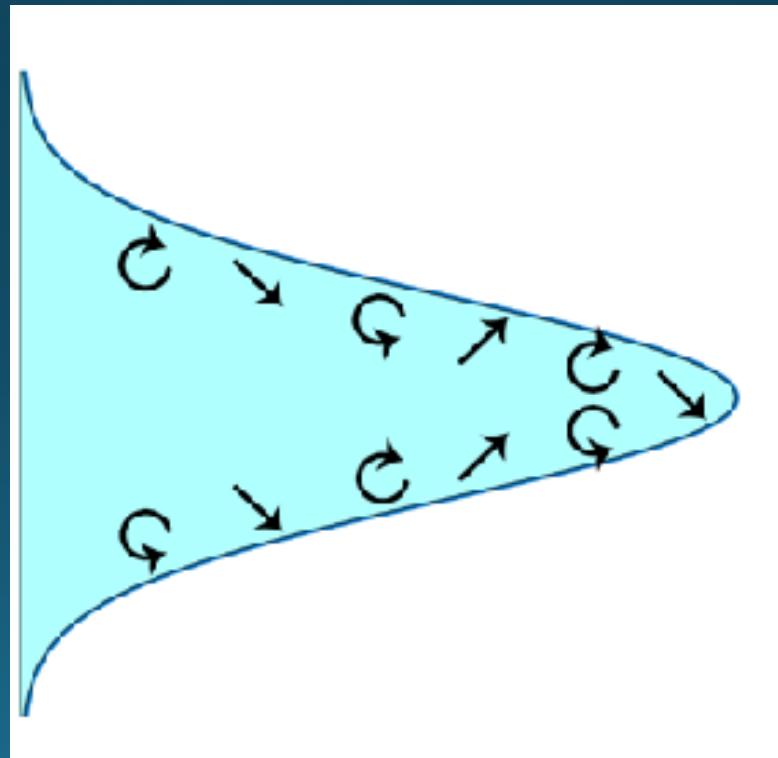
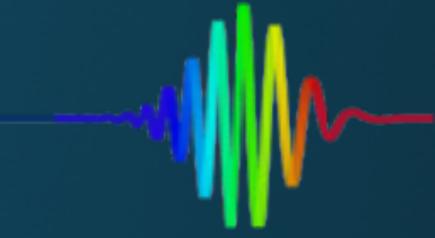
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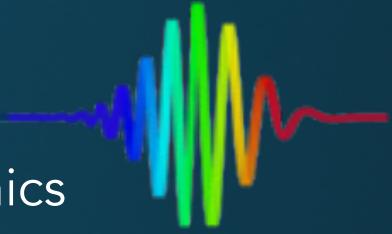
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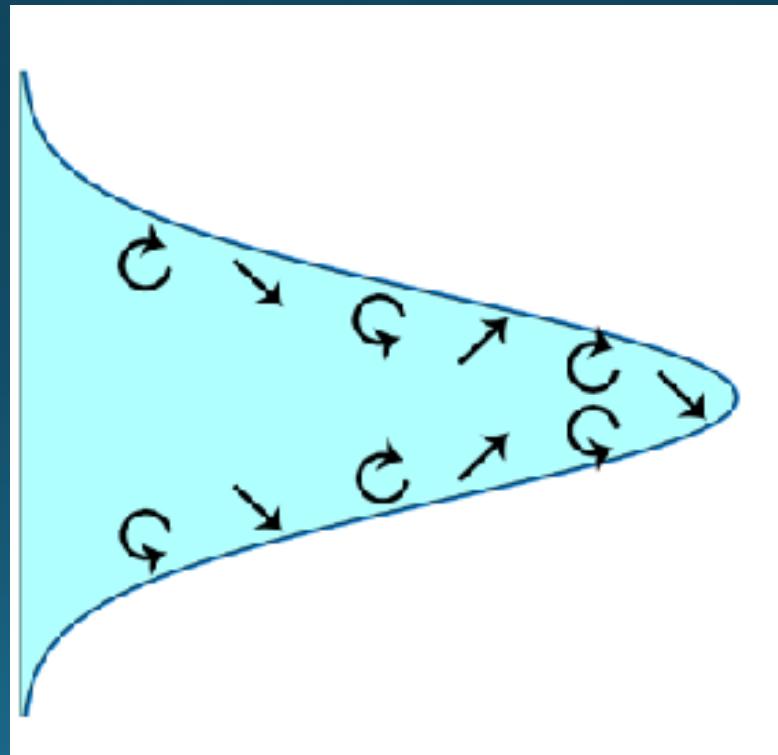
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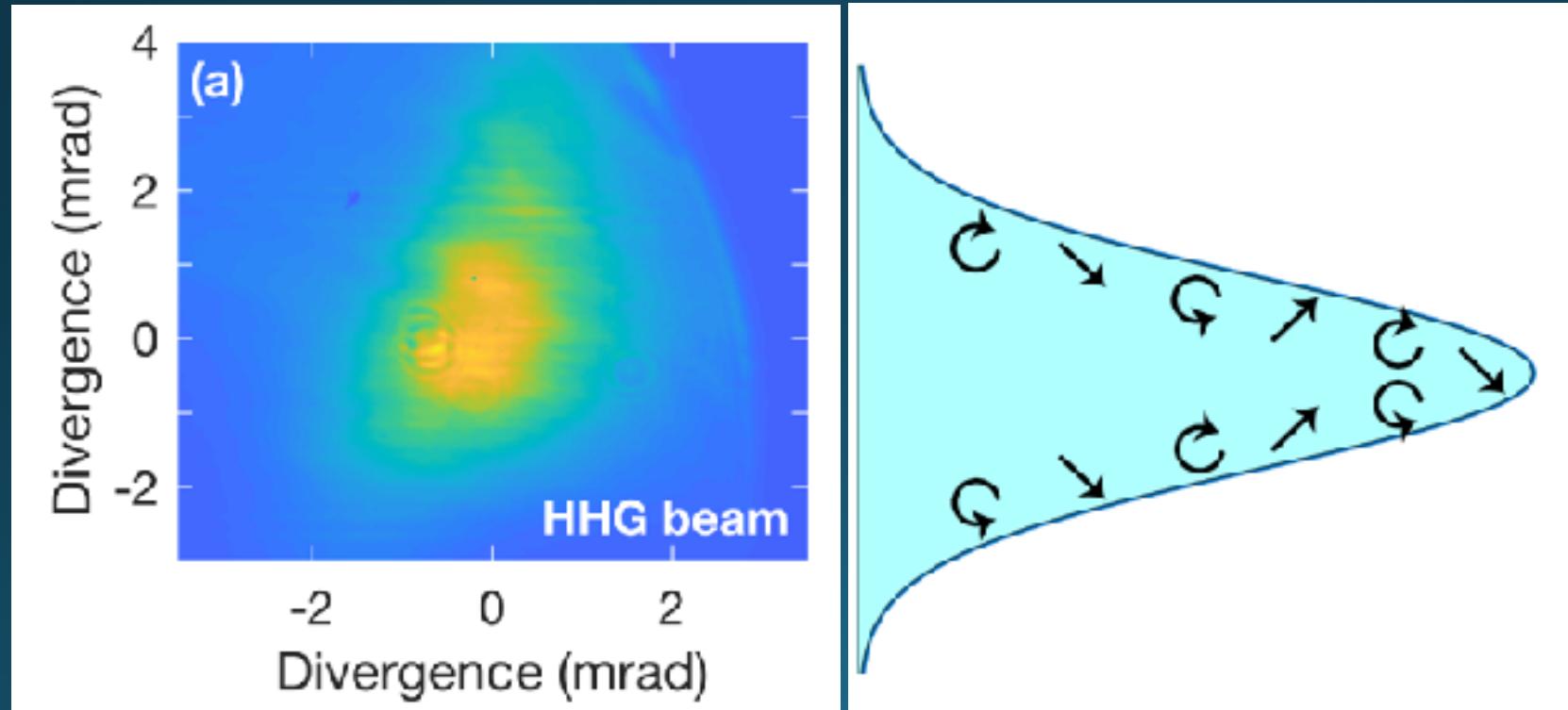


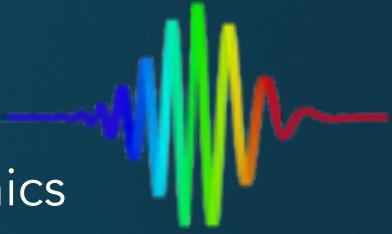
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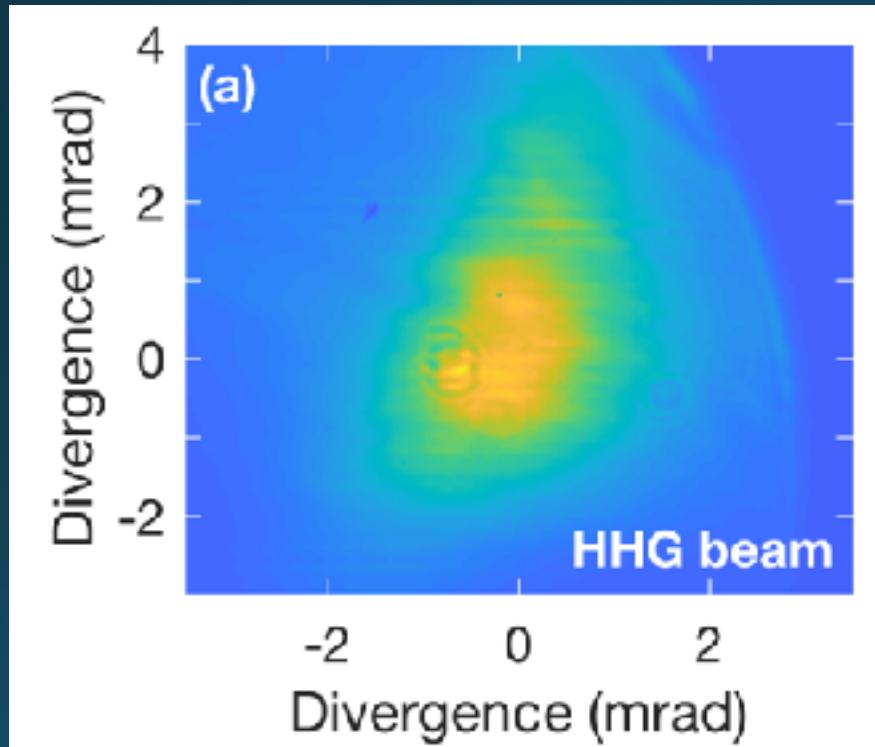


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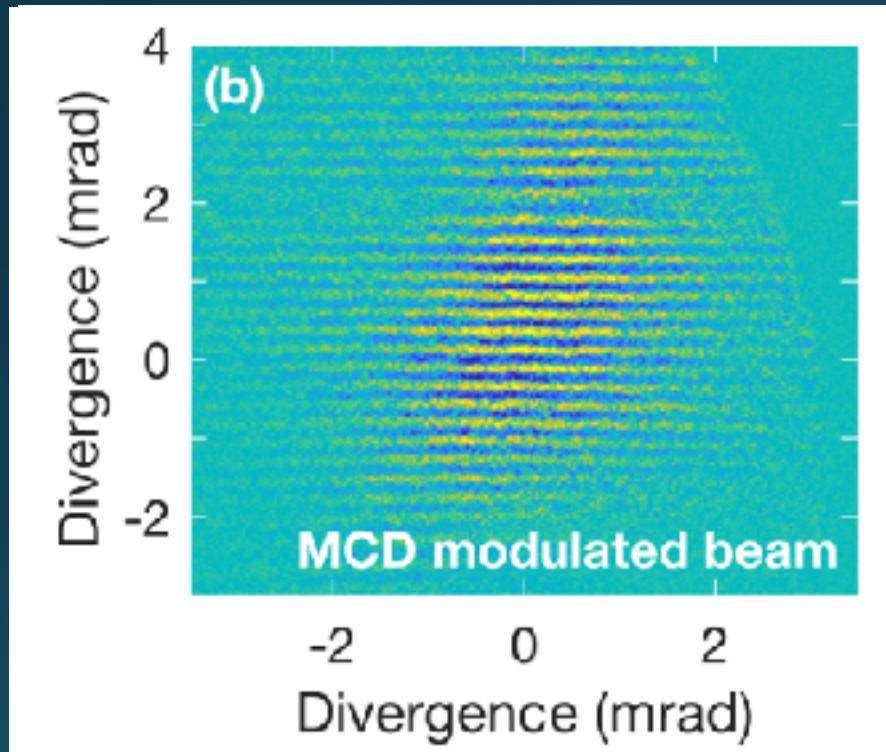
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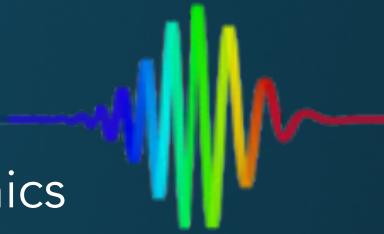
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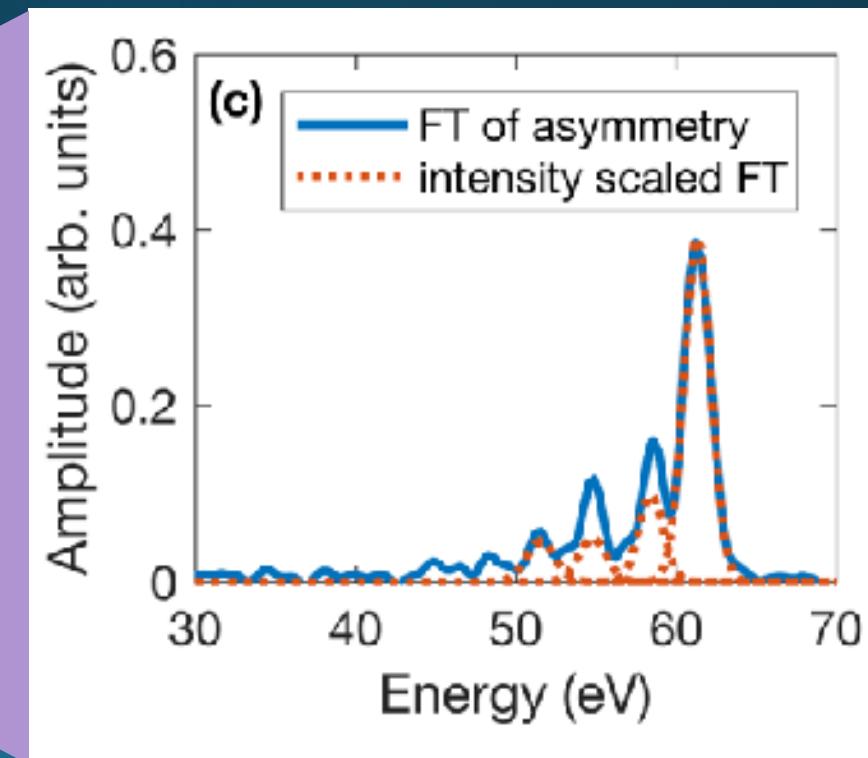
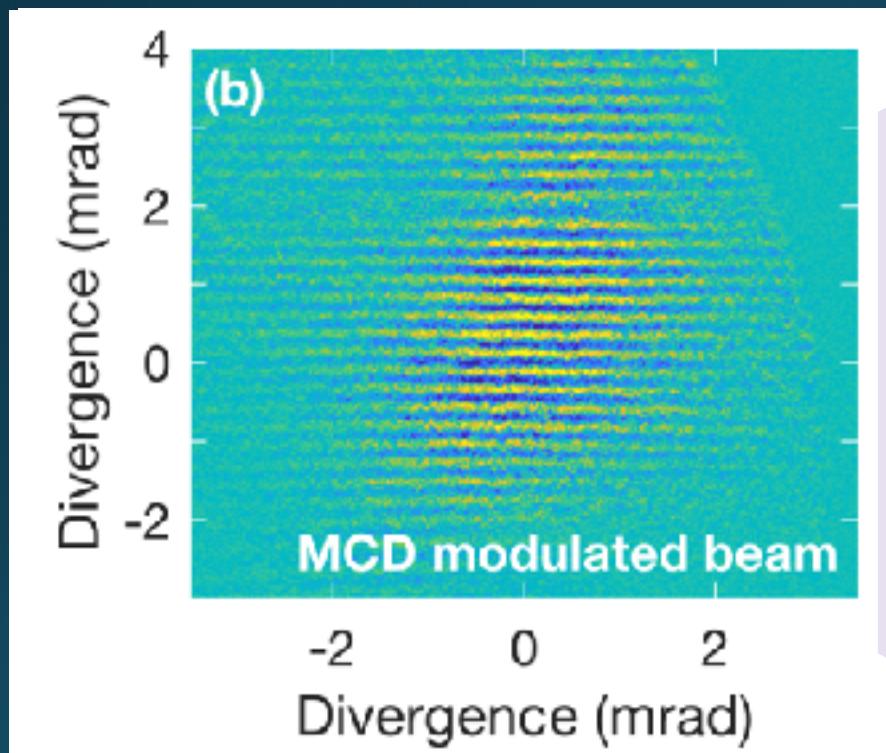
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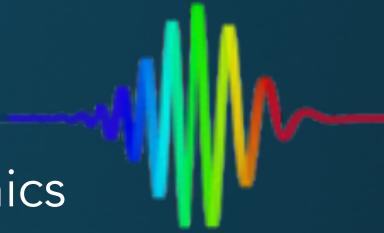
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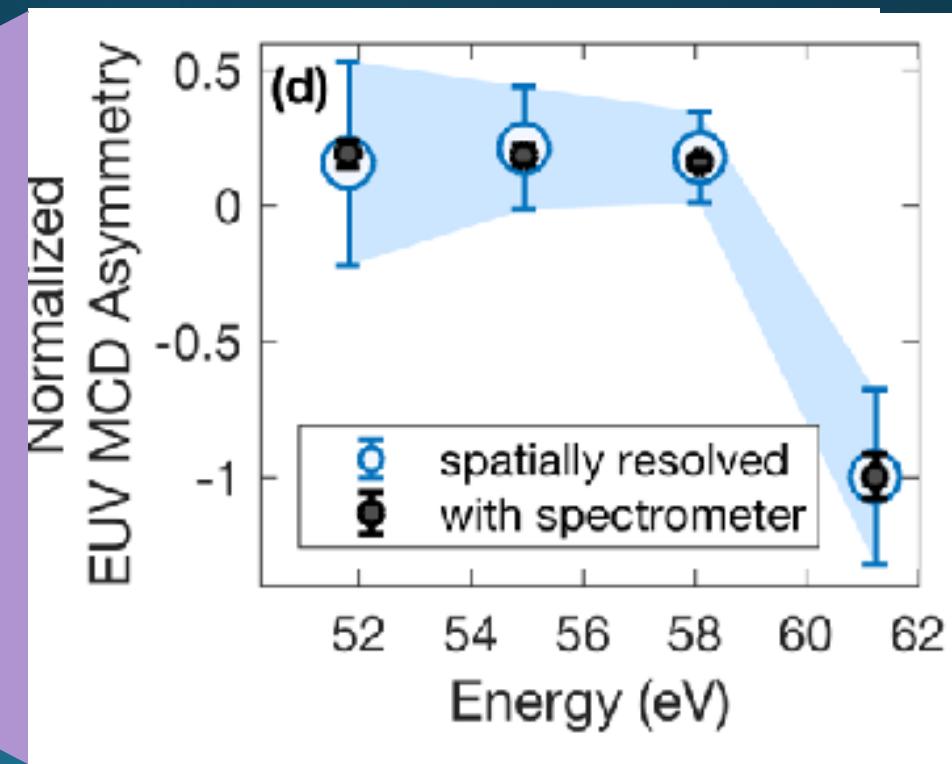
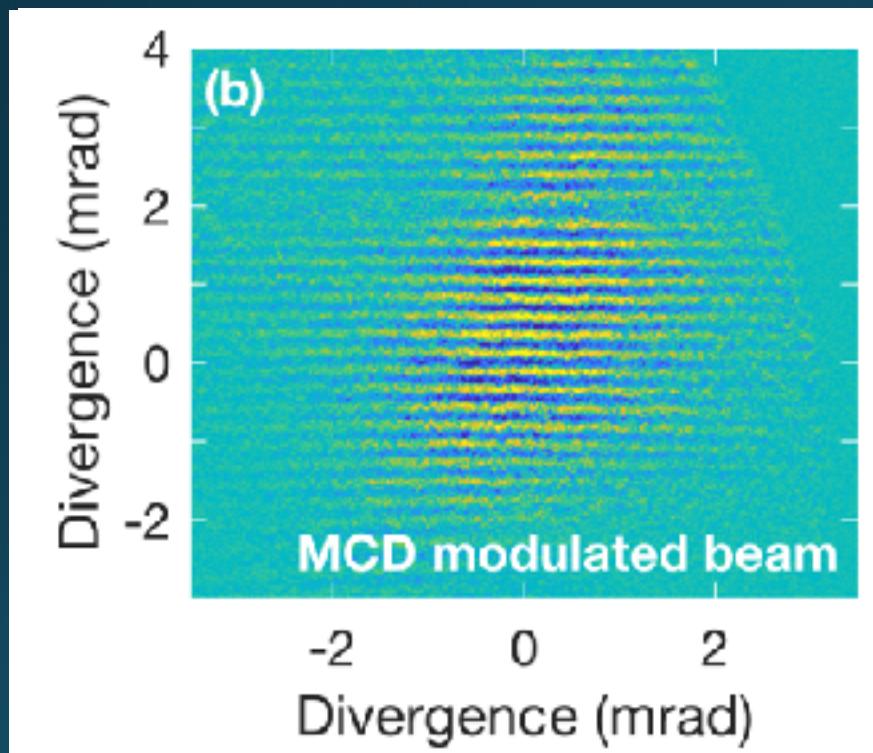
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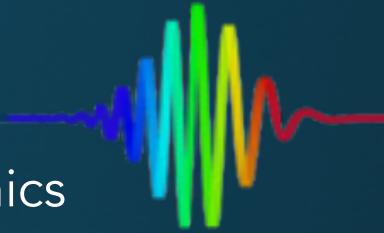


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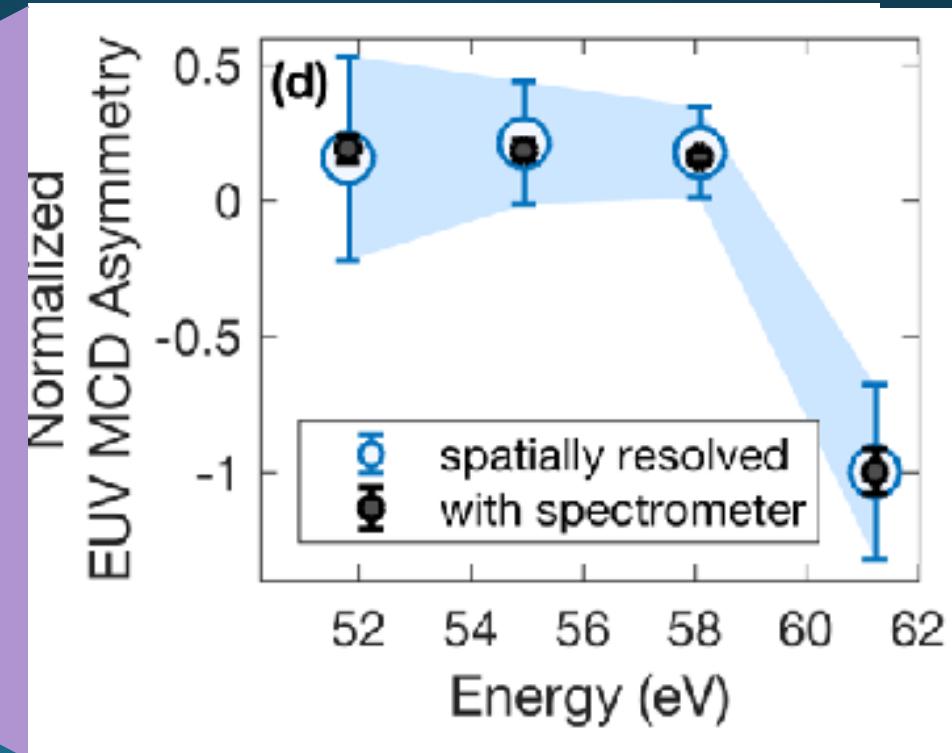
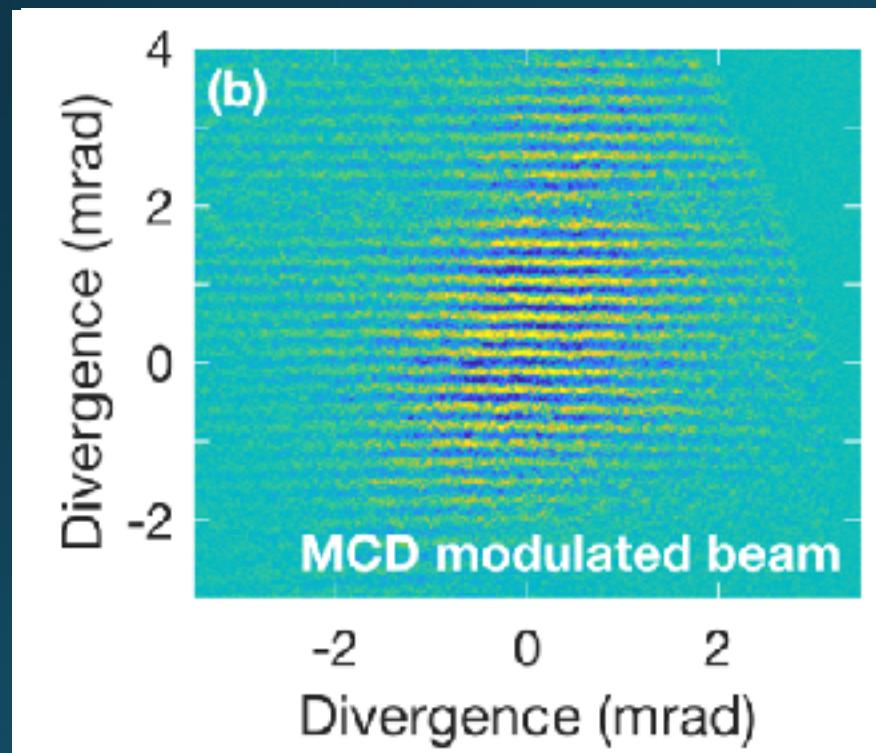
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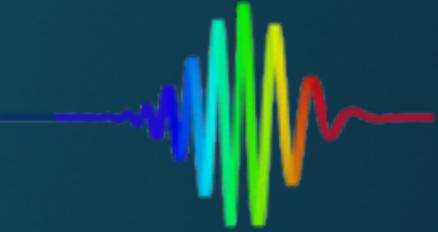
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Nearly Identical MCD Spectra from Different Samples Measured Months Apart

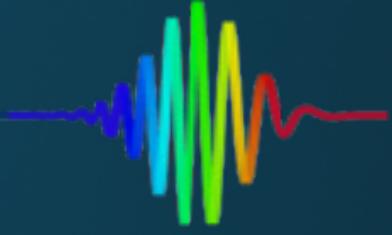




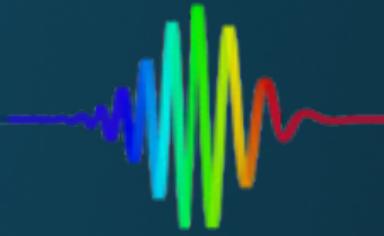
Temporal Evolution of Two-Focal Spot CPHHG: Supercontinua and Isolated, Circular Attosecond Pulses!



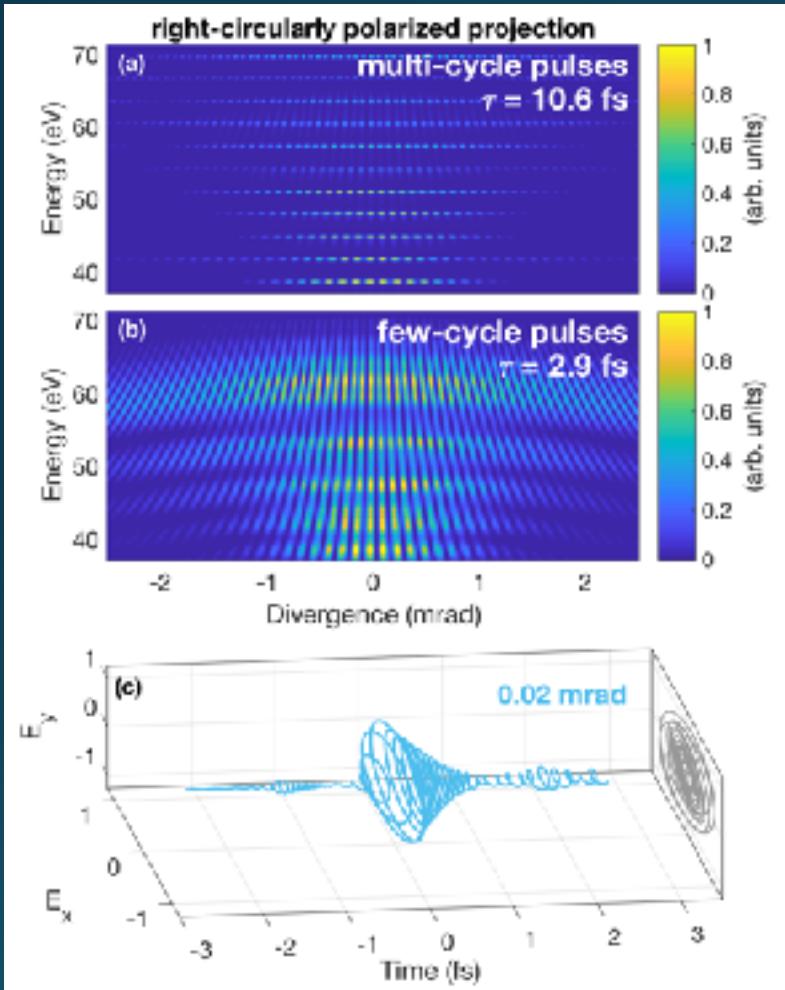
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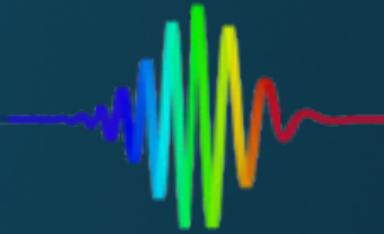
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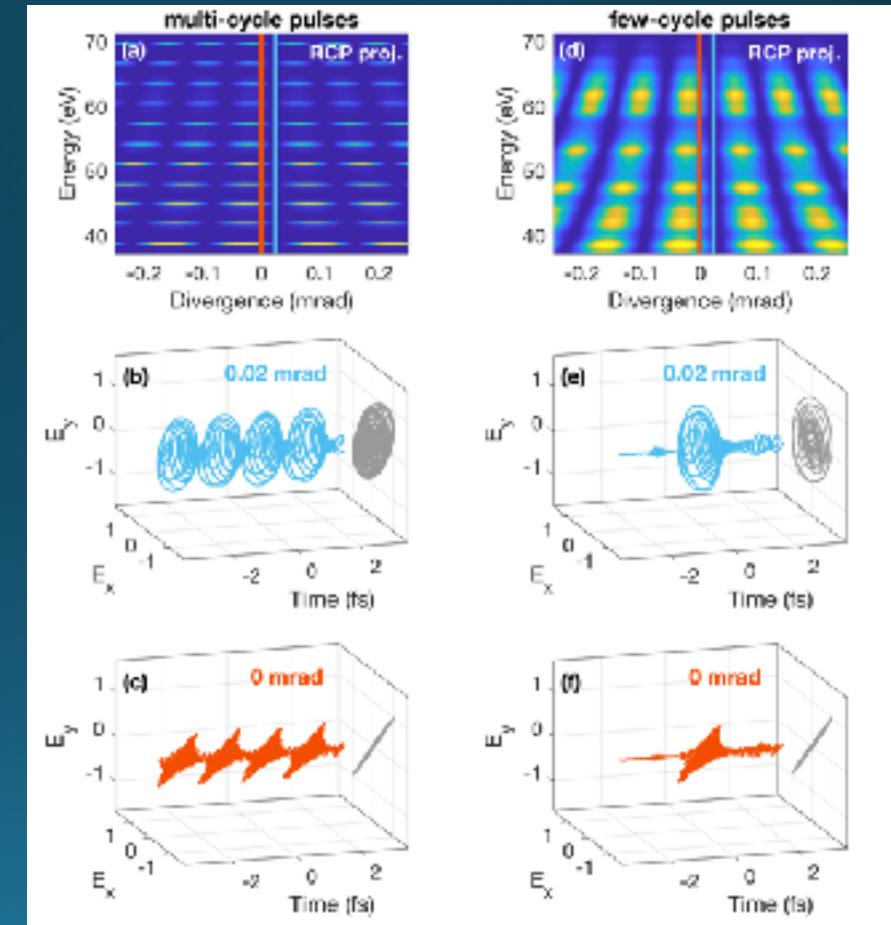
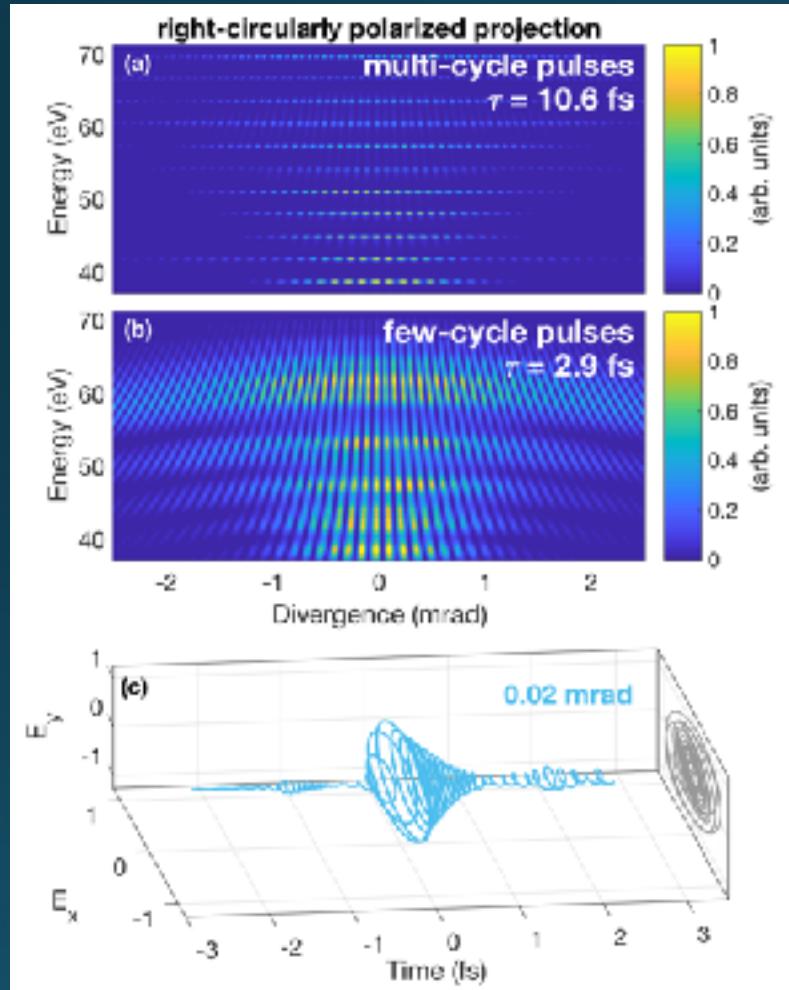
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Temporal Evolution of Two-Focal Spot CPHHG: Supercontinua and Isolated, Circular Attosecond Pulses!

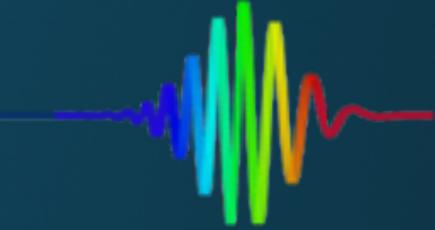


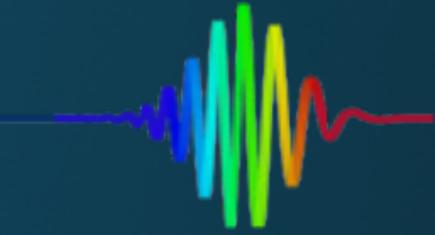
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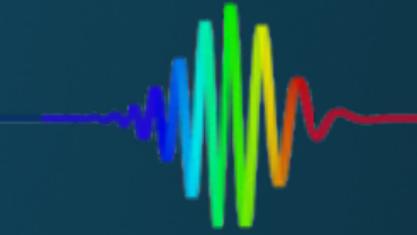


Production and Control of Elliptically Polarized Waveforms: Versatile Light Sources for Advanced Spectroscopies



➤ **Helicity-selective CPHHG via ω - 2ω amplitude control**

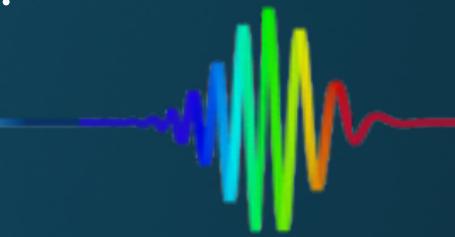
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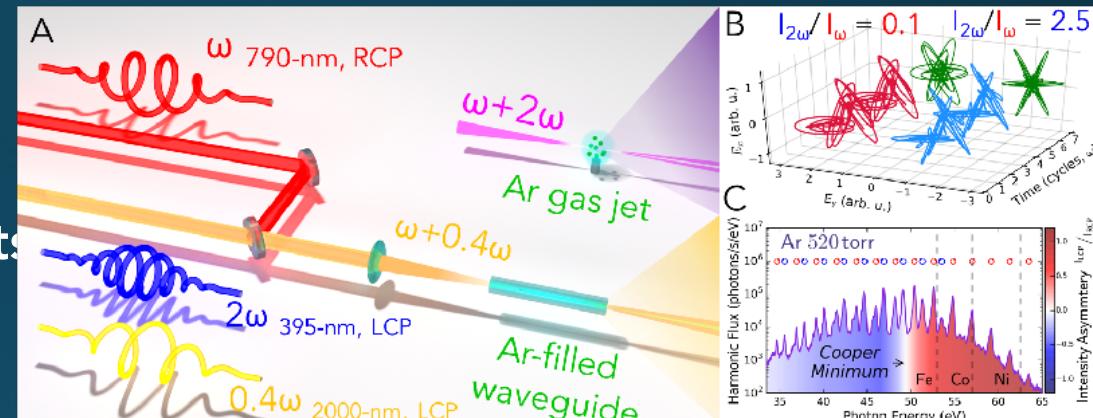


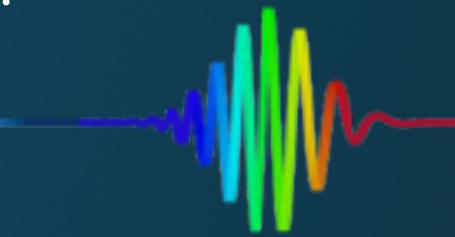
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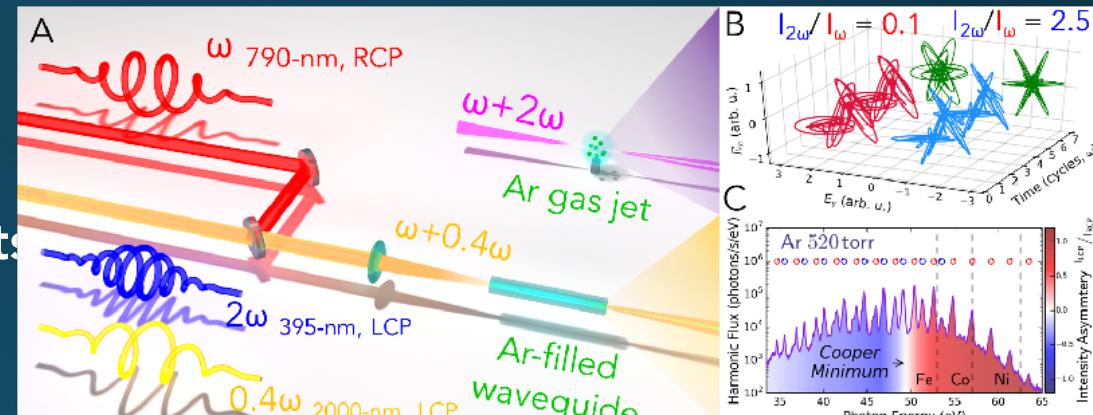
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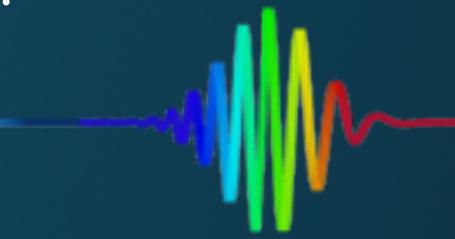
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- Microscopic physics is identical to linear, one-color HHG
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- Combined with timing information, can allow for hyperspectral imaging of chiral systems





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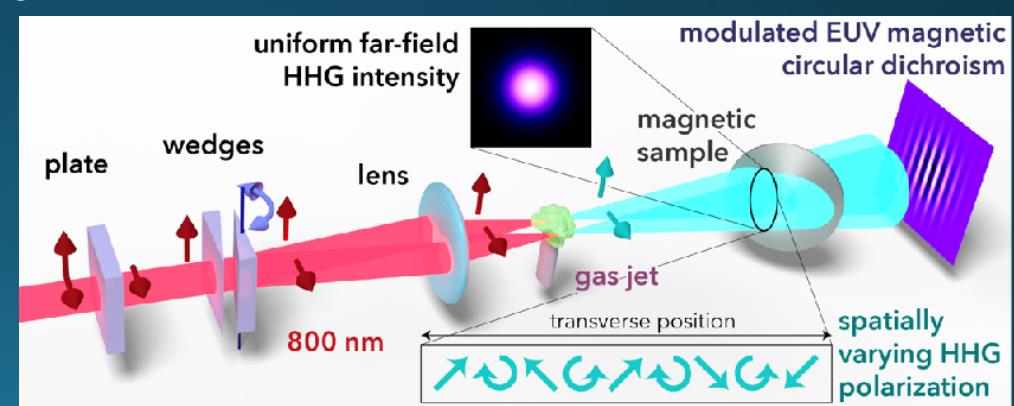
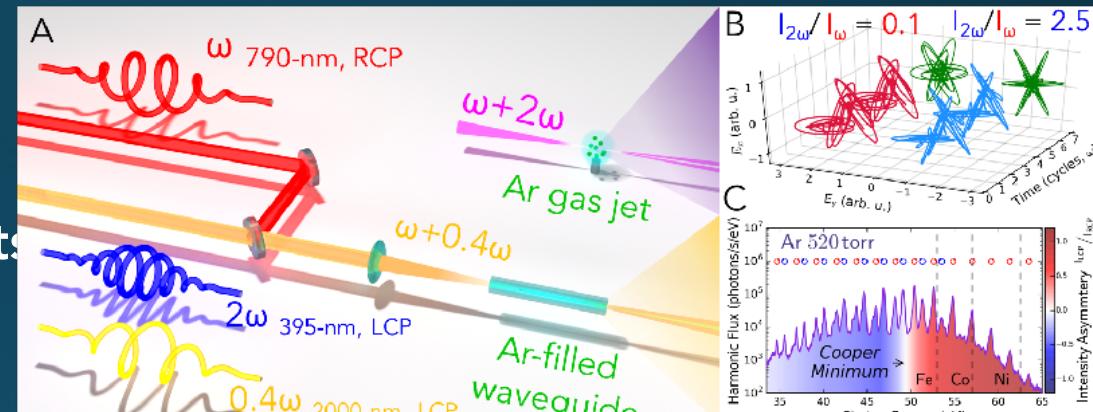
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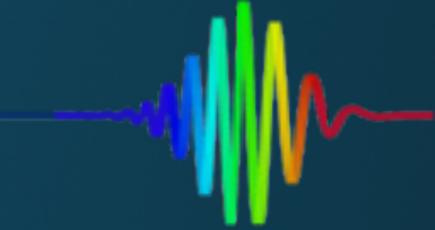
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