Controlling Quantum Electrodynamics in Circularly Polarized High Harmonic Generation: Bright, High-Energy Attosecond Waveforms with Tailored Spectro-Temporal Polarization Properties

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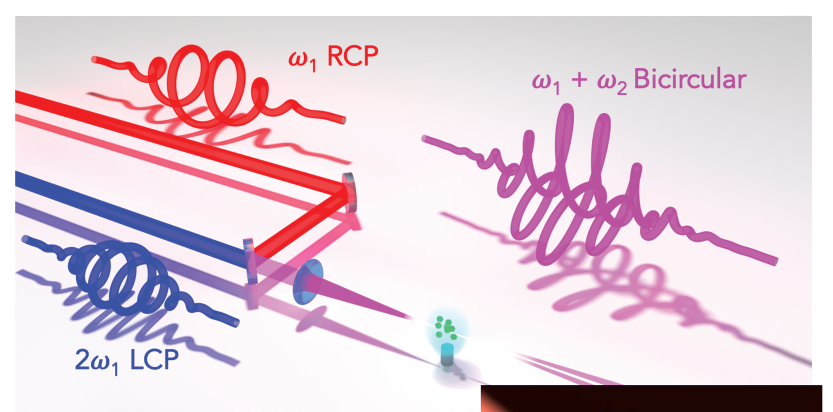
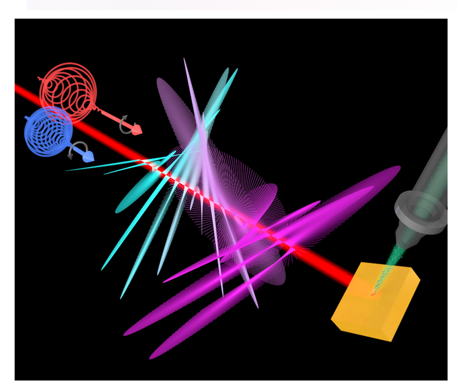
Abstract

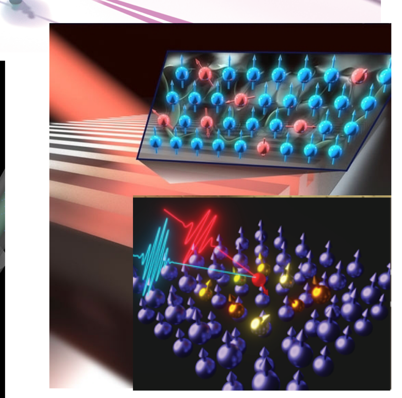
- **Motivation** - Circularly polarized high-harmonic generation (CPHHG) is a breakthrough light-science technique that yields laser-like beams of high-energy, ultrashort, circularly polarized light on a table-top scale system. Typically, CPHHG results in a comb of high-harmonics with alternating circularities, *while the attosecond pulse trains (APTs) are linearly polarized, thus precluding CPHHG-based studies of sub-fs chiral dynamics.*

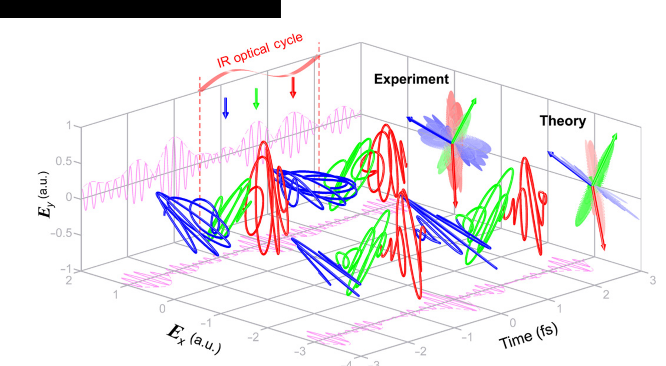
- **Experiment** - We present experimental and theoretical efforts that demonstrate active control over the quantum electrodynamics in CPHHG, *resulting in full control over the spectrotemporal polarization properties of the harmonics.*

- **Results** -
  1. The spectral helicity distribution in CPHHG can be actively controlled via the intensity ratio of the bicircular field, *yielding direct control over the polarization of the APTs.*
  2. Collective multielectron effects can be exploited in CPHHG to yield a bright harmonic spectrum composed of a single helicity, *thus generating fully circularly polarized APTs.*



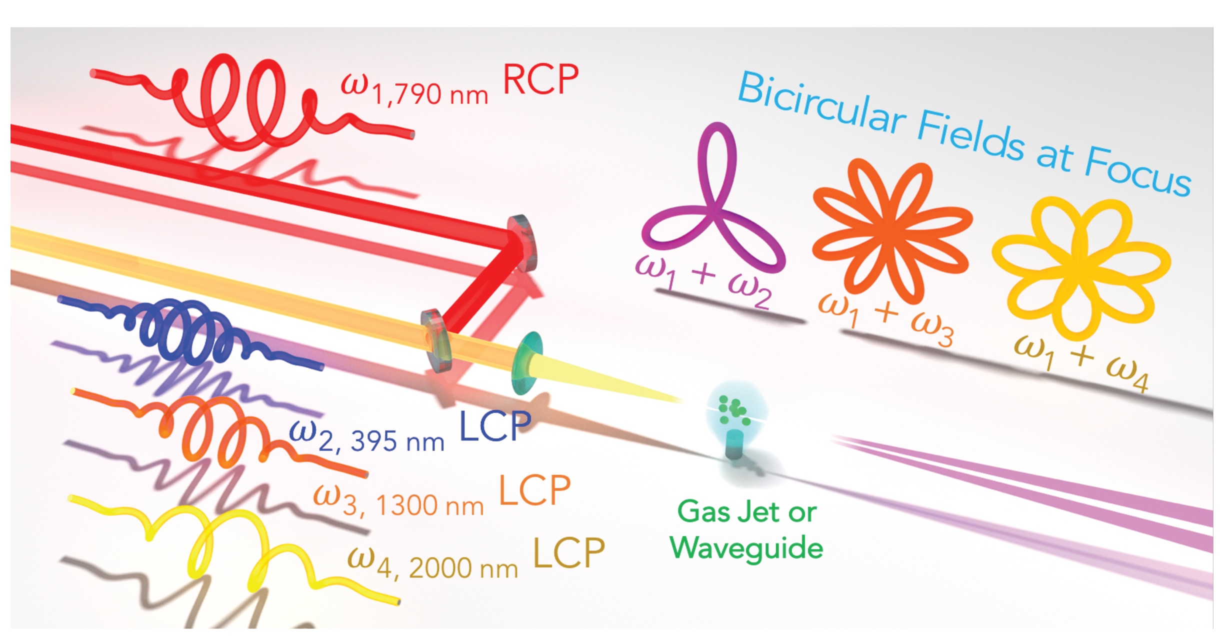




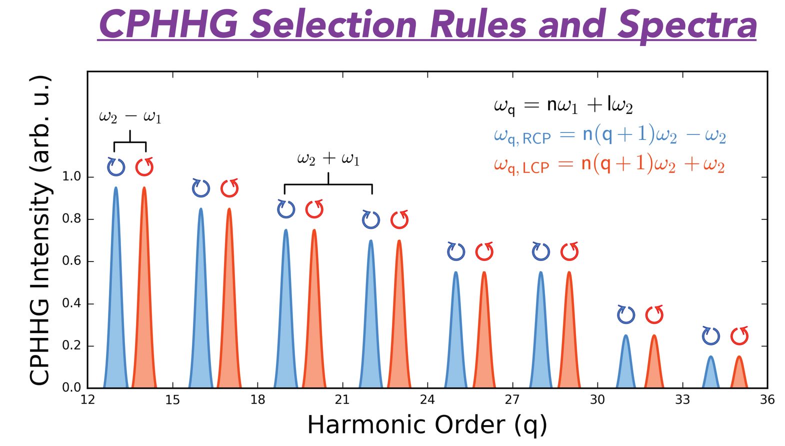


Multi-Color Bicircular Driven CPHHG

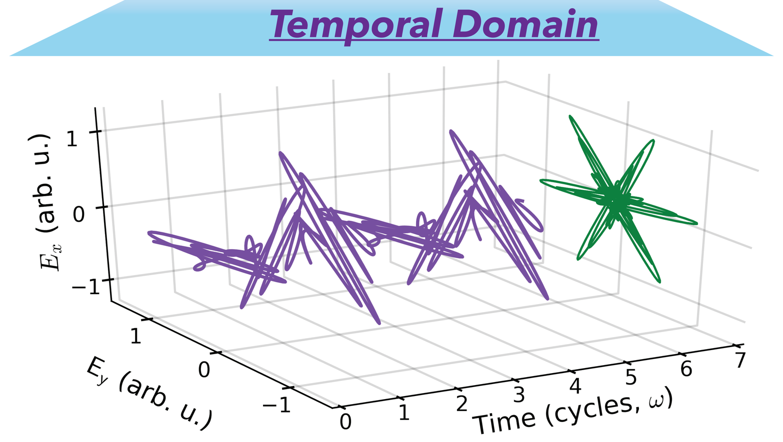
- Single-stage, high-energy Ti:Al_2O_3 amplifier (790 nm, 9 mJ, 45 fs).
- Second-harmonic generation in BBO crystal (395 nm, 4 mJ, 40 fs).
- Short-wave IR OPA (1200-2400 nm, 3.5 mJ (signal+idler), <50 fs).
- CPHHG performed in gas jet (790+395) or waveguide (790+OPA).



CPHHG Selection Rules and Spectra



Temporal Domain



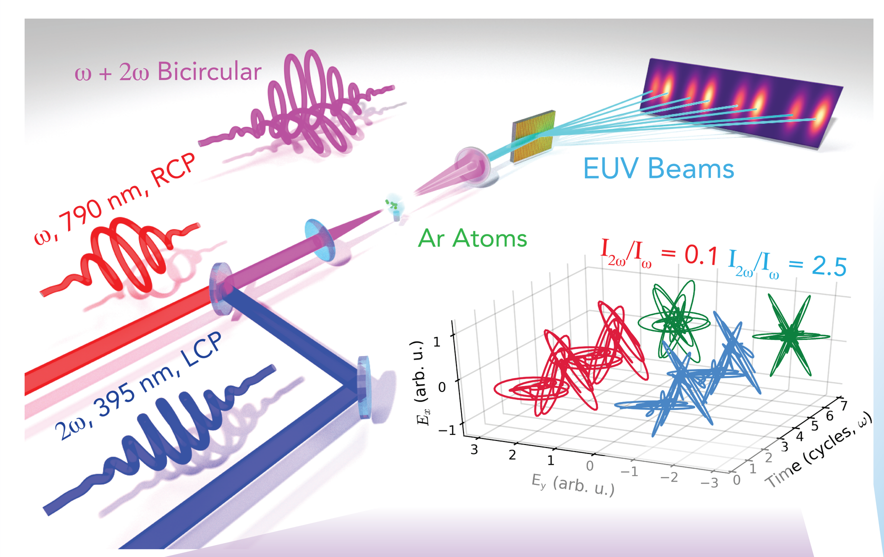
Controlling Quantum Electrodynamics in CPHHG: Custom Spectrotemporal Waveforms for Attosecond Chiral Spectroscopy

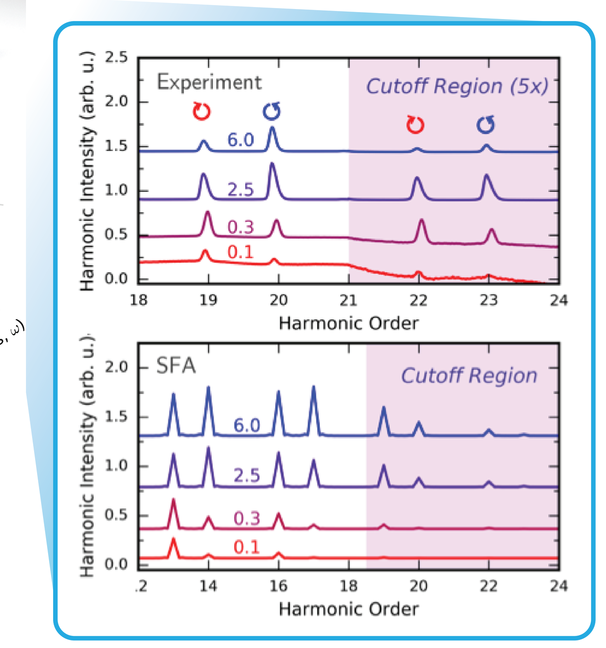
Controlling the Polarization State of Attosecond High-Harmonic Waveforms[1]

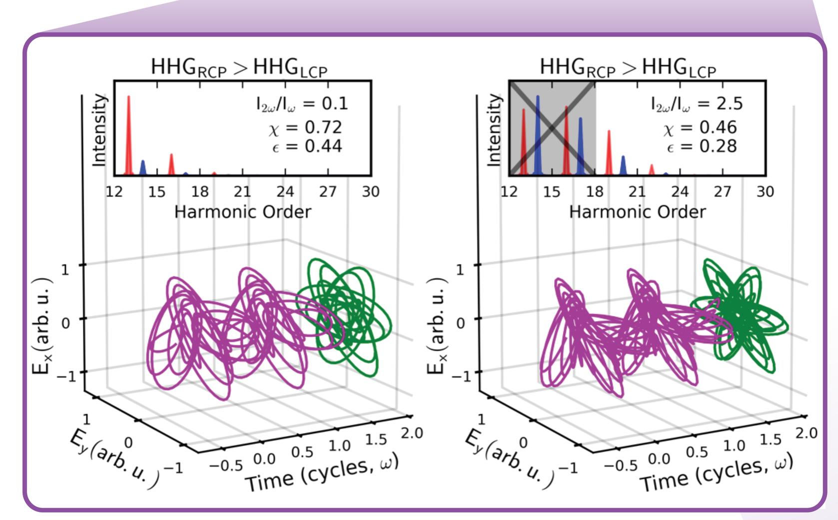
- The polarization of the underlying APTs produced via CPHHG is directly coupled to the spectral intensities of RCP and LCP harmonics.

$I_{q,RCP} \approx I_{q,LCP} \Rightarrow$ Linear APTs! $I_{q,RCP} \neq I_{q,LCP} \Rightarrow$ Elliptical APTs!

- By simply altering the intensity ratio, I_B/I_R, of the of the bicircular field, we can enhance either RCP or LCP harmonics, while still preserving their circularity!

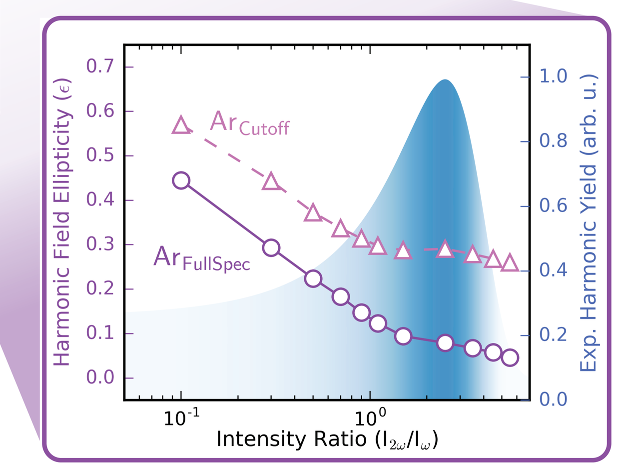






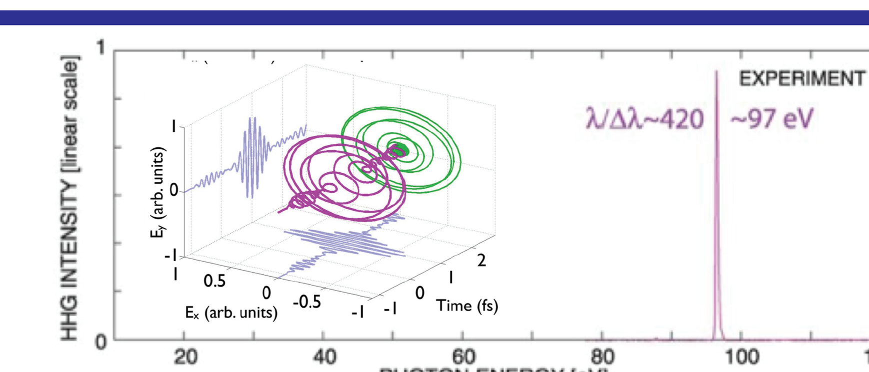
- The intensity ratio of the bicircular field allows for *arbitrary polarization control over the underlying APTs in real time.*
- Temporal gating of an individual burst in the APT can yield *isolated, circularly polarized attosecond pulses.*

APT Polarization Control Over Entire CPHHG Spectrum!



Conclusions and Outlook

- We demonstrate active control over the spectrotemporal structure of CPHHG high-harmonic waveforms, *yielding user-defined harmonic beams for ultrafast chiral spectroscopies.*
- These methodologies are *straightforward, robust, and easily integrated into existing setups.*
- Future work involves extension of these techniques to ultraviolet and mid-infrared driven CPHHG, as well as *isolating single attosecond bursts from the APTs.*



References

[1]Dorney, K. M. et al. Helicity-selective enhancement and polarization control of attosecond high-harmonic waveforms driven by bichromatic circuarly polarized laser fields. Phys. Rev. Lett. **119**, 063201 (2017).
[2]Dorney, K. M. et al. Electronic structure-induced spectrotemporal shaping of attosecond waveforms. ***In Preparation.***
[3]Schoun, S. B. Attosecond pulse shaping around a Cooper minimum. Phys. Rev. Lett. **112**, 153001 (2014).