

# Controlling Quantum Electrodynamics in Circularly Polarized High Harmonic Generation:

## Bright, High-Energy Attosecond Waveforms with Tailored Spectro-Temporal Polarization Properties



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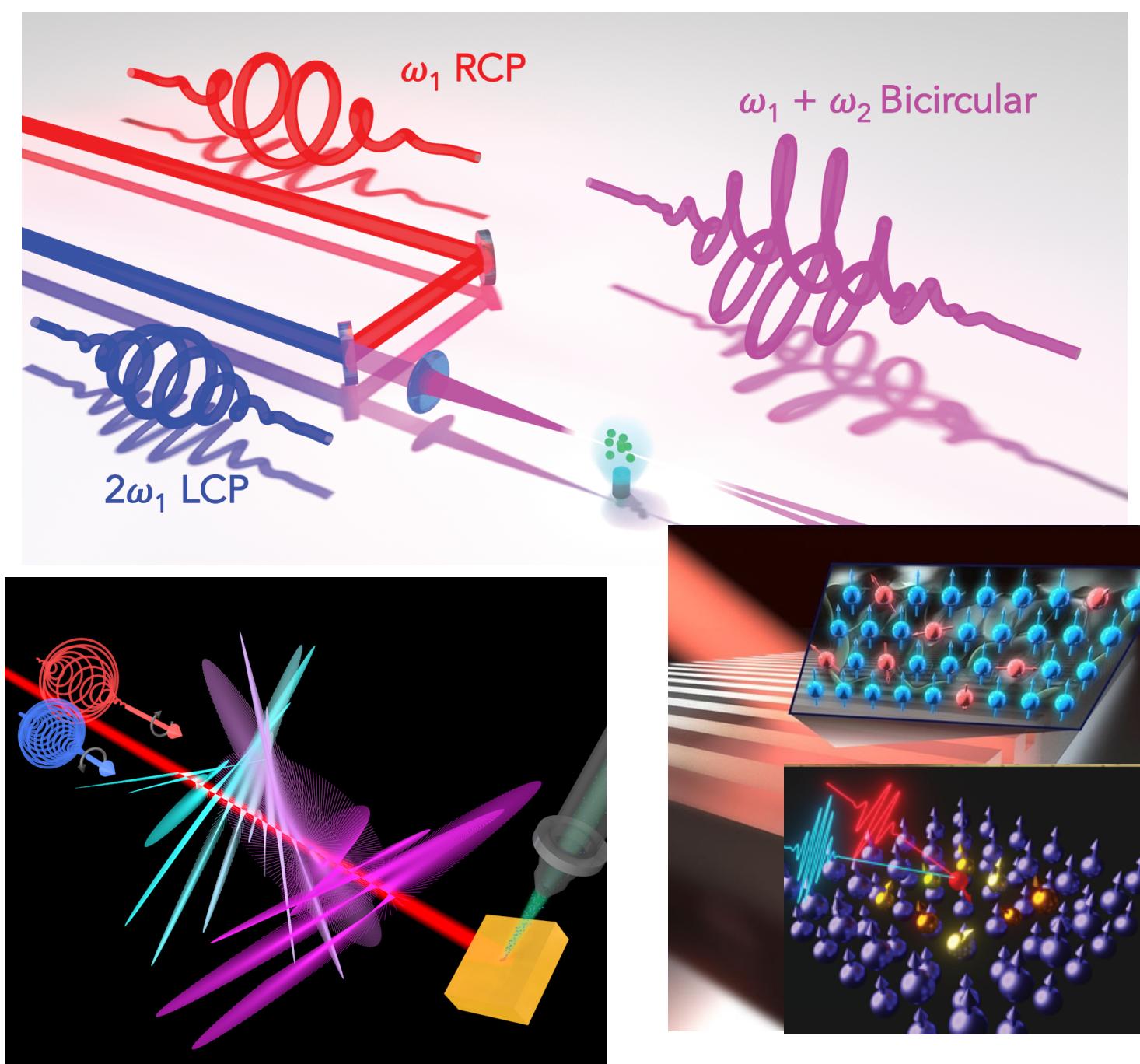
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### ABSTRACT

- 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.*



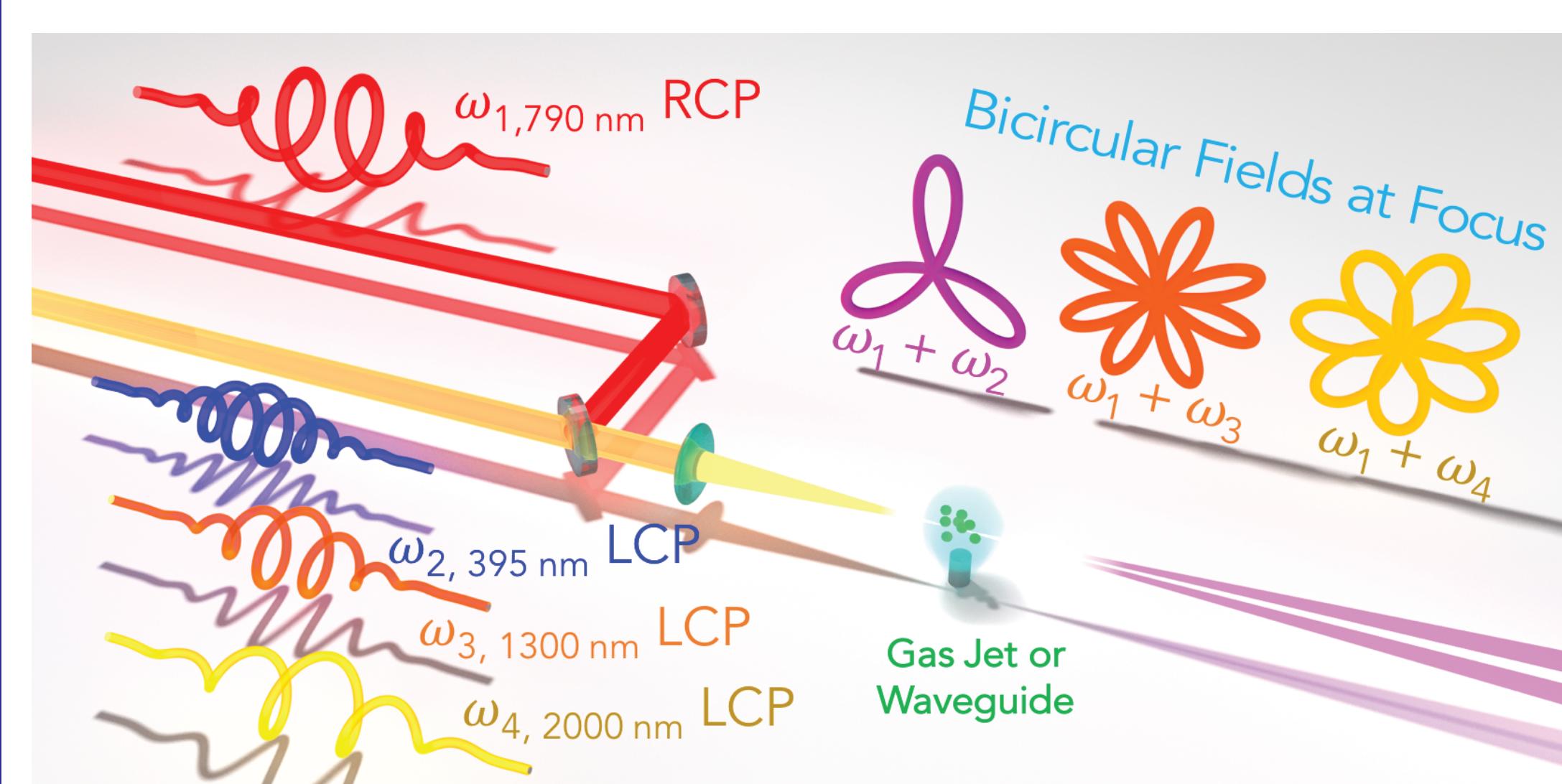
- EXPERIMENT** - We present experimental and theoretical efforts that demonstrate active control over the quantum electrodynamics in CPHHG, *resulting in full control over the spectrotemporal polarization properties of the harmonics.*

### RESULTS

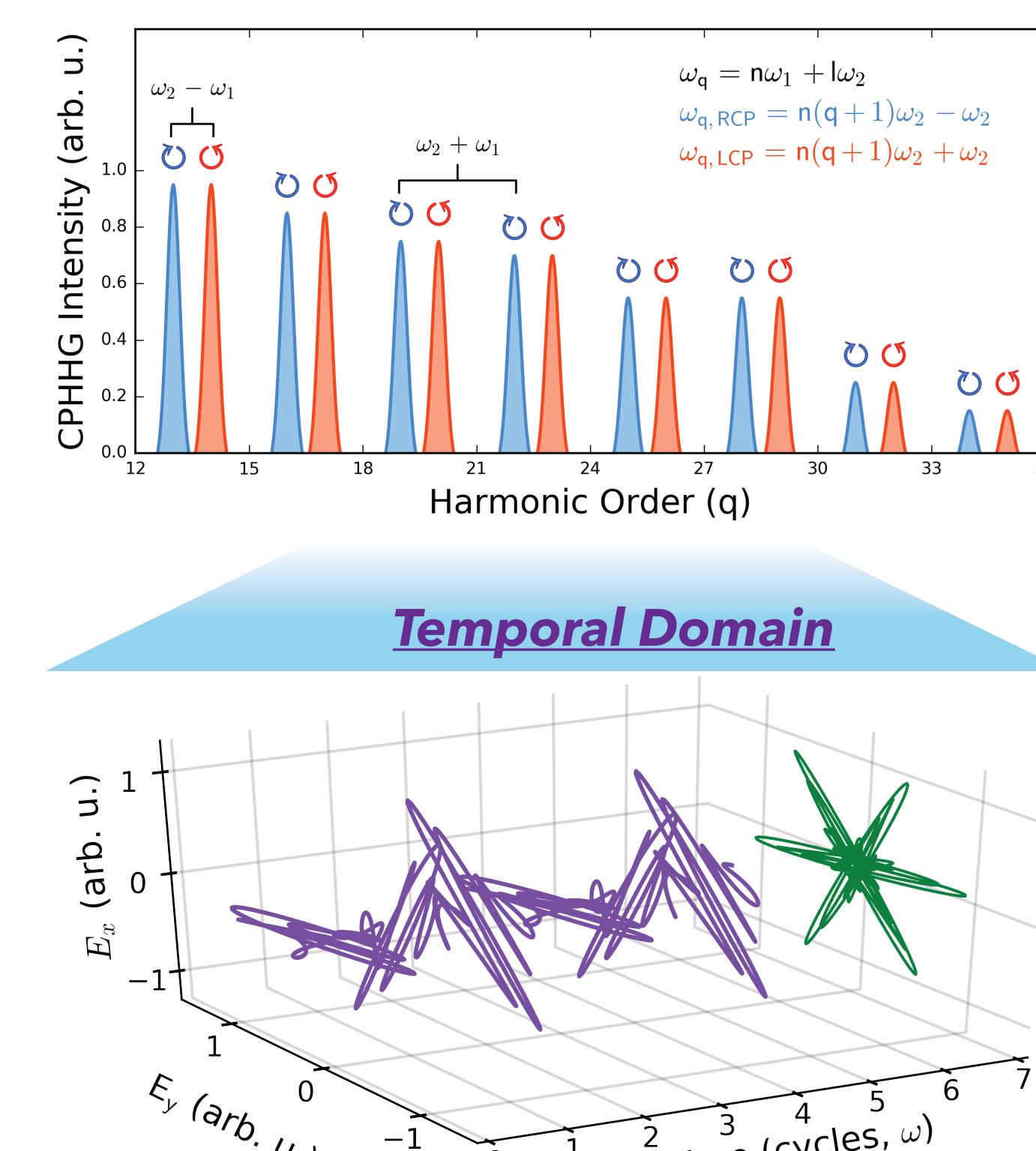
1. The spectral helicity distribution in CPHHG can be actively controlled via the intensity ratio of the bicircular field, *yielding direct control over the polarization of the APTs.*
2. Collective multielectron effects can be exploited in CPHHG to yield a bright harmonic spectrum composed of a single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

### MULTI-COLOR BICIRCULAR DRIVEN CPHHG

- Single-stage, high-energy Ti:Al<sub>2</sub>O<sub>3</sub> 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



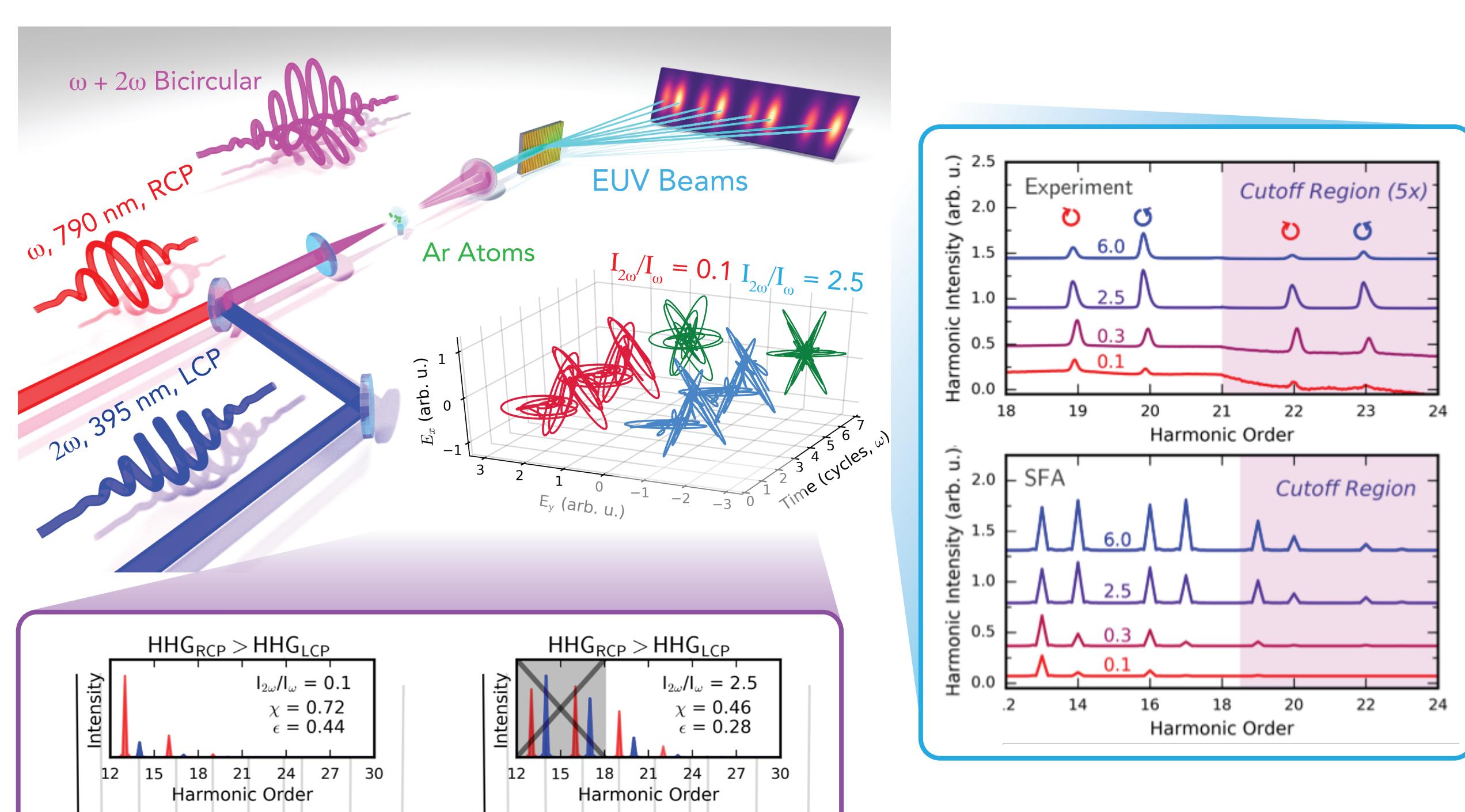
## CONTROLLING QUANTUM ELECTRODYNAMICS IN CPHHG: CUSTOM SPECTROTEMPORAL WAVEFORMS FOR ATTOSECOND CHIRAL SPECTROSCOPY

### CONTROLLING THE POLARIZATION STATE OF ATTOSECOND HIGH-HARMONIC WAVEFORMS<sup>1</sup>

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

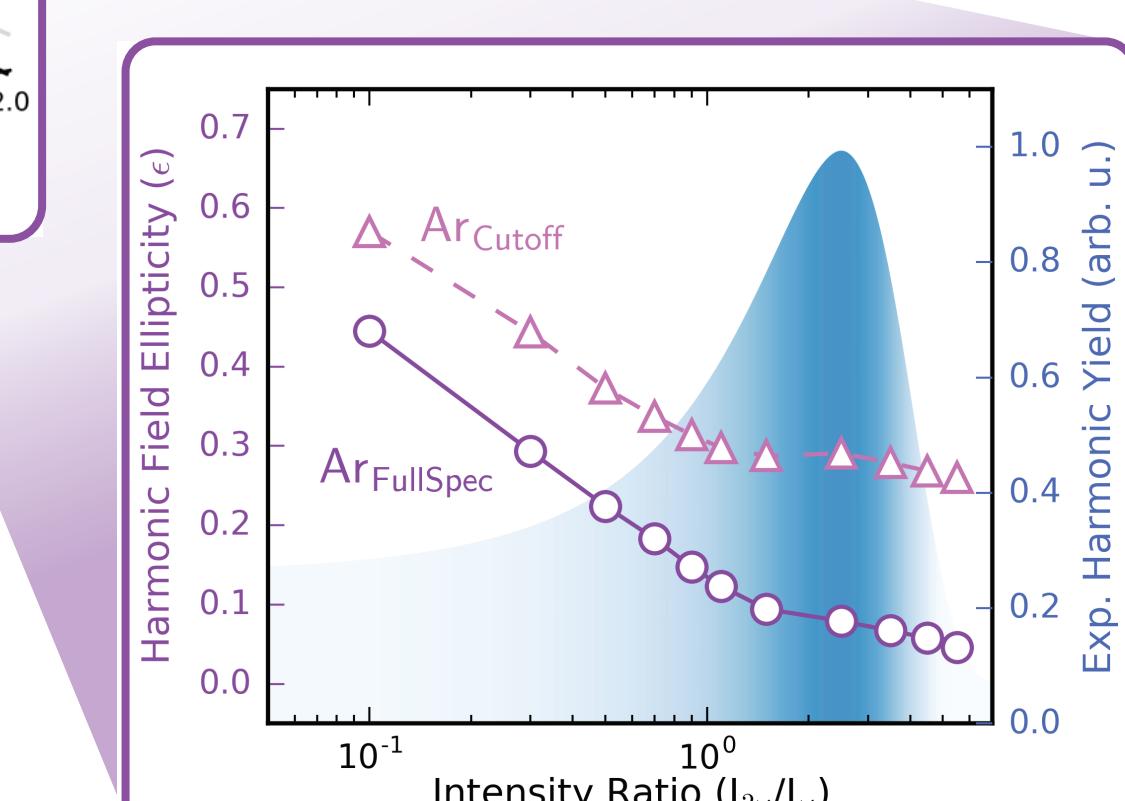
$$I_{q,RCP} \approx I_{q,LCP} \Rightarrow \text{Linear APTs!} \quad I_{q,RCP} \neq I_{q,LCP} \Rightarrow \text{Elliptical APTs!}$$

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



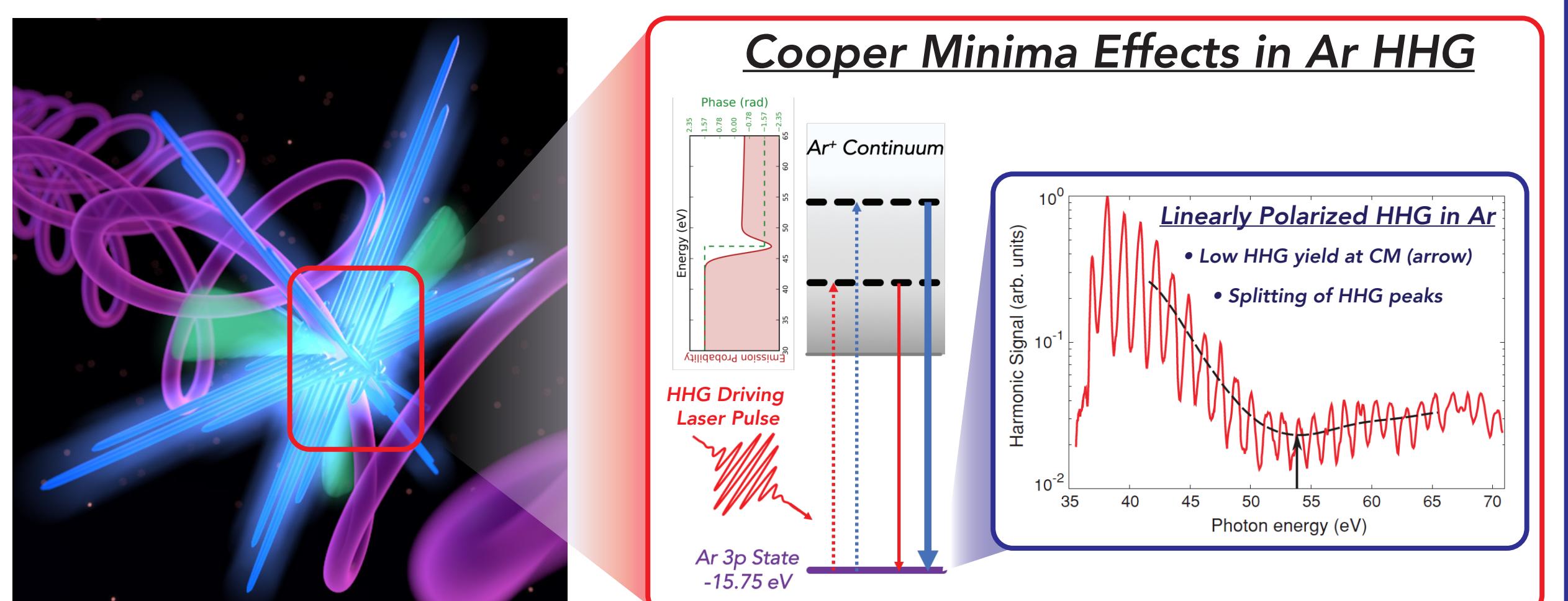
#### APT Polarization Control Over Entire CPHHG Spectrum!

- 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.*

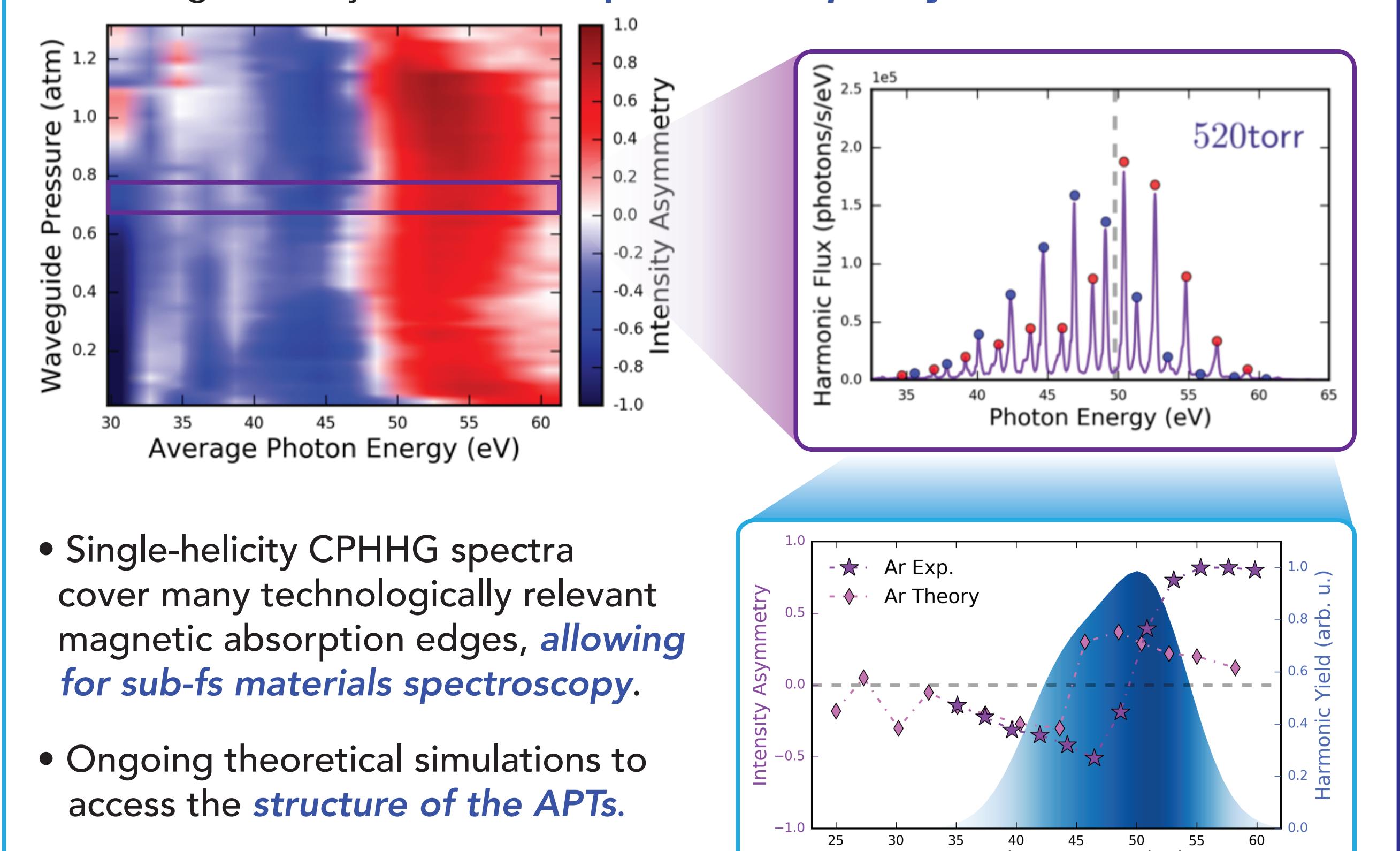


### EXPLOITING ELECTRONIC STRUCTURE TO YIELD SINGLE HELICITY ATTOSECOND PULSE TRAINS<sup>2</sup>

- Formally, CPHHG emission is the result of bound-free dipole transitions from laser-dressed continuum states to the ground state of the parent ion (e.g. Ar<sup>+</sup>).
- Electronic structure effects, such as a Cooper Minimum (CM), can be exploited to reshape the spectrotemporal structure of harmonics in a natural way.<sup>3</sup>



- Under optimal conditions, the CM effect can be exploited to yield harmonics of a single helicity, *which correspond to completely circular APTs!*



### REFERENCES

<sup>1</sup>Dorney, K. M. et al. Helicity-selective enhancement and polarization control of attosecond high-harmonic waveforms driven by bichromatic circularly polarized laser fields. Phys. Rev. Lett. **119**, 063201 (2017).

<sup>2</sup>Dorney, K. M. et al. Electronic structure-induced spectrotemporal shaping of attosecond waveforms. *In Preparation.*

<sup>3</sup>Schoun, S. B. Attosecond pulse shaping around a Cooper minimum. Phys. Rev. Lett. **112**, 153001 (2014).

### 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.*

