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

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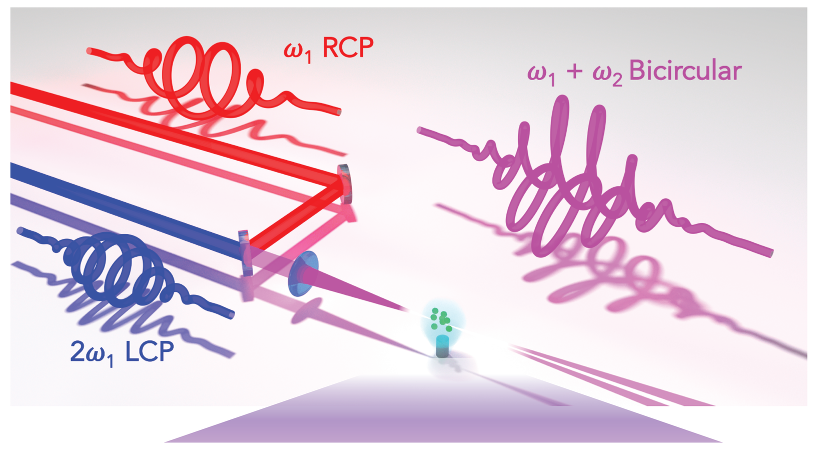
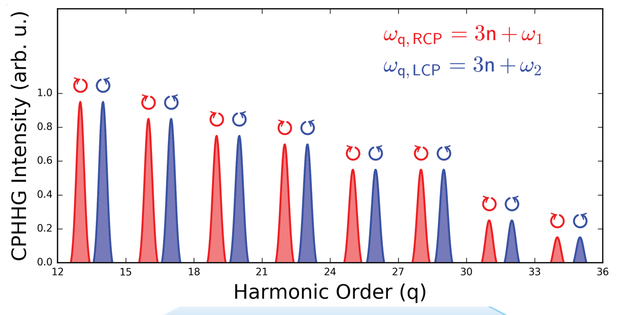
Abstract

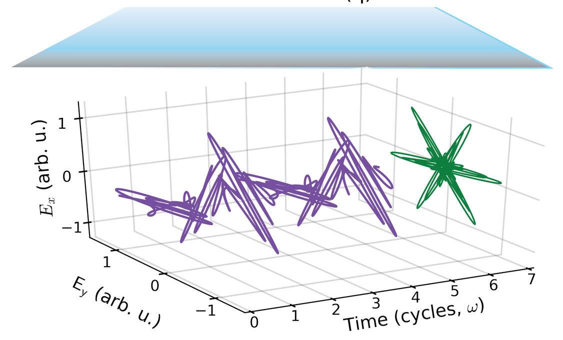
- **Motivation** - Circularly polarized high-harmonic generation (CPHHG) has recently emerged as a breakthrough light science technique to produce laser-like beams of high-energy, ultrashort, circularly polarized light on a table-top scale system. Typically, CPHHG results in a comb of high-harmonics 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 generated high-harmonics.

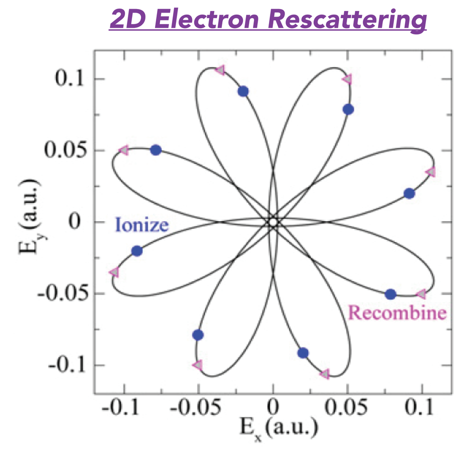


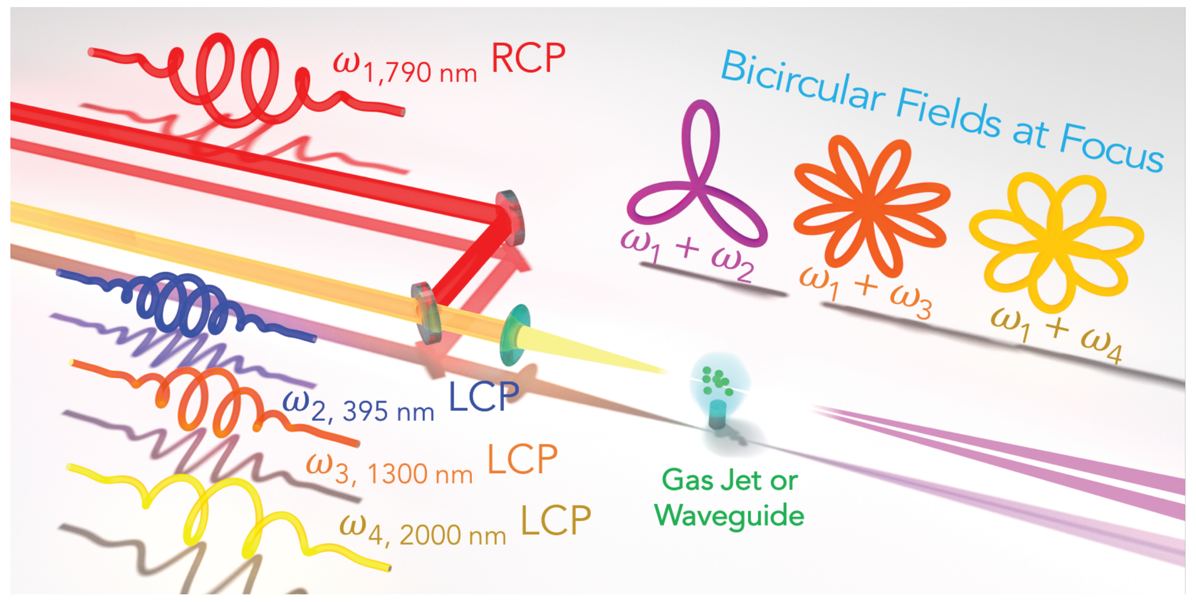
- **Results** -
  1. The spectral helcity distribution in CPHHG can be actively controlled via the intensity ratio of the bicircular driving field, resulting in direct control over the polarization of the underlying APTs.
  2. Collective multielectron effects can be exploited in CPHHG to yield a bright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

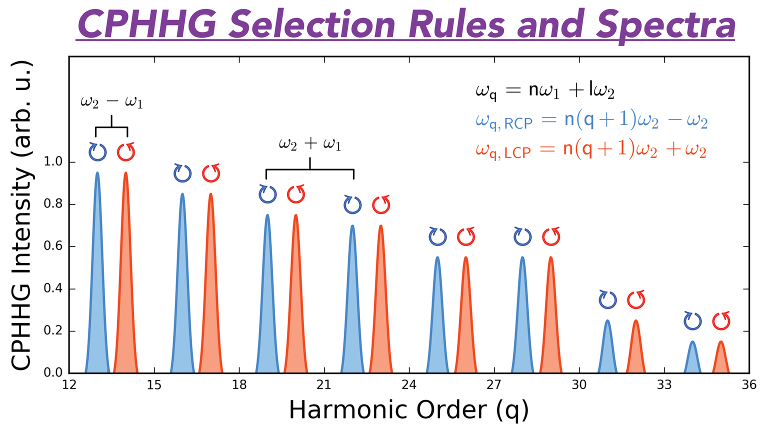


Bicircular Driven CPHHG

- Single-stage, high-energy Ti:sapphire amplifier (790 nm, 9 mJ, 45 fs, 1 kHz).
- Second harmonic generation (395 nm) and short-wave IR OPA (1300 nm or 2000 nm).
- Fundamental and SHG/OPA beams spatiotemporally overlapped in a collinear geometry in a gas jet (790 nm + 395 nm) or hollow core waveguide (790 nm + OPA).







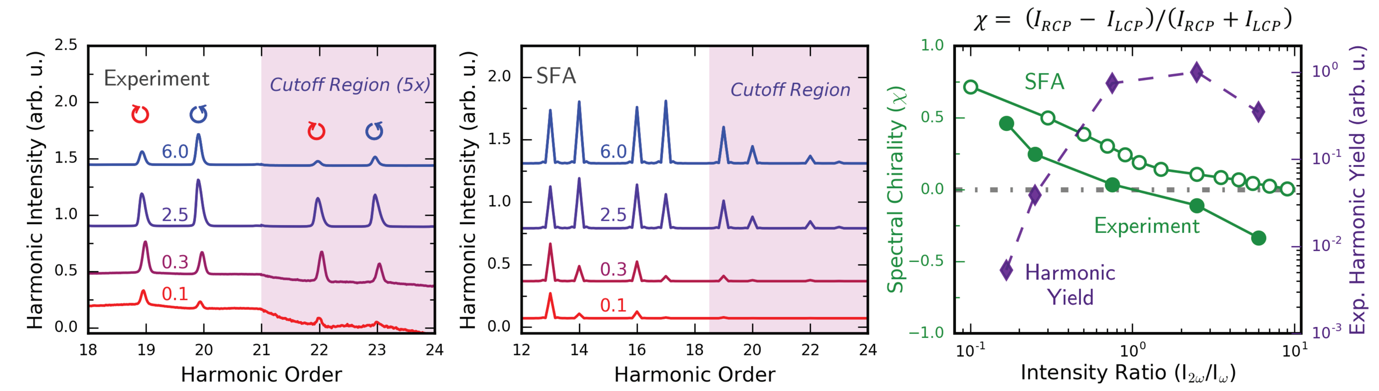
Controlling Quantum Electrodynamics in CPHHG: Custom Spectrotemporal Waveforms for Attosecond Chiral Spectroscopy

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

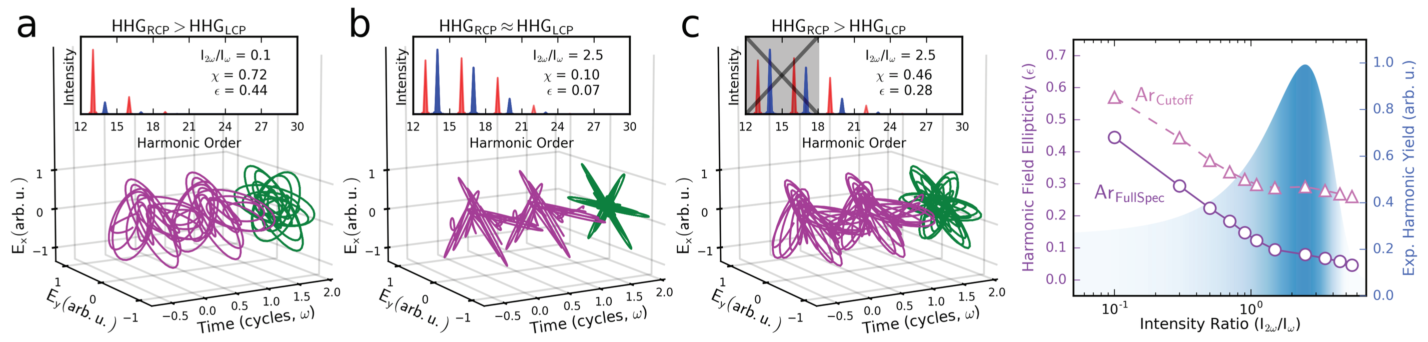
- The polarization of the underlying APTs produced via CPHHG is directly coupled to the spectral intensities of RCP and LCP harmonics.

$I_{q,RCP} \approx I_{q,LCP} \Rightarrow$ 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!



- Numerical SFA calculations allow for direct access to the subcycle dynamics of the APTs as the intensity ratio is varied.

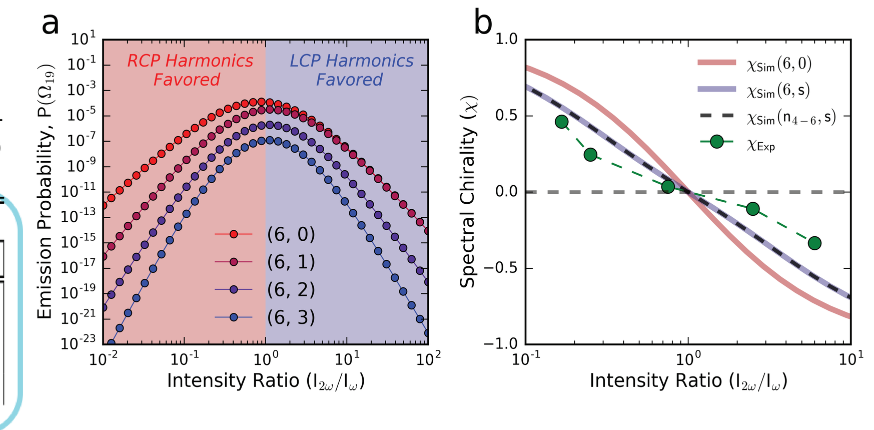


- Remarkably, a simple perturbative photon model for CPHHG emission accurately recaptures the spectral control afforded by altering the intensity ratio.

$$I_q \propto P(\Omega) \propto \sum_{i \neq j}^{\infty} p_R^{|n_R^i|} p_B^{|n_B^j|}$$

$$p_R = \frac{I_R}{I_R + I_B} = \frac{1}{1 + I_B/I_R} \qquad p_B = \frac{I_B}{I_R + I_B} = \frac{1}{1 + {}^{1}/_{(I_B/I_R)}}$$

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)	$11\omega_1 + 12\omega_2$	$p_{2\omega}$



Conclusions and Outlook

- We demonstrate active control over the spectrotemporal structure of high-harmonic waveforms produced via CPHHG, yielding user-defined harmonic beams for ultrafast chiral spectroscopies.
- The methods presented here are both straightforward and robust, allowing for easy integration into existing setups.
- Future work involves a rigorous theoretical treatment of the CM effect in Ar (and other species) and extension of these techniques to ultraviolet and mid-infrared driven CPHHG, thus enabling transient absorption studies of sub-fs chiral dynamics.

References

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[4]Schoun, S.B. Attosecond pulse shaping around a Cooper minimum. *Phys. Rev. Lett.* **112**, 153001 (2014).