

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

Kevin M. Dorney,<sup>1,\*</sup> Jennifer L. Ellis,<sup>1</sup> Carlos Hernández-García,<sup>2</sup> Daniel D. Hickstein,<sup>1</sup> Christopher A. Mancuso,<sup>1</sup> Nathan Brooks,<sup>1</sup> Tingting Fan,<sup>1</sup> Guangyu Fan,<sup>3</sup> Dmitriy Zusin,<sup>1</sup> Christian Gentry,<sup>1</sup> Patrik Grychtol,<sup>1</sup> Henry C. Kapteyn,<sup>1</sup> and Margaret M. Murnane<sup>1</sup>

<sup>1</sup>*JILA, Department of Physics, University of Colorado Boulder and NIST, Boulder, Colorado 80309, USA*

<sup>2</sup>*Grupo de Investigación en Aplicaciones del Láser y Fotónica, Departamento de Física Aplicada, University of Salamanca, E-37008 Salamanca, Spain*

<sup>3</sup>*Photonics Institute, Vienna University of Technology, A-1040 Vienna, Austria*

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High harmonics driven by two-color counterrotating circularly polarized laser fields are a unique source of bright, circularly polarized, extreme ultraviolet, and soft x-ray beams, where the individual harmonics themselves are completely circularly polarized. Here, we demonstrate the ability to preferentially select either the right or left circularly polarized harmonics simply by adjusting the relative intensity ratio of the bichromatic circularly polarized driving laser field. In the frequency domain, this significantly enhances the harmonic orders that rotate in the same direction as the higher-intensity driving laser. In the time domain, this helicity-dependent enhancement corresponds to control over the polarization of the resulting attosecond waveforms. This helicity control enables the generation of circularly polarized high harmonics with a user-defined polarization of the underlying attosecond bursts. In the future, this technique should allow for the production of bright highly elliptical harmonic supercontinua as well as the generation of isolated elliptically polarized attosecond pulses.

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High-harmonic generation (HHG) is an extreme nonlinear optical process, producing bright coherent extreme-ultraviolet (EUV) and soft x-ray beams with pulse durations in the femtosecond-to-attosecond regime on a tabletop [1–10]. The high temporal coherence of HHG makes it an ideal tool to capture the fastest charge and spin dynamics in atomic, molecular, and materials systems [11–17], while the high spatial coherence has enabled imaging with subwavelength spatial resolution in the EUV region for the first time [18–20]. To date, most studies have used linearly polarized laser fields to drive the HHG process, which results in linearly polarized harmonic radiation—a direct consequence of the nanoscale single-atom HHG mechanism [3,21–23]. In the quantum picture of HHG, harmonics are emitted as the result of a dipole transition of an electron in a laser-dressed continuum state back to the ground state of the atom. In the semiclassical picture of HHG, an atom first undergoes strong-field ionization, followed by acceleration of an electron in the laser field, before it recombines with its parent ion and emits a high-energy photon. In either picture, as the ellipticity of the driving laser field is changed from linear to circular polarization, the electron trajectory deviates from simple linear motion, and the probability for recombination strongly decreases, precluding the emission of circularly polarized harmonics using one-color circularly polarized driving laser fields [23,24].

Fortunately, if the HHG process is driven by two-color counterrotating circularly polarized laser fields (bichromatic CPHHG), bright, phase-matched, circularly polarized harmonics can be generated that span from the EUV to

the soft x-ray regions of the spectrum, with very unique spectral and temporal signatures [25–33]. The primary spectral characteristic of bichromatic CPHHG is the suppression of any harmonic orders that do not conserve photon spin angular momentum, resulting in pairs of harmonic orders with opposite helicity [i.e., left-circular (LCP) or right-circular polarization (RCP)] [25–36]. Moreover, the driving laser field parameters determine the spectral, temporal, and polarization properties of CPHHG, enabling the creation of tailored CPHHG waveforms. For instance, the frequencies of the bichromatic laser field can be chosen to tune the photon energy and bandwidth of the emitted harmonics from the EUV to the soft x-ray region [29]. Additionally, the ellipticity of the individual harmonics can be controlled by varying the ellipticity of the driving lasers [37] or by driving the CPHHG process with a near-degenerate bichromatic field [38]. Precise control of the driving field allows for the generation of highly circular EUV and soft x-ray harmonics, which can probe chiral-specific phenomena such as magnetic dichroism [28,29], discrete molecular symmetries [39–41], and even lead to enhanced detection of chiral enantiomers via photoelectron circular dichroism [42–44].

The spectral control afforded by a bichromatic driving laser field also has direct consequences on the underlying temporal waveform generated during the CPHHG process. In the time domain, the presence of phase-locked harmonics with alternating helicities manifests as a train of attosecond bursts with exotic polarization properties [27,28,33,35,36,45–47]. Under typical experimental conditions, the unique attosecond pulse trains (APTs) of CPHHG consist of *linearly*

polarized bursts of EUV and soft x-ray radiation [29,30,39,45], each rotated in the polarization plane by an angle that depends on the central frequencies of the combined laser field [28,29,48]. A linearly polarized APT results when the RCP and LCP harmonics possess nearly equal amplitudes (i.e., an achiral spectrum), which follows naturally from a Fourier analysis of the CPHHG spectrum. Consequently, if the spectral amplitudes are unequal (i.e., a chiral spectrum), then the individual attosecond bursts become elliptically polarized, while the harmonics retain their circular polarization [45,48–50]. This rather unique spectrottemporal coupling in CPHHG has recently led to a strong interest in producing chiral CPHHG spectra [45,48–52], since such spectra allow for a straightforward way to generate bright highly elliptical APTs. Moreover, the high brightness and relative simplicity of collinear, bichromatic CPHHG make it an attractive alternative to more complex methods of generating highly elliptically or circularly polarized APTs [53–57].

To date, several schemes [45,48–51] have been proposed to induce chirality in bichromatic CPHHG by exploiting helicity-dependent aspects of the macroscopic or microscopic response to the external field. In the macroscopic approach, the helicity-selective nature of phase matching [5,28,45,51] in CPHHG has been suggested as a viable method to suppress harmonics of a given circularity, thus enabling the production of highly chiral CPHHG spectra [45,51]. Microscopically, the harmonic emission in atoms containing  $p$ -type ground states has been theoretically shown to exhibit a helicity dependence, yielding elliptical APTs at the single-atom level [48,49]. Additionally, a judicious choice of the frequencies of the two-color driving field can result in electron trajectories that preferentially produce LCP or RCP harmonics [50]. Although these methods offer promising routes for producing elliptically polarized attosecond pulses, it is unclear if they can achieve the desired chiral specificity in all situations, with a reasonably large chirality, or across a broad range of photon energies. Moreover, the practical challenges associated these schemes has, to the best of our knowledge, precluded an experimental demonstration of active helicity control in CPHHG. Thus, a straightforward and robust method for controlling the polarization state of the APTs has yet to be realized.

In this Letter, we experimentally show that by adjusting the intensity ratio of the two driving lasers,  $I_{2\omega}/I_{\omega}$ , we can achieve significant control over the chirality of the CPHHG process, making it possible to selectively enhance either the LCP or RCP harmonics. We quantify the degree of chiral control via the spectral chirality,  $\chi = (I_{\text{RCP}} - I_{\text{LCP}})/(I_{\text{RCP}} + I_{\text{LCP}})$ , where  $I_{\text{RCP}}$  and  $I_{\text{LCP}}$  refer to the integrated intensity of all RCP and LCP harmonics, respectively, across the entire spectrum. We find that this control extends across much of the high-harmonic spectrum, providing an energy-independent method to produce chiral CPHHG waveforms. This control over the spectral chirality of CPHHG inherently allows for the manipulation of the polarization of the resulting APTs (see Fig. 1). We confirm our experimental results by performing numerical

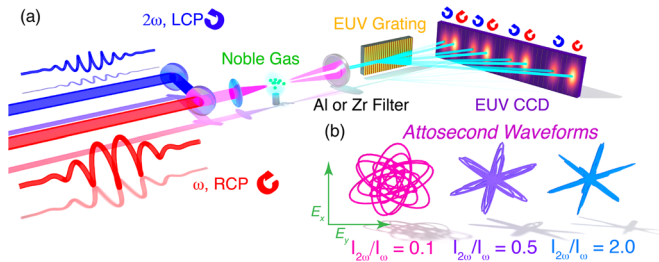


FIG. 1. Selective enhancement of RCP HHG. (a) Experimental scheme for bicircular ( $\omega$ ,  $2\omega$ ) generation of circularly polarized high harmonics of controllable chirality. (b) Polarization plane projections of the temporal structure for the theoretical attosecond pulse trains supported by experimental CPHHG spectra generated at different intensity ratios ( $I_{2\omega}/I_{\omega}$ ) of the driving field, plotted for one cycle of the  $\omega$  field (2.67 fs).

simulations of high-harmonic generation including propagation [58], which show that the observed chiral CPHHG spectra can support elliptical APTs. We describe both the theoretical and experimental results in terms of an intuitive photon model, in which harmonics that rotate in the same direction as the more intense laser of the bicircular field are preferentially enhanced. In the future, this technique should allow for the production of bright highly elliptical harmonic supercontinua as well as the generation of isolated elliptically polarized attosecond pulses.

The circularly polarized high harmonics are generated via collinear mixing of the fundamental (790 nm, RCP) and second harmonic (395 nm, LCP) of a Ti:sapphire regenerative amplifier (1 kHz, 8.5 mJ, 45 fs, KMLabs Wyvern HE). The circularly polarized beams are spatially and temporally overlapped, then focused onto a supersonic expansion of Ar gas. The intensity of each beam is controlled via half-wave-plate-polarizer pairs in each arm, and the CPHHG spectra as a function of the  $I_{2\omega}/I_{\omega}$  ratio are recorded in the far field using a dispersive EUV grating and CCD camera (see the Supplemental Material [59]).

As the intensity ratio is varied, either RCP or LCP ( $3n + \omega$  or  $3n + \omega_2$ ) harmonics are preferentially produced, which imparts a net chirality to the harmonic spectrum (Fig. 2). Remarkably, this spectral chirality can be introduced without significantly altering the bandwidth of the individual harmonic orders or the spectral envelope, indicating that the up-conversion process is not strongly altered as the intensity ratio is scanned [Fig. 2(a)]. For instance, we can safely rule out effects from a dynamically changing harmonic envelope because these effects would be strongest for the near-cutoff orders, in which the envelope itself can lead to more intense RCP harmonics as compared to their higher energy LCP counterparts (i.e.,  $\chi > 0$ ). However, our experimental spectra demonstrate that significant chiral control can be achieved in the near-cutoff region, which allows for controlling the ellipticity of high-energy attosecond waveforms.

The degree of chiral control over the CPHHG spectra to changes in the  $I_{2\omega}/I_{\omega}$  ratio are further reinforced by

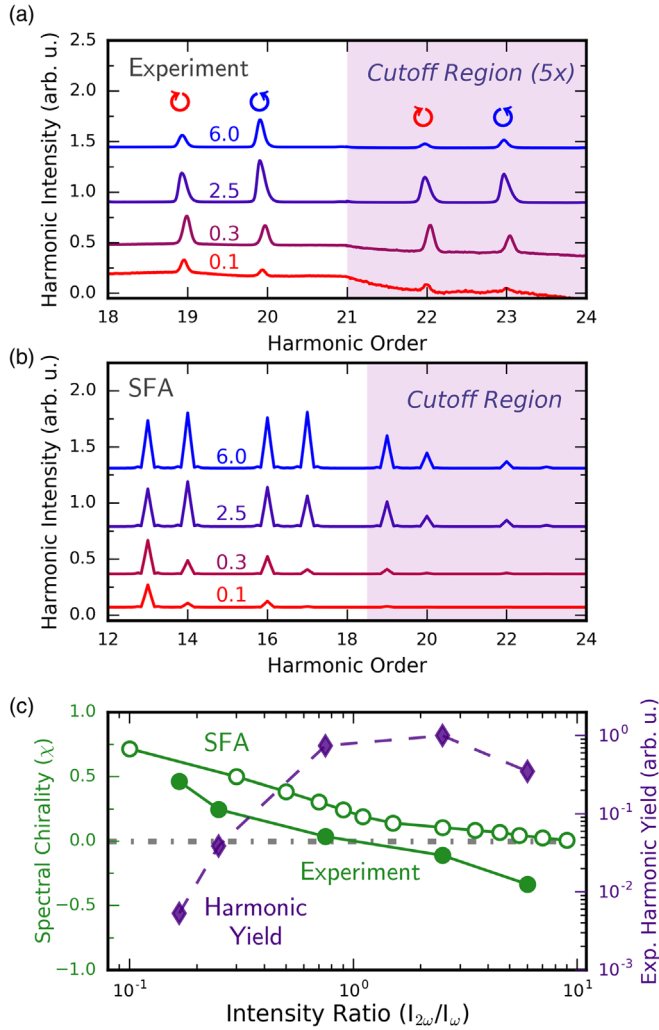


FIG. 2. Circularly polarized high harmonic spectra in Ar. (a) Experimental and (b) theoretical CPHHG spectra recorded at increasing  $I_{2\omega}/I_{\omega}$  ratios (values given next to the spectra) at total intensities of  $\sim 2 \times 10^{14}$  W/cm<sup>2</sup>. The harmonics in the cutoff region in (a) have been scaled for clarity. (c) Experimental and theoretical spectral chirality  $\chi = (I_{\text{RCP}} - I_{\text{LCP}})/(I_{\text{RCP}} + I_{\text{LCP}})$  (left axis) and experimental CPHHG yields (right axis) observed in Ar as a function of the  $I_{2\omega}/I_{\omega}$  ratio of the two-color field. Harmonic yields are obtained as the total integrated signal for all observed harmonic orders in the experimental spectra.

numerical simulations of the CPHHG process [Fig. 2(b)], which include both the microscopic (single-atom) and macroscopic physics. To this end, we use the electromagnetic-field-propagator method [58], where the single-atom dipole acceleration is calculated using the strong-field approximation (SFA) [59]. Phase-matching effects are included by propagating the single-atom response to the detector plane; however, we note that under the experimental conditions employed phase matching does not play a significant role in the observed chiral control [59]. Aside from a slight difference in cutoff, the theoretical spectra accurately reproduce the experimentally observed trends in spectral

chirality. Interestingly, the theoretical spectra indicate that the spectral chirality may only be controlled in favor of RCP harmonics (i.e.,  $\chi > 0$ ), which could be the result of helicity-dependent absorption in the generating medium [28,45] or simply a stronger envelope effect in the theoretical spectra. Additionally, the theoretical and experimental spectra display a similarly strong suppression of the spin-forbidden  $3n$  harmonic orders, indicating a high degree of circularity of the individual harmonic orders across the CPHHG spectra [27,28,35,36,44]. This high degree of suppression is maintained as the intensity ratio of the bicircular field is altered. Therefore, the circular polarization of the individual harmonics themselves is maintained while the spectral chirality, and thus the ellipticity of the APT, is tuned. However, there is a practical limitation—adjusting the  $I_{2\omega}/I_{\omega}$  ratio in favor of one color over the other results in a decreased CPHHG yield [Fig. 2(c)]. Fortunately, this reduction in harmonic signal is not as strong as the ellipticity dependence of HHG using one-color driving lasers, which leads to complete suppression of harmonic emission for ellipticity values of  $\sim 0.2$  [23,24].

The chiral control imposed on the CPHHG output through the intensity ratio of the bicircular field can be understood by making an analogy between the HHG up-conversion process and a parametric process [28,29,34]. For circularly polarized driving lasers, conservation of energy and photon spin angular momentum requires that the  $q$ th harmonic in the CPHHG spectrum is generated as follows:  $q\omega = l\omega + m2\omega$ , where  $l - m = \pm 1$ , which then results in RCP and LCP harmonics [28,29,35,36,46]. In this model, the probability of emission for a given harmonic order is largely determined by the photon density of the beam that is copolarized with that particular harmonic channel [35,39,59]. Owing to the parametric nature of HHG, detailed balance allows CPHHG emission via a particular harmonic channel to be viewed as being initiated by ionization from the corresponding co-polarized driving laser. Owing to the highly nonlinear dependence on the electric-field strength for tunneling ionization rates [70], the ionization rates are highly sensitive to the relative strengths of the two components of the bicircular field. Thus, as the relative photon density of each wavelength is altered via the intensity ratio of the two-color field, the probability of emission for a given harmonic channel is altered as well. Consequently, the spectral intensities of RCP or LCP harmonics can be arbitrarily increased or decreased via the  $I_{2\omega}/I_{\omega}$  ratio, leading to a net chirality across the CPHHG spectrum, independent of the CPHHG bandwidth [59]. However, as the number of photons involved in generating a particular harmonic order increases, perturbative arguments are expected to break down, which may limit the degree of chiral control attainable for very high harmonic orders or for mid-IR-driven HHG. Fortunately, employing UV-driving lasers can mitigate this problem, which should yield high-brightness, highly chiral spectra in the soft x-ray region of the spectrum [71]. Furthermore, we note that this model is also in agreement with recent results in which the sensitivity

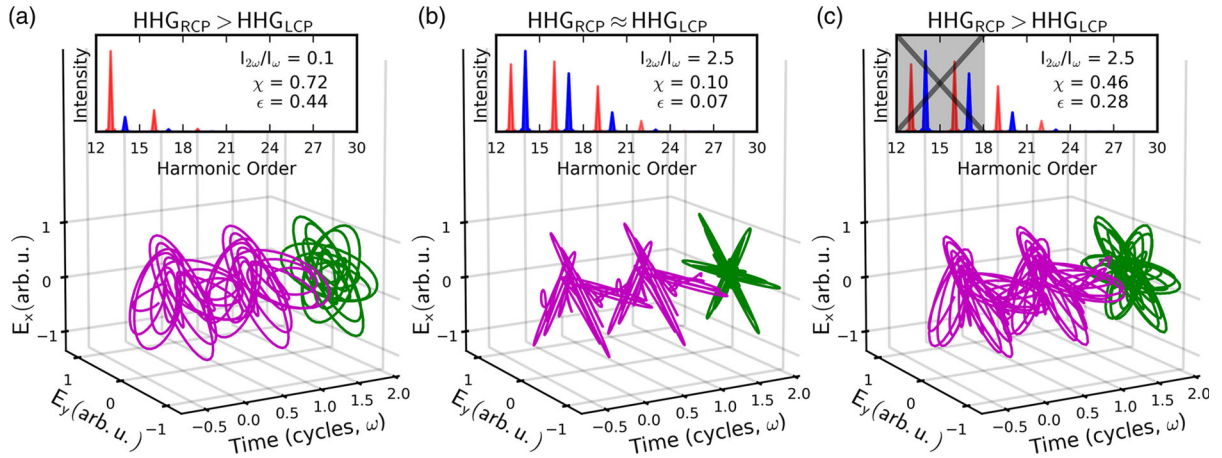


FIG. 3. Theoretical attosecond pulse trains (APTs) and corresponding spectra in Ar [(a)–(c)] at a total intensity of  $2.0 \times 10^{14}$  W/cm<sup>2</sup>. The temporal profile is obtained via an inverse Fourier transformation of the corresponding CPHHG spectra at the indicated  $I_{2\omega}/I_\omega$  ratio, spectral chirality ( $\chi$ ) and harmonic field ellipticities ( $\epsilon$ ) [insets]. Here, light red HHG orders are RCP, while dark blue orders are LCP. As the CPHHG spectra evolve from being dominated by RCP harmonics to a preference of LCP harmonics, the APTs evolve from elliptical to linear (a, b). Selecting a small bandwidth near the cutoff (c, unshaded region) restores the ellipticity of the APTs, even when the  $I_{2\omega}/I_\omega$  ratio is optimized for the brightest harmonic signal.

of the strong field ionization rates to the initial orbital angular momentum of  $p$ -type electrons [72–75] can yield elliptically polarized APTs at the single-atom level [48,49].

The theoretical CPHHG spectra provide a means to quantify the effect of the spectral chirality on the ellipticity of the corresponding APTs. By performing a Fourier transform of the simulated CPHHG spectra, we can access the subcycle dynamics of the CPHHG emission process. As the intensity ratio of the driving laser field is increased—which induces an increase in the relative intensity of the LCP harmonics—the polarization of the individual attosecond bursts within the harmonic field evolve from being elliptical [Fig. 3(a)] to near linear [Fig. 3(b)]. Interestingly, for very large intensity ratios (i.e.,  $I_{2\omega}/I_\omega \gg 1$ ), we find the polarization of the CPHHG pulse train remains nearly linear [59]. However, the application of an appropriate spectral filter for the high-energy cutoff harmonics can restore the ellipticity of the CPHHG waveform [Fig. 3(c)], thus allowing for a straightforward method to produce high-energy elliptically polarized attosecond pulses [50]. Furthermore, this method, in conjunction with phase-matching techniques to extend the CPHHG cutoff [5,42], could yield bright, highly elliptical attosecond pulses in the soft x-ray region of the spectrum. In this case, proper gating of these elliptical APTs could also yield a practical way to generate isolated elliptically polarized attosecond bursts, which are difficult to produce via conventional methods [44,50–52,56,57,76].

The experimental results and simulations offer corroborating evidence that the attosecond waveform of CPHHG can be controlled by adjusting the intensity ratio of the bicircular fields. However, at each intensity ratio, the CPHHG process is driven by a unique electric field waveform, which affects the ionization [77], propagation [78], and recombination [79,80] dynamics of the electrons. Consequently, we find that controlling the ellipticity of

the APTs comes at the cost of a reduction in the CPHHG flux [Figs. 2(c) and 4]. We find that the harmonic yield in Ar is maximized when  $I_{2\omega}/I_\omega$  is approximately 2.5, which agrees with theoretical predictions of the CPHHG yield in these fields [26]. These findings are supported by recent photoelectron and photoion spectroscopy measurements [78–80], which show that the yield of high-energy electron

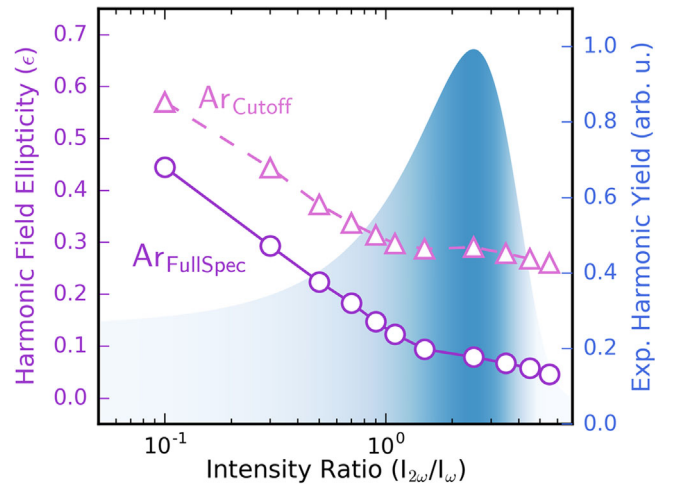


FIG. 4. Practical implications of controlling the ellipticity of the attosecond bursts in Ar. As the intensity ratio of the two-color field is increased in favor of the  $2\omega$  component, the ellipticity of the attosecond bursts (circle symbols) is quickly reduced, such that near-linearly polarized bursts are obtained when the experimental CPHHG signal is brightest (shown here as a Gaussian fit of the experimental data, blue gradient, right axis). However, spectral filtering of the CPHHG spectra to select the cutoff harmonics can significantly increase the ellipticity of the attosecond bursts (triangle symbols), even when the intensity ratio of the driving field is optimized for the CPHHG yield.

recollisions is optimized when the vector potentials of the two fields are approximately equal. However, the intensity ratio required to optimize the CPHHG yield corresponds to achiral experimental spectra, and thus linear APTs. Fortunately, this limitation can be overcome by employing readily available commercial metal foils to isolate the near-cutoff region of the CPHHG spectra, which can double the ellipticity of the harmonic field (Fig. 4). Consequently, appropriate spectral filtering should lead to increased sensitivity in experiments employing CPHHG to probe chiral processes.

In conclusion, we have shown that the chirality of circularly polarized high harmonics can be controlled via the intensity ratio of the two drivers in the counterrotating two-color field, which makes it possible to selectively enhance either the left or right circularly polarized harmonic orders. We have shown that the effects of changing the intensity ratio on the CPHHG chirality can be intuitively understood by treating the up-conversion process parametrically, where the harmonics are generated by absorption of photons with a definite spin. Most importantly, we find that the induced chirality in the CPHHG spectra allows for direct control over the attosecond polarization of the CPHHG pulse trains. Moreover, our results suggest a straightforward route for generating bright, high-energy, elliptically polarized APTs, without extensive modification of existing setups. As such, this method could be exploited to produce polarization-tailored attosecond waveforms and even yield isolated elliptically polarized attosecond bursts. Furthermore, the results presented here do not rely on the choice of frequencies for the bichromatic field, thus allowing this method to be applied to CPHHG driven by both UV and mid-IR lasers. The results presented here will not only aid development of novel sources of circularly polarized extreme ultraviolet and x-ray radiation, but also extend the applicability of these light sources to studying chiral dynamics on the attosecond time scale.

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\*Corresponding author.

kevin.dorney@colorado.edu

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