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

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High harmonics driven by two-color counter-rotating 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 sub-wavelength 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 semi-classical 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 counter-rotating 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 spectro-temporal 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, $\chi = (I_{RCP} - I_{LCP}) / (I_{RCP} + I_{LCP})$, where I_{RCP} and I_{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 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.

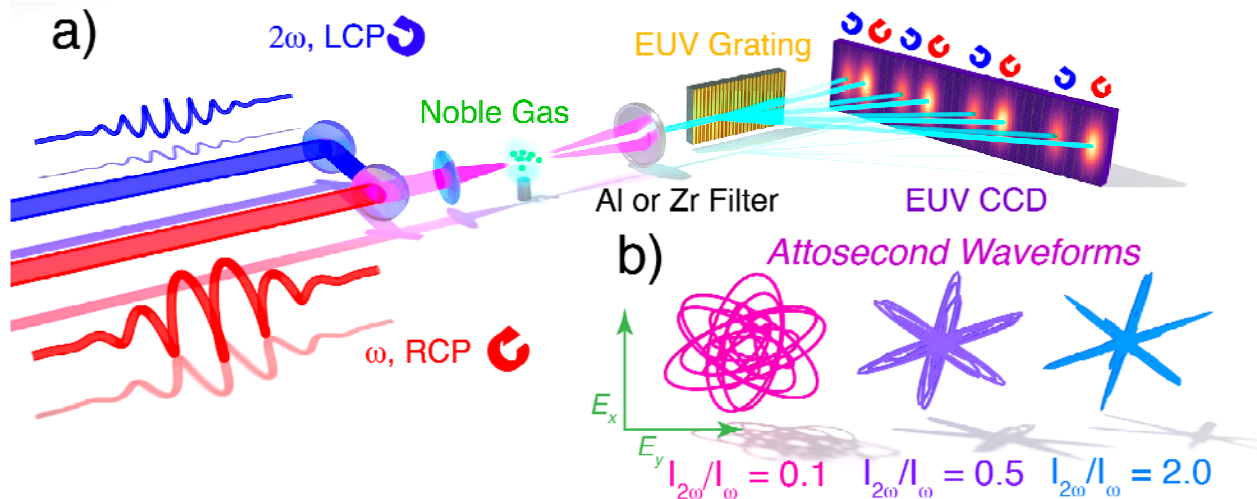


Figure 1. Selective enhancement of RCP HHG. (a) Experimental scheme for bicircular (ω , 2ω) generation of circularly polarized high harmonics of controllable chirality. (b) Polarization plane projections of the temporal structure for the theoretical attosecond pulse trains supported by experimental CPHHG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

The circularly polarized high harmonics are generated via collinear mixing of a 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-waveplate-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 Supplemental Material, [59]).

As the intensity ratio is varied either RCP or LCP, ($3n + \omega$, $3n + \omega_2$ for the commensurate laser frequencies employed in this study), harmonics are preferentially produced, which imparts a net chirality to the harmonic spectrum [Figure 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 upconversion process is not strongly altered as the intensity ratio is scanned [Figure 2a]. 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 numerical simulations of the CPHHG process [Figure 2b], 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 [Figure 2c]. 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 > 0.2 [23, 24].

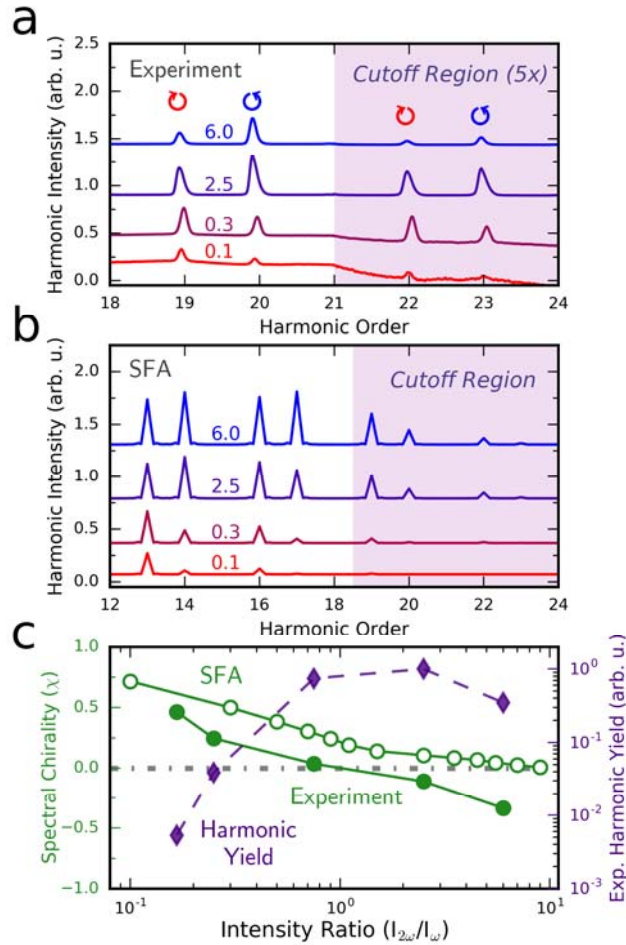


Figure 2. Circularly polarized high harmonic spectra in Ar. (a) Experimental CPHHG spectra recorded at increasing $I_{2\omega}/I_\omega$ ratios (values given next to the spectra) at a total intensity of 2×10^{14} W/cm². (b) Simulated CPHHG spectra for Ar at a total intensity of 2.0×10^{14} W/cm² and the same intensity ratios as the spectra in (a). The harmonics in the cutoff region in (a) have been scaled for clarity. (c) Experimental and theoretical spectral chirality, $\chi = (I_{RCP} - I_{LCP}) / (I_{RCP} + I_{LCP})$, (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.

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 upconversion 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 co-polarized with that particular harmonic channel [35, 39, 59]. This is largely because a parametric process like HHG must obey detailed balance, which demands that photorecombination is conjugated to the photoionization process. As such, CPHHG emission via a particular harmonic channel can be viewed as being initiated by ionization from the corresponding co-polarized component of the driving field. Owing to both 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. Therefore, bright CPHHG emission favors harmonics that rotate with the more intense component of the combined field. Consequently, the spectral intensities of RCP or LCP harmonics can be arbitrarily increased or decreased, 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 breakdown, which may limit the degree of chiral control attainable through this method for very high harmonic orders or 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 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].

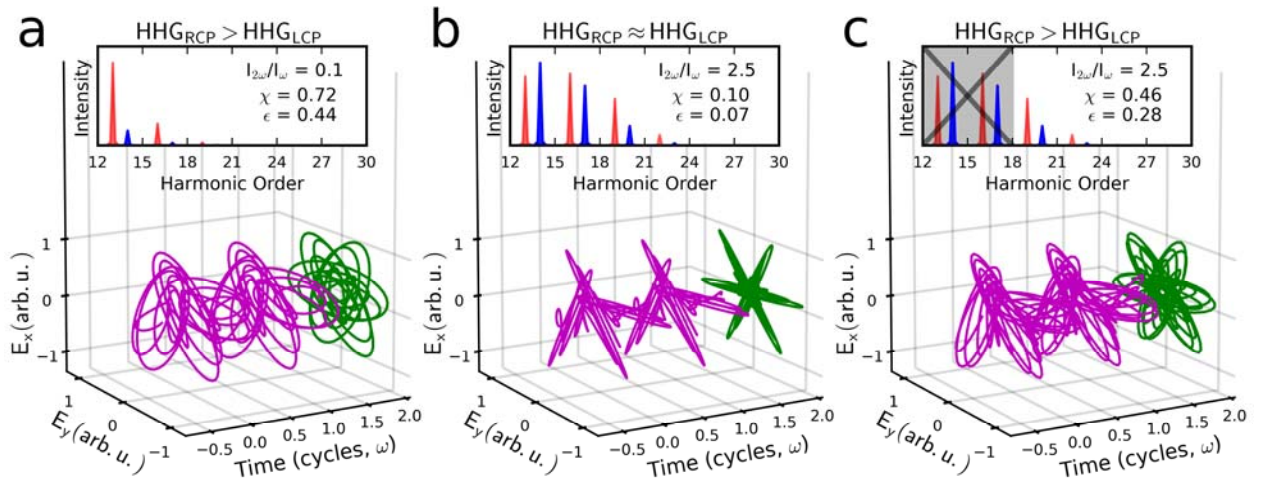


Figure 3. Theoretical attosecond pulse trains (APTs) and corresponding spectra in Ar (a, b, c) at a total intensity of 2.0×10^{14} W/cm². The temporal profile is obtained via an inverse Fourier transformation of the corresponding CPHHG spectra at the indicated $I_{2\omega}/I_\omega$ ratio, spectral chirality (χ) and harmonic field ellipticities (ϵ) [insets]. Here,

light red HHG orders are RCP and rotate with the fundamental, while dark blue orders are LCP and rotate with the second harmonic field. 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). If, however, a small bandwidth near the cutoff is selected (c, unshaded region) the ellipticity of the APTs can be restored, especially when the $I_{2\omega}/I_\omega$ ratio of the driving field is optimized for the brightest harmonic signal.

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 sub-cycle dynamics of the CPHHG emission process. As the intensity ratio of the driving laser field is increased – which induces a corresponding increase in the relative intensity of the LCP harmonics – the polarization of the individual attosecond bursts within the harmonic field evolve from being elliptical [Figure 3(a)] to near linear [Figure 3(b)], with a corresponding decrease in the spectral chirality. This demonstrates how the intensity ratio of the driving fields actively controls both the spectral chirality and the polarization of the corresponding attosecond bursts, allowing for the generation of CPHHG waveforms with user-defined polarization states. 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 [Figure 3c], thus allowing for a straight-forward 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].

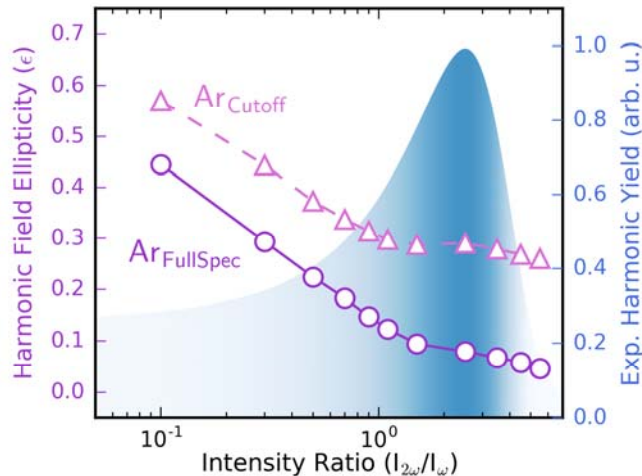


Figure 4. Practical implications of controlling the ellipticity of the attosecond bursts in Ar. As the intensity ratio of the two-color field is increased in favor of the 2ω component, the ellipticity of the attosecond bursts (circle symbols) is quickly reduced, such that near linearly polarized bursts 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.

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 quantum dynamics of the field-driven electrons. Consequently, we find that controlling the ellipticity of the APTs comes at the cost of a reduction in the CPHHG flux [Figure 2c & 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]. Additionally, these findings are supported by recent photoelectron spectroscopy measurements [77-80], which have shown that the yield of high-energy electron 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 easily overcome by employing readily available commercial metal foils to isolate the near-cutoff region of the CPHHG spectra, which can effectively double the ellipticity of the harmonic field [Figure 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 counter-rotating 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 upconversion 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 timescale.

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