

Control of spectral chirality in high-harmonic generation using two-color bicircular laser fields

N. Zhavoronkov^{1,*}, U. Bengs,¹ and D. B. Milošević^{1,2,3}¹*Max-Born Institute for Nonlinear Optics and Short Pulse Spectroscopy, Max-Born-Straße 2A, D-12489 Berlin, Germany*²*Faculty of Science, University of Sarajevo, Zmaja od Bosne 35, 71000 Sarajevo, Bosnia and Herzegovina*³*Academy of Sciences and Arts of Bosnia and Herzegovina, Bistrik 7, 71000 Sarajevo, Bosnia and Herzegovina*

(Received 16 January 2026; accepted 20 April 2026; published 11 May 2026)

We have extended the two-color bicircular approach for generating chiral XUV radiation via high-harmonic generation by combining femtosecond driving laser fields with 1:3 frequency ratio of wavelengths 1200 and 400 nm. With optimized driving-field intensities, we produced highly chiral spectra in which the vast majority of harmonics sharing the same helicity constitute the vast majority of spectral content. We support these experimental finding with a numerically comprehensive analytical model that describes the propensity (selection) rules for emission of circularly polarized high harmonics. We identify and confirm the rules responsible for the contrast between the allowed odd harmonics of orders $4q + 1$ and $4q - 1$ (q an integer), which possess opposite helicities, as well as the complete prohibition of even-order harmonics in the high-harmonic spectra. We further apply these rules to explain the spectral shape and ellipticity of the emitted odd harmonics as functions of both the intensity and the ellipticity of the driving-field components. Using XUV polarimetry, we retrieved full Stokes vector, confirming achieved ellipticities higher than 0.9.

DOI: [10.1103/xtck-9s9n](https://doi.org/10.1103/xtck-9s9n)

I. INTRODUCTION

Many frontiers of attosecond science—which investigates the behavior of electrons in atoms and molecules with unprecedented temporal resolution on the electron's natural time scale—rely primarily on attosecond pulses ($1 \text{ as} = 10^{-18} \text{ s}$) produced through high-harmonic generation (HHG). High-order harmonics from gas targets have attracted considerable attention in both experiment and theory, establishing the semiclassical three-step model as the main mechanism for harmonic generation [1–7]. Naturally, control over the macroscopic parameters of the resulting XUV radiation, including its amplitude, frequency, and polarization, is essential for realizing the full potential of HHG as a source for experiments. In particular, the availability of circularly polarized attosecond pulses, enabling attosecond-resolved measurements of chiral-sensitive electron dynamics, remains both a challenging and an important task. In principle, synchrotron and free-electron-laser sources with tunable polarization provide sufficient brightness and cover the spectral range for such measurements. Although the generation and initial application of subfemtosecond pulses were recently demonstrated at the Linac Coherent Light Source [8,9], access to such large-scale facilities for chiral-sensitive experiments is very limited. This makes table-top HHG sources highly attractive for broadly accessible experimental platforms.

Over the past decade, numerous methods have been developed to address the challenge of generating the circularly polarized XUV radiation suitable for producing attosecond pulses. An overview of these approaches, including various combinations of driving laser fields and different techniques for utilizing of generating media, can be found elsewhere (see Refs. [10,11]). Recently, another HHG scheme driven by copropagating bichromatic $\omega + 2\omega$ fields with orthogonal polarizations has attracted considerable attention. The field polarizations may be linear or slightly elliptical, with opposite helicities and perpendicular major axes [12]. Highly helically polarized harmonics have been reported, emitted without particular symmetry constraints, i.e., with odd or even harmonics of comparable strengths [13,14]. As a result, the overall spectral chirality, where the chirality in spectra refers to the ability of spectroscopic methods to distinguish between enantiomers (mirror-image isomers) that exhibit identical physical properties in an achiral environment, is quite pure, leading to the low ellipticity of the attosecond pulses train. To obtain attosecond pulses with polarization close to circular, it is necessary to ensure that one helicity component dominates over the other within a specific spectral range. Such suppression is achieved in a noncollinear scheme [15], but this approach is inherently accompanied by nonlinear spatial chirp, spatiotemporal distortions, and transverse walk-off. In the two-color bicircular (TCBC) approach, which relies on tailored driving laser fields [16–18], an effective helicity-dependent filter, favoring harmonics of only one helicity and producing highly elliptically polarized pulse trains, can be realized through optimal configuration of the applied scheme [19–22]. This strategy also paves the way for generating chiral isolated attosecond pulses [10].

In this paper, we address the challenge of increasing and controlling the spectral chirality of XUV radiation gener-

*Contact author: zhavoron@mbi-berlin.de

Published by the American Physical Society under the terms of the Creative Commons Attribution 4.0 International license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

ated in form of high harmonics by exploiting, for the first time, the TCBC approach in the $\omega + 3\omega$ configuration of the driving fields. First, we use results from quantum-mechanical multielectron theory based on the S -matrix formalism, the single-active-electron approximation, and the strong-field approximation (SFA) to formulate specific selection rules, to model harmonic spectra, and to reveal the characteristic features of this configuration. Second, we experimentally generate high harmonics in Ar and Ne and investigate their dependence on the experimental parameters, particularly an ellipticity and an intensity ratio of driving femtosecond radiation. Finally, we apply XUV polarimetry based on the Mueller-matrix formalism to determine the polarization state of the generated harmonics by retrieving the full Stokes vector.

II. SELECTION RULES FOR ELLIPTICAL DRIVING RADIATION

A theory of high-order harmonic generation by a bichromatic elliptically polarized laser field, composed of two coplanar components with frequencies $r\omega$ and $s\omega$ (where r and s are integers) and defined in the xy plane, was presented in Ref. [23]. The study employs a decomposition of both the driving and harmonic fields into circularly polarized components with opposite helicities. By invoking the conservation laws for energy and for the projection of the total angular momentum of atom, as well as of the laser and harmonic photons, onto the z axis, the following general selection rule was derived for circularly polarized driving fields of opposite helicities ($\varepsilon_r = -\varepsilon_s = \pm 1$):

$$n = q(r + s) + \varepsilon_r \varepsilon_n r, \quad (1)$$

where q is an integer and $\varepsilon_n = \pm 1$ denotes the ellipticity of the n th harmonic. The selection rule (1) reduces to the well-known result for a bicircular field composed of two counter-rotating circularly polarized components with $\varepsilon_r = 1$. For $r = 1$ and $s = 2$, the harmonics $n = 3q \pm 1$, $q = 1, 2, \dots$, having helicities $\varepsilon_n = \pm 1$, are emitted, while the harmonics $3q$ are absent. For the configuration with $r = 1$ and $s = 3$, the allowed harmonics satisfy the condition $n = 4q \pm 1$; i.e., only odd harmonics are emitted and all even harmonics are forbidden. In this case, the neighboring harmonics carry opposite helicities.

For an arbitrary elliptically polarized bichromatic field, the dynamical symmetry is broken and the selection rule (1) no longer applies. The emitted harmonics are elliptically polarized and have the frequency $n\omega$, where n is an arbitrary integer. The n th harmonic field $\mathcal{E}_n$ can be expressed as the sum of two components $\mathcal{E}_{n+}$ and $\mathcal{E}_{n-}$, with helicities $+1$ and -1 , respectively. These components interfere coherently to produce an elliptically polarized n th harmonic. Denoting by $p_{\pm}$ and $q_{\pm}$ the integers associated with the number of exchanged photons of frequencies $r\omega$ and $s\omega$, respectively, and with their corresponding helicities ± 1 , the mentioned angular-momentum conservation leads to the relation

$$p_+ = p_- - q_+ + q_- \pm 1. \quad (2)$$

By additionally applying energy conservation, one arrives at the following selection rule for the $n_{\pm}$ harmonic-field compo-

nent [23]:

$$n_{\pm} = (2p_- + q_- - q_+ \pm 1)r + (q_- + q_+)s. \quad (3)$$

For arbitrary integers p_- , q_- , and q_+ , the numbers $n_{\pm}$ can also be arbitrary integers. The dominant contribution to the generation of the n th harmonic arises from the smallest values of $|p_-|$, $|q_-|$, and $|q_+|$ that satisfy relation (3). This allows us to estimate which harmonics are expected to be most prominent in the spectrum.

In a more straightforward configuration where the ellipticity ε_s is fixed to $+1$, while the ellipticity $-1 \leq \varepsilon_r < 0$ is arbitrary, we have $p_- = p$, $q_- = 0$, and $q_+ = q$, so the selection rule takes the form

$$n_{\pm} = (2p - q \pm 1)r + qs, \quad (4)$$

where p and q are integer, and $n_{\pm}$ is the harmonic order n of the positive or negative helicity of harmonic-field component, depending on the combinations of p and q . Substituting into Eq. (4) the values for r and s corresponding to specific configurations, we find a remarkable difference in the development of the spectral shape of generated XUV radiation, when one of the driving laser fields is elliptical. For the $\omega + 2\omega$ case, Eq. (4) leads to $n_{\pm} = 2p + q \pm 1$ so, both even and odd harmonics are allowed and emitted, as already observed in numerous experiments [20,24]. However, it is particularly notable that for the $\omega + 3\omega$ configuration the emitted harmonics satisfy $n_{\pm} = 2(p + q) \pm 1$, i.e., only odd harmonics are allowed, while even harmonics remain forbidden. The fundamental reason for this is the symmetry of this field with respect to the half cycle of the ω field [25]. This distinctive feature of the $\omega + 3\omega$ configuration offers a potential route for improving the spectral chirality of the generated XUV radiation and preventing depolarization of the attosecond pulses [26], which is especially critical for few-cycle laser drivers [10].

III. EXPERIMENTAL SETUP

We performed the experiments using the setup schematically presented in Fig. 1. A commercial Ti:sapphire-based laser system with a single-stage regenerative amplifier (SPIT-FIRE ACE) was used, delivering 35 fs pulses with energies up to 6 mJ and a central wavelength of $\lambda = 795$ nm at 1 kHz repetition rate. The carrier-envelope phase (CEP) of the pulses was not stabilized. Because CEP fluctuations cause only an azimuthal rotation of the combined laser field, without altering its shape or symmetry [27], and because the resulting change in the field intensity is estimated to be smaller than the experimental noise, we conclude that CEP fluctuations do not noticeable influence the parameters of generated radiation.

The original beam was split into two parts in an 80/20 ratio, and each part was compressed separately to achieve optimal parameters for subsequent nonlinear conversion. The first beam was directed into an optical parametric amplifier system (TOPAS, Light Conversion) to generate 50 fs pulses tunable within the 1120–1680 nm spectral range. The radiation from the second beam was up-converted to the second harmonic ($\lambda = 400$ nm) using a 100- μ m-thick type-I beta barium borate (BBO) crystal producing pulses of 40 fs duration. Both beams then passed through achromatic broadband half-wave plate and quarter-wave plate pairs, yield-

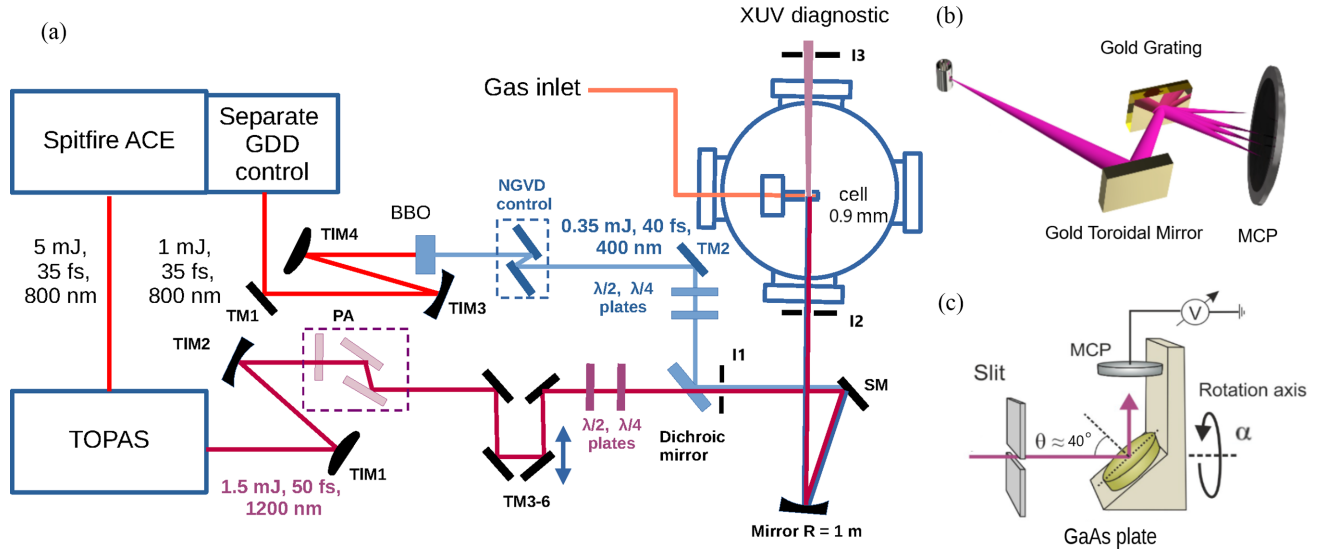


FIG. 1. Experimental setup: (a) TM1–TM6—turning mirrors, PA—attenuator, TIM1–TIM4—telescope mirrors, BBO—beta-barium-borate crystal for second-harmonic generation, NGVD—mirror pair for precompression, SM—silver mirror, MCP—detector for XUV radiation, I1–I3—auxiliary iris diaphragm to align and control collinearity of the two-color components of the driving field, (b) XUV spectrometer, and (c) XUV polarimeter.

ing nearly circular polarization ($\varepsilon_1 \approx -0.98 \pm 0.02$) with clockwise (right-rotating) helicity for the infrared beam, and $\varepsilon_3 \approx 0.93 \pm 0.02$ counterclockwise (left-rotating) helicity for the 400 nm beam. The two driving beams were carefully overlapped spatially in collinear geometry and focused with a single Ag mirror at $f/100$ into a 0.9-mm-long gas cell filled with the target gas (Ar or Ne). The cell was initially sealed with a metal foil, which was burned through by the laser beam at the start of the experiment. The resulting cell opening, $d_0 \approx 60 \mu\text{m}$, was comparable to the focal spot size, allowing the gas pressure inside the cell to be maintained between 5 and 100 mbar while keeping the vacuum in the interaction chamber at $P_{\text{rest}} \approx 10^{-3}$ mbar. The waist of the infrared beam ($75 \pm 10 \mu\text{m}$ ($1/e^2$ level)) was measured to be slightly larger than that of the 400 nm beam [$(60 \pm 10) \mu\text{m}$]. The beam waists were carefully adjusted to coincide at the same position along the propagation axis. The cell was positioned approximately 2 mm after the focus to improve phase-matching conditions [20]. The maximal achievable peak intensities at the cell position were estimated to be $I_{1200} \approx 6.5 \times 10^{14} \text{ W/cm}^2$ and $I_{400} \approx 1.8 \times 10^{14} \text{ W/cm}^2$. After passing through the cell, the driving beams were blocked by a 200-nm-thick aluminium foil, and the transmitted XUV radiation was directed to an XUV spectrometer based on a 1 m toroidal mirror and a 600 grooves/mm plane grating (for details, see Refs. [10,28]), schematically shown in Fig. 1(b). The spectrally resolved XUV radiation was detected using a microchannel-plate-based detector (MCP) with a phosphor screen and recorded by a fast complementary metal-oxide semiconductor camera.

The relative time delay between the two-color components was precisely controlled by translating the fundamental beam with a motorized delay stage. The XUV radiation was observed only within the temporal window where the two driving pulses overlapped and the temporal overlap was optimized to maximize the harmonic signal. The IR beam passes through a half-wave-plate/polarizer pairs to control its inten-

sity. The intensity of the second-harmonic beam was adjusted using 1-mm-thick fused-silica beamsplitters with different split ratios in 10% increments. The harmonic spectra presented in this work were obtained by integrating the MCP images along the vertical axis.

IV. EXPERIMENTAL RESULTS

The experimental spectra obtained from the evaluation of the data sets are presented in this section together with the corresponding theoretical results, calculated using simulations based on the strong-field approximation. For the spectra shown in Fig. 2 (Ar) and Fig. 3 (Ne), the total intensity $I = I_{1200} + I_{400}$ of the two-color field was kept approximately constant at $I = (4\text{--}5.5) \times 10^{14} \text{ W/cm}^2$, while the ratio $\gamma = I_{1200}/I_{400}$ was varied. The generated XUV spectra exhibit the characteristic shape of the TCBC scheme. According to the selection rule (1), only odd harmonics are observed for all values of γ , while even harmonics are forbidden. This is a distinctive spectral feature of bichromatic HHG in the $\omega + 3\omega$ configuration, in contrast to the $\omega + 2\omega$ case, where the appearance of forbidden $n = 3q$ harmonics has been reported in several experiments. For the $\omega + 2\omega$ bicircular field, only the harmonics $n = 3q \pm 1$ are allowed according to the selection rule (1), so the $n = 3q$ harmonics are forbidden. The observations of $n = 3q$ harmonics for bichromatic elliptical polarization of the driving field have been attributed either to trapped population in Rydberg states during strong-field ionization [29], or to the breaking of the dynamical symmetry of the driving bicircular field [20].

We continued with another experimental series for Ar, in which the intensity of the IR driving field was kept constant while the intensity of the 400 nm field was varied. The results are shown in Fig. 4. Analyzing the results presented in Figs. 2–4, we draw the following conclusions. First, the overall harmonics cutoff (on the linear scale [30]) exhibits a

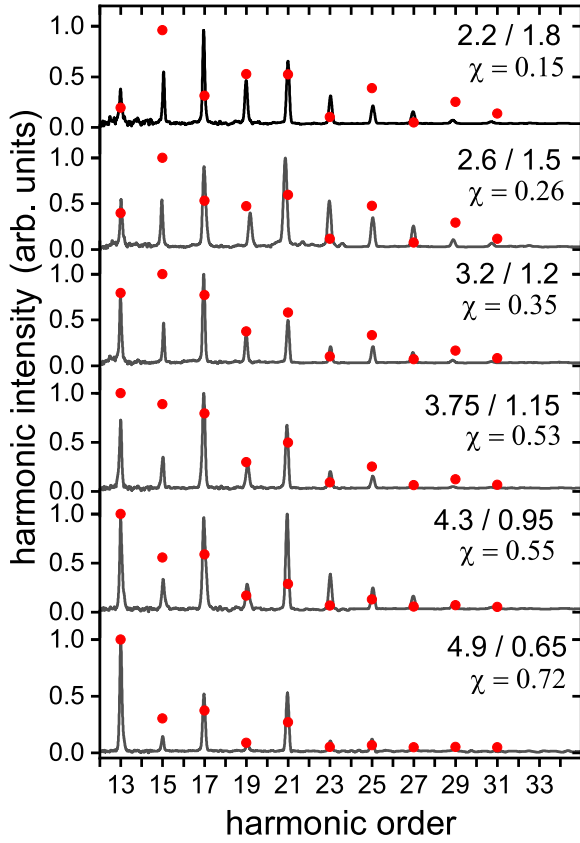


FIG. 2. XUV spectra of high harmonics generated in Ar using two-color bicircular driving fields of different intensity. The values in the insets indicate the driving-field intensities in units of $\times 10^{14}$ W/cm², with the first value corresponding to the 1200 nm IR beam and the second to the 400 nm beam. The experimental spectra are shown by the black solid curves, while the red filled circles present simulated harmonic spectra obtained under the experimental conditions. The calculated values of the asymmetry parameter χ for the corresponding experimental spectra are shown in the legend.

dependence proportional to the total intensity $I = I_{1200} + I_{400}$, but with a more efficient contribution from the 3ω component. Second, the key parameter governing the suppression of the harmonics $n = 4q - 1$, $q = 3, 4, 5, \dots$, which have helicity $\varepsilon_n = 1$ of the 3ω driver, is the ratio $\gamma = I_{1200}/I_{400}$, with a clearly dominant IR component for $\gamma \geq 3$. Such behavior has already been reported for the $\omega + 2\omega$ configuration, where the intensity of specific harmonic lines was controlled by varying the relative intensities of the two driving fields [19,21]. In our experiments, we achieve the highest degree of suppression for $\gamma \geq 7$ in Ar and $\gamma \geq 4$ in Ne. We quantify the degree of chiral control by calculating the spectral asymmetry in the form

$$\chi = (I_{4q+1} - I_{4q-1}) / (I_{4q+1} + I_{4q-1}), \quad (5)$$

where I_{4q+1} and I_{4q-1} denote the integrated intensities of all harmonics of the corresponding orders. The spectra are obtained integrating the MCP images along the vertical axis, within the range of interest for the relevant signal of corresponding harmonic, as described in Sec. III. The values of χ indicated in Figs. 3 and 4 were calculated for harmonics

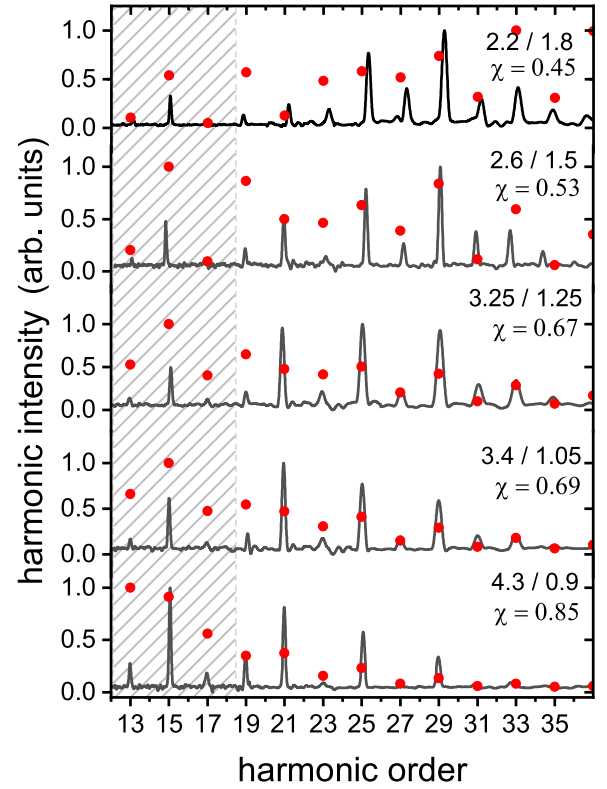


FIG. 3. Same as in Fig. 2, but for the XUV spectra generated in Ne. The shaded area indicates the spectral region where the dominance of the harmonics is reversed.

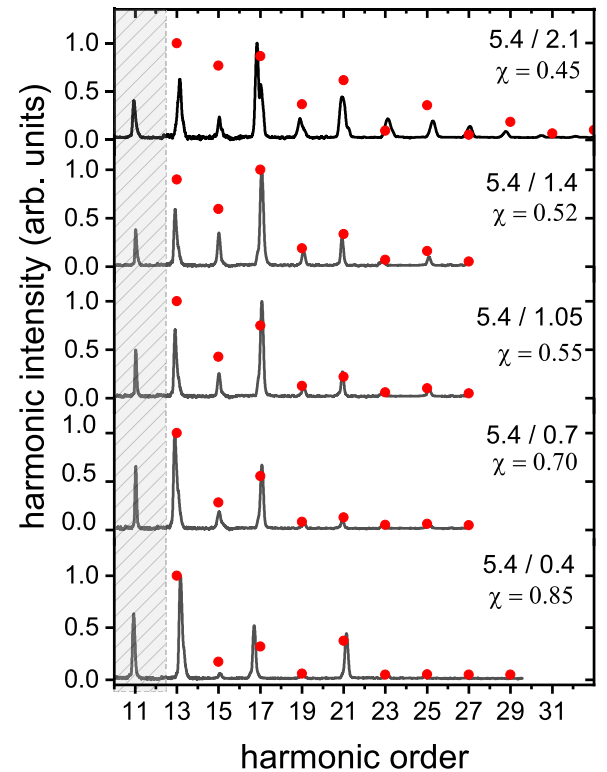


FIG. 4. Same as in Fig. 2 for Ar, but with the intensity of the ω component kept fixed.

H13 and higher for Ar spectra and H19 and higher for Ne spectra, corresponding to nonshadowed area in figures. Our experimental results demonstrate that substantial control can be achieved in the near-cutoff region. In particular, the suppression of the $4q - 1$ harmonics can be tuned over a wide range, down to the level less than 0.1 (corresponding to $\chi = 0.85$) of the intensity of the neighboring $4q + 1$ harmonics, providing a relatively straightforward means for efficiently controlling the ellipticity of the attosecond bursts. It is worth noting that the calculated values of the spectral asymmetry parameter χ represent an upper limit for spectral chirality, under the assumption that the contributing high harmonics are circularly polarized without any unpolarized component. A realistic estimate of spectral chirality requires incorporating the precise values of the harmonic ellipticity and polarization state.

As mentioned above, a remarkable feature of the $\omega + 3\omega$ configuration is the strong prohibition against the generation of even-order harmonics. Theory predicts that deviations of the driving field polarization from pure circular $\varepsilon = 1$ toward elliptical $0 < \varepsilon < 1$ do not lift the selection rules that forbid even harmonics. This behavior contrasts with the $\omega + 2\omega$ case, where the harmonics of order $3q$, forbidden for bicircular case, appear in the spectrum as soon as $\varepsilon < 1$. Their emergence is attributed either to modified selection rules [23] or to the opening of generation channels due to spin angular momentum conservation [24]. We validate this prediction experimentally by performing a series of measurements in a straightforward configuration where the ellipticity of the 400 nm field is fixed at its maximum available value, $\varepsilon_3 \simeq 0.93$, while the ellipticity ε_1 of the IR field is tuned over a wide range. The results are shown in Figs. 5 and 6. The ionization potential of Ar is 15.76 eV, while for Ne it is 21.56 eV. This explains the higher intensities used in Fig. 6. As predicted by the selection rules, even harmonics do not appear in the spectrum as shown in Figs. 5 and 6. In contrast, the appearance of forbidden $3q$ harmonics is routinely observed in the $\omega + 2\omega$ configuration in different experiments when the driving-field polarization is varied from circular toward elliptical [20,24,31]. This effect can lead to spectral overlap with neighboring orders, particularly in the cutoff region, thereby mixing their polarization properties. As a consequence, the harmonic ellipticity associated with emission over the full pulse duration is significantly reduced, accompanied by a rapid drop in ellipticity and increased depolarization [26]. Therefore, the commonly proposed simple method for controlling harmonic ellipticity for applications [32] is not free from these drawbacks. The remarkable robustness of the $\omega + 3\omega$ scheme against the generation of forbidden harmonics, which is in accordance with the selection rule (4) for bichromatic elliptically polarized driving field, offers a clear advantage for improving harmonic ellipticity. Furthermore, we observe a strong dependence of the spectral asymmetry on the ellipticity ε_1 of the IR driving field. As the IR field ellipticity decreases, the intensity of the $4q + 1$ orders remains approximately constant, while the intensity of the $4q - 1$ harmonic increases, thereby modifying the spectral asymmetry. Consequently, the harmonic ellipticity can be fully controlled by adjusting the ellipticity of the IR driver, without detrimental contributions from forbidden harmonics.

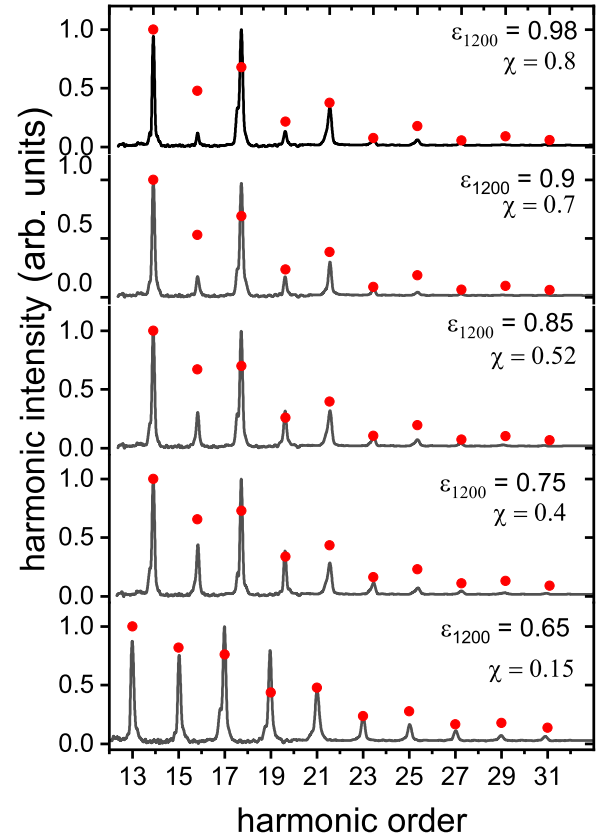


FIG. 5. XUV spectra of high harmonics generated in Ar for $I_{1200}/I_{400} = (4.5/1.2) \times 10^{14} \text{ W/cm}^2$. The black solid curves show experimental spectra, while the red solid circles represent the simulated harmonic spectra obtained under the experimental conditions. The values in the legends indicate the ellipticity ε_1 and the calculated asymmetry parameter χ .

V. THEORETICAL RESULTS

The theoretical results presented in proceeding section are obtained using the method described in Ref. [33]. In this approach, the HHG spectra are simulated by performing incoherent averaging of fixed-intensity SFA results, over the laser intensity distribution in the focus. By the SFA results we refer to the calculation of the T -matrix element, which determines the harmonic intensity. This requires numerical integration over the ionization and recombination times of the product of the corresponding dipole matrix elements and an exponential factor with the strong-field-dependent action. The dipole matrix element is between the ground-state wave function, represented in the form of atomic orbitals obtained by solving the Roothan-Hartree-Fock equations in terms of Slater-type orbitals, and the laser-dressed continuum Volkov state. For the Ar and Ne atoms considered here, the ground state is a p state (orbital quantum number is $\ell = 1$). Consequently, the T -matrix element is obtained as a coherent sum of the contributions corresponding to the magnetic quantum number $m = \pm 1$ [34]:

$$T_n = \sum_m \int_0^T dt_{\text{rec}} \hat{\mathbf{e}}_k^* \cdot \mathbf{d}_m(t_{\text{rec}}) e^{i n \omega t_{\text{rec}}} / T, \quad (6)$$

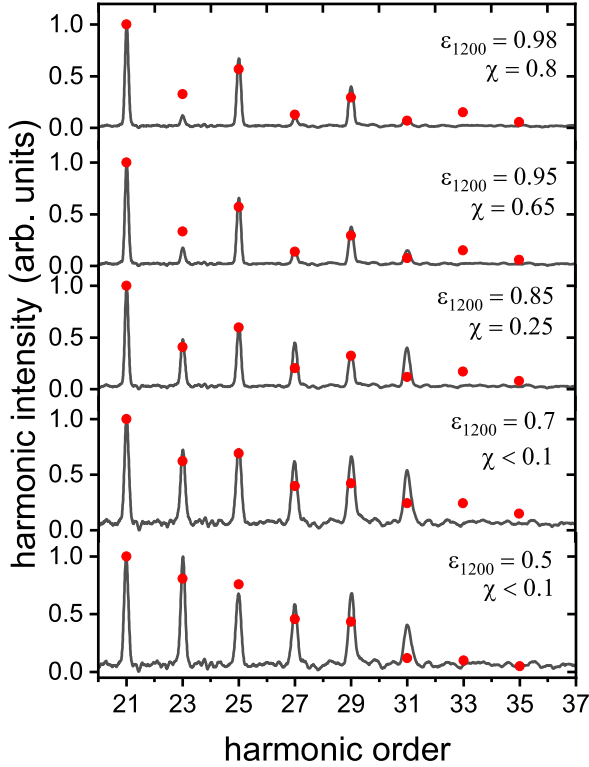


FIG. 6. Same as in Fig. 5, but for Ne and $I_{1200}/I_{400} = (6.5/1.5) \times 10^{14} \text{ W/cm}^2$.

where $\omega = 2\pi/T$ is the fundamental laser-field frequency, $\hat{\mathbf{e}}_k$ is the complex harmonic-field polarization vector, and the T -matrix element T_n is the Fourier component of the recombination-time-dependent dipole $\mathbf{d}_m(t_{\text{rec}})$, which is calculated by numerical integration over the ionization time t_{ion} , for particular value of the ground-state magnetic quantum number m . The n th harmonic intensity is $P_n \propto (n\omega)^4 |T_n|^2$.

Figure 7 shows an example of the fixed-intensity SFA calculations for the Ar atom, which has an ionization potential $I_p = 15.76 \text{ eV} = 15.25\omega$. The driving field is bicircular, with component helicities $\varepsilon_1 = -1$ and $\varepsilon_3 = 1$, and fixed component intensities $I_{1200} = 3.4 \times 10^{14} \text{ W/cm}^2$ and $I_{400} = 2.0 \times 10^{14} \text{ W/cm}^2$. According to the selection rule (1), the allowed harmonics are $n = 4q \pm 1$, $q = 1, 2, 3, \dots$, with helicities $\varepsilon_n = \mp 1$. Two spectral regions can be distinguished: (1) the low-energy harmonics, where experimental data are available, and (2) the high-energy plateau and cutoff region, where the semiclassical model of Refs. [17,18,23,34–37] provides an accurate description [38]. The SFA spectra in Fig. 7(a) show that harmonics with positive helicity dominate for $n \leq 15$ and $n \geq 39$. A pronounced intensity asymmetry appears between the $\varepsilon_n = -1$ and $\varepsilon_n = +1$ harmonics, especially for $n = 21, 25, 29, 33$, all of which have $\varepsilon_n = -1$. This asymmetry can be understood by analyzing the partial $m = \pm 1$ contributions to the harmonic intensity, shown in Figs. 7(b) and 7(c). For harmonics with $\varepsilon_n = -1$, the $m = +1$ channel dominates at low orders ($n \leq 21$), whereas at higher orders, the $m = -1$ channel becomes dominant (with the exception of the cutoff region $n > 53$, where the intensity decreases exponentially). Conversely, for harmonics with $\varepsilon_n = 1$ in Fig. 7(c),

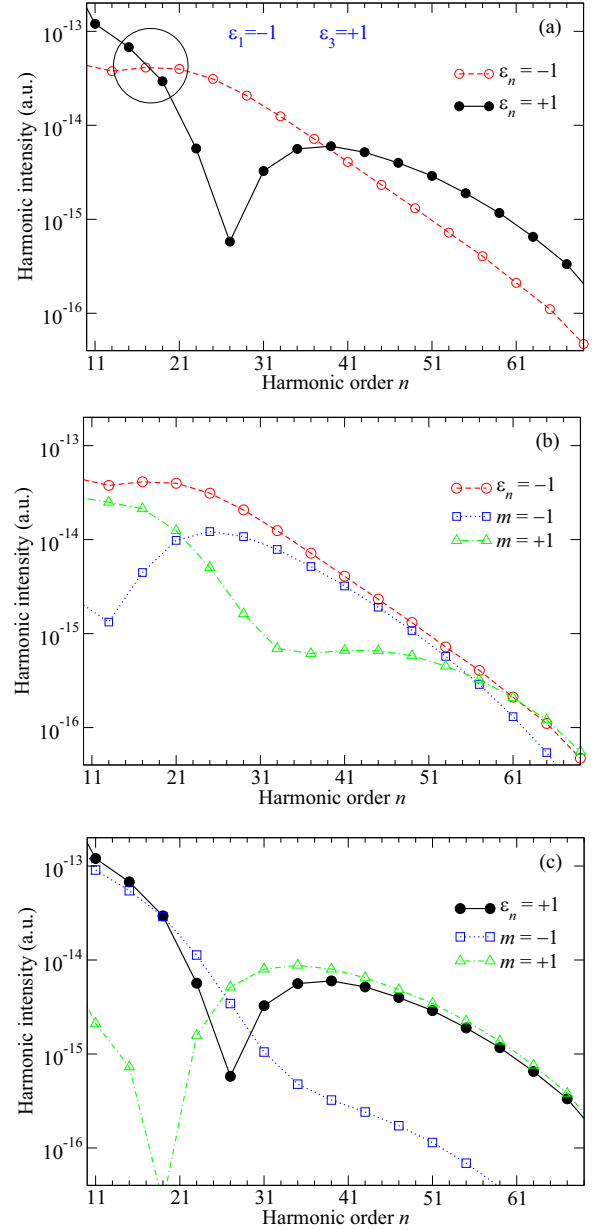


FIG. 7. Harmonic intensity as a function of harmonic order for HHG in Ar driven by a bicircular field with parameters $\varepsilon_1 = -1$, $\varepsilon_3 = +1$, $I_{1200} = 3.4 \times 10^{14} \text{ W/cm}^2$, and $I_{400} = 2.0 \times 10^{14} \text{ W/cm}^2$. Panel (a) shows the intensities of harmonics with helicities $\varepsilon_n = \pm 1$. Panels (b) and (c) display the partial contributions of the $m = \pm 1$ channels for harmonics with $\varepsilon_n = -1$ and $\varepsilon_n = 1$, respectively.

the $m = -1$ contribution dominates at low orders $n \leq 23$, while the $m = +1$ contribution becomes dominant throughout the plateau and cutoff region. The coherent sum of the two m contributions to the harmonic-emission T -matrix element (6) produces the spectra shown in Fig. 7(a). Overall, our analysis shows that for $n < 33$ the $m = -1$ channel provides the dominant contribution to the harmonic intensity.

The spectra shown in the right-hand panels of Figs. 2–6 are SFA simulations performed using incoherent focal-intensity averaging with the same field parameters as in the experiments. The agreement is notable, both in the overall spectral

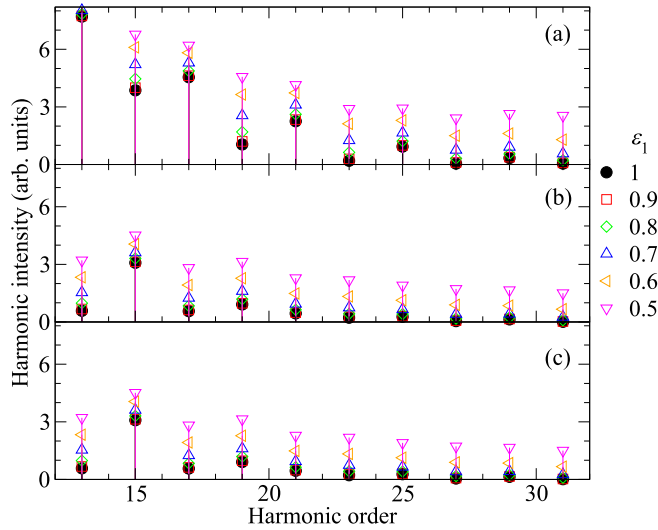


FIG. 8. Result of the SFA simulation for Ar driven by a bicircular field with $I_{1200} = 4.5 \times 10^{14}$ W/cm², $I_{400} = 1.2 \times 10^{14}$ W/cm², $\varepsilon_3 = -1$, and variable ε_1 . Panel (a) shows the harmonic intensities. Panels (b) and (c) display the partial contributions of the $m = +1$ and $m = -1$ channels, respectively.

shape and in the dependence on the driving-field intensity and ellipticity. Another characteristic feature, observed experimentally and reproduced by the simulations, is the reversal in dominance from the harmonics with $n = 4q - 1$ orders toward harmonics with $n = 4q + 1$ orders at the low-energy edge of the spectrum (shadowed regions in Figs. 3 and 4). For Ar, this reversal occurs around H11–H13, and for Ne around H17–H19, indicating a clear dependence on the ionization potential of the target medium. This behavior is also reproduced qualitatively in the theoretical spectra of Fig. 7(a), at the location marked by the circle.

Results of the simulation with the ε_1 as a parameter are summarized in Fig. 8. Most noteworthy are the results for the $n = 4q + 1$ harmonic orders H13, H17, and H21, co-rotating with 1200 nm driver. For each individual $m = +1$ or $m = -1$ contribution, the intensities of these harmonics are lower than those of the neighboring $n = 4q - 1$ harmonics of opposite helicity, as shown in Figs. 8(b) and 8(c). However, the total intensities in Fig. 8(a) are unexpectedly higher, most prominently for H13. This indicates that the $m = +1$ and $m = -1$ channels are in phase for both the x and y components, so their contributions add coherently and interfere constructively. This interpretation is confirmed by calculating the phases of the corresponding T -matrix-element components for $m = +1$ or $m = -1$ shown in Fig. 9. For $\varepsilon_1 = 1$, the phases of the T -matrix-element components are nearly identical for the harmonics H13, H17, H21, H25, and H29. For the remaining harmonics, the phases differ, with the exception of $n = 15$. When $\varepsilon_1 = 0.5$, the phases of the two m components are close only for the x component and only for harmonic orders $n < 27$; this is particularly evident for H13, H17, H21, and H25.

A more insightful semiclassical interpretation of the role of the magnetic quantum number m and the associated electric ring current will be provided after introducing the

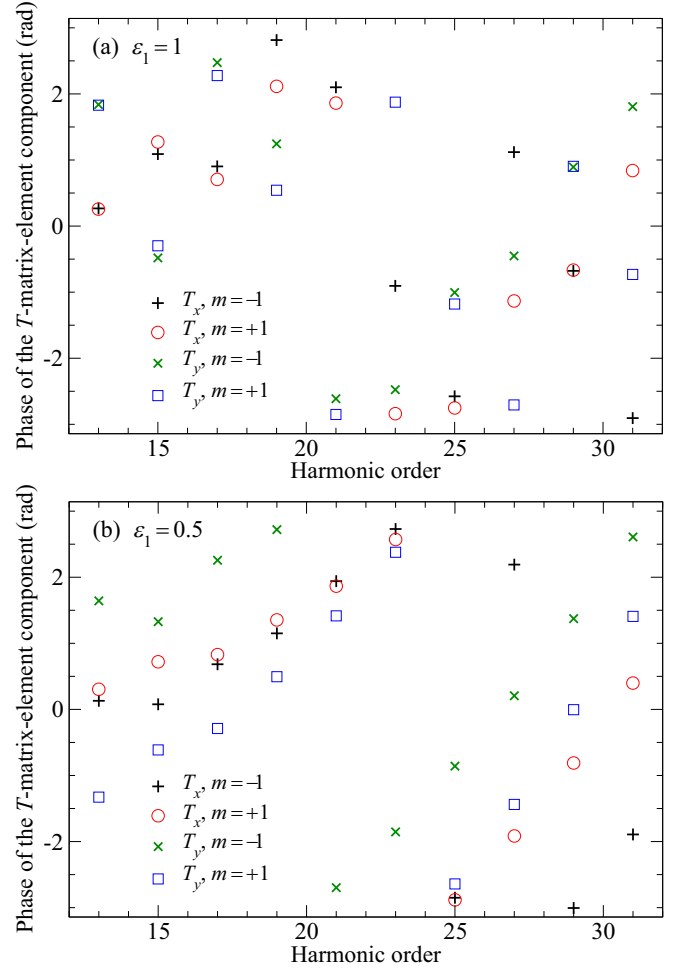


FIG. 9. Phase of the T -matrix-element components of the harmonics calculated for (a) $\varepsilon_1 = 1$ and (b) $\varepsilon_1 = 0.5$. Other parameters are as in Fig. 8.

quantum-orbit theory. The integral over the ionization time t_{ion} and the recombination time t_{rec} can be evaluated approximately using the saddle-point method and quantum-orbit theory [35,39]. The solutions of the corresponding saddle-point equations are classified by a multiindex (α, β, μ) [40], with each solution defining a quantum orbit that determines a specific electron trajectory. The index μ approximately measures the electron travel time in units of the optical period T , while the index β enumerates the solutions within the laser-field period corresponding to the index μ . Finally, the index α identifies the “short” ($\alpha = +1$) and “long” ($\alpha = -1$) orbits of the pair of solutions characterized by the indices β and μ (the nomenclature of short and long orbits was first introduced in Ref. [3]). The dominant contribution to the T -matrix element arises from the short quantum orbits, which are characterized by the index $\mu = 0$. Figure 10 illustrates the (α, β, μ) classification for the parameters of Fig. 7, restricted to the case $\mu = 0$. The harmonic photon energy $n\omega$, in units of the ponderomotive energy U_p , is shown as a function of the electron travel time $\text{Re } \tau = \text{Re } (t_{\text{rec}} - t_{\text{ion}})$, given in units of the optical period $T = 2\pi/\omega$. The resulting curves appear in pairs labeled by $\alpha = \pm 1$, with each pair exhibiting its own characteristic maximum energy (cutoff).

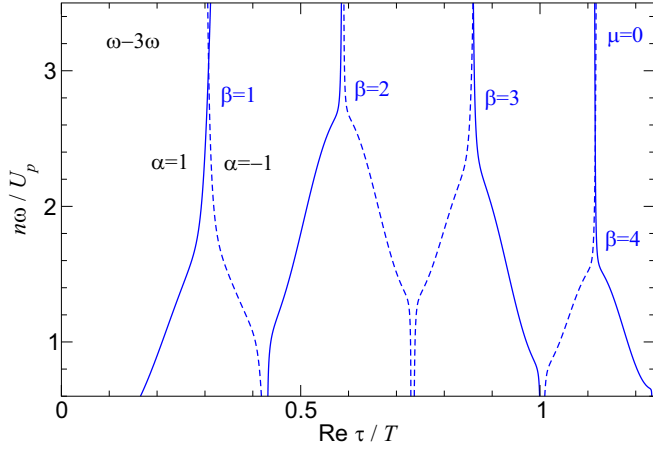


FIG. 10. Saddle-point solutions (α, β, μ) for $\mu = 0$, obtained for the same parameters as in Fig. 7.

For the present parameters, there are $r + s = 4$ such pairs, indexed by $\beta = 1, 2, 3, 4$. Among these, the dominant contribution to the HHG T -matrix element originates from the shortest quantum orbit $(\alpha, \beta, \mu) = (1, 1, 0)$. In what follows, we use this orbit to provide a physical interpretation of the experimental observations and theoretical simulations presented in Figs. 2–6.

In Fig. 11, we present our semiclassical quantum-orbit analysis for the harmonic $n = 25$. The upper-left panel shows that, for the harmonic H25, the electric field at the ionization time (denoted by I) is relatively strong and evolves along an almost linear segment up to the recombination time (denoted

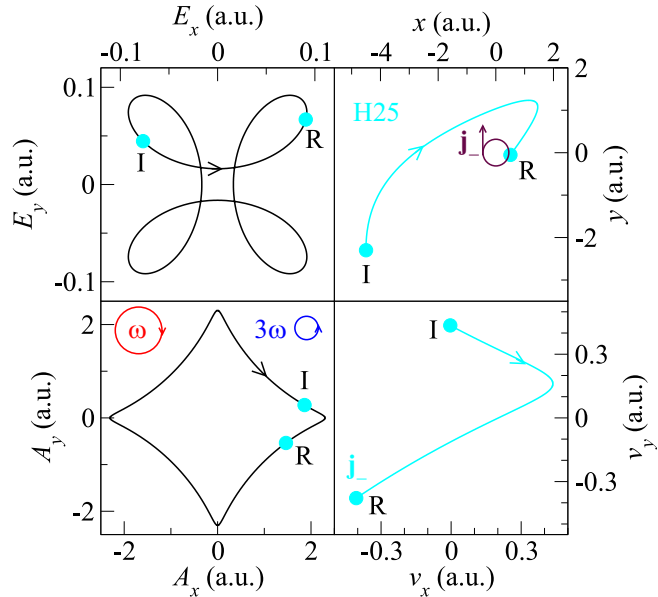


FIG. 11. Bicircular electric-field vector (upper-left panel) and vector potential (lower-left panel) in the polarization plane. The right panels show the electron trajectory (upper) and velocity (lower) between the ionization (I) and recombination (R) times corresponding to harmonic order $n = 21$. The associated electron probability current is also shown. Atomic and laser parameters are identical to those used in Fig. 7.

by R). The presence of such near-linear field segments is generally crucial for efficient generation of high-order harmonics. The lower-left panel illustrates the corresponding evolution of the vector potential. The insets depict the polarization of the two component fields; the smaller blue circle indicates the lower intensity of the 3ω component of the bicircular field. The field, vector potential, electron coordinate, and velocity at the ionization and recombination times are marked by filled cyan circles. The upper right panel shows the electron trajectory between ionization and recombination. The electron is born a few atomic units away from the nucleus (located at the origin) and returns to it along the cyan path. The trajectory is short and differs from those associated with higher harmonic-photon energies (compare Fig. 11 in Ref. [37]). After emerging at the tunnel exit, the electron initially moves in the positive y direction. A positive initial velocity t_{ion} is required to compensate the positive value of the field component E_y , ensuring that the electron can return to the origin and recombine. For $t > t_{\text{ion}}$, the force $-\mathbf{E}(t)$ causes v_y to decrease while v_x increases. When the x component of the electric field changes sign, v_x begins to decrease, while v_y continues to decrease until recombination, at which point the electron has sufficient kinetic energy to emit the $n = 25$ harmonic. As shown in Fig. 7(b), the probability of ionization from the atomic ground state having $m = -1$ is higher for $n = 25$. The corresponding electric ring current $\mathbf{j}_m = \mathbf{j}_- \propto m\hat{\mathbf{e}}_\varphi$ (with $\hat{\mathbf{e}}_\varphi$ the unit vector in spherical coordinates) is displayed in the right-hand panels of Fig. 11. Since $x(t_{\text{ion}}) < 0$, the y component of $\mathbf{j}_-$ is positive, making it more likely for the electron to emerge with a large positive $v_y(t_{\text{ion}})$ than in the $m = +1$ case. This explains the stronger harmonic intensity for $m = -1$ observed in Fig. 7. Furthermore, recombination is more probable when the bound state corotates with the circular field of the emitted harmonic, i.e., when $m = -1$ and $\varepsilon_n = -1$, which is indeed the case for $n = 25$. A similar analysis applies to other harmonics. For example, for $n = 15$ the trajectory resembles that in Fig. 11, but the electron does not encircle the origin, instead $y < 0$ throughout the excursion. In this case, the electron returns with a positive velocity component $v_x(t_{\text{rec}}) = 0.4$ a.u.

In conclusion, our theoretical analysis leads to the following interpretation of the experimental results. Both Ar and Ne have a p -ground state, and ionization and recombination in the HHG process can occur for either the $m = +1$ or $m = -1$ state. Consequently, the T -matrix element is a coherent superposition of these two contributions, Eq. (6). Their interference varies across different harmonic ranges, as illustrated in Fig. 7 where the partial m contributions are also shown. This interpretation is further supported by Figs. 8 and 9, which demonstrate that for certain harmonics and laser parameters the T -matrix contributions from the $m = +1$ and $m = -1$ channels become in phase. Additional insight is obtained using quantum-orbit theory and analyzing the electron trajectory associated with the dominant, shortest quantum orbit (see Fig. 11). The enhanced intensity of a specific harmonic for a given value of the ground-state magnetic quantum number m is linked to the corresponding electric ring current, which causes a more likely emergence of the electron with the velocity required for emission of that harmonic. Moreover, recombination is more probable when the bound state corotates

with the circular polarization of the emitted harmonic. Such an analysis does not apply to atoms with an s ground state and $m = 0$, such as He.

VI. XUV POLARIMETRY

Exact knowledge of the polarization state of radiation is crucial for a quantitative understanding of the underlying physical processes in a chiral environment, because chiral effects are proportional to the degree of circular polarization. The polarization of light is completely characterized by the Stokes vector $\vec{S}$ whose elements are four Stokes parameters S_0, S_1, S_2 , and S_3 , defined by [41,42]

$$\vec{S} = \begin{pmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{pmatrix} = \begin{pmatrix} I_x + I_y \\ I_x - I_y \\ I_{+45^\circ} - I_{-45^\circ} \\ I_R - I_L \end{pmatrix}, \quad (7)$$

where I_x, I_y , and $I_{\pm 45^\circ}$ are the intensities measured after a polarizer aligned along x, y , and $\pm 45^\circ$ directions, respectively, and I_R (I_L) is the intensity measured along the $+45^\circ$ (-45°) direction after a $\pi/2$ retardation. In this work, we define the light polarized along the x direction as p polarized and light polarized along the y direction as s polarized. For convenience, we use the normalized Stokes parameters, defined by $\tilde{S}_i = S_i/S_0, i = 1, 2, 3$. Moreover, the Stokes parameters allow one to describe partially polarized light, with the degree of polarization

$$\eta = \sqrt{\tilde{S}_1^2 + \tilde{S}_2^2 + \tilde{S}_3^2} \leq 1, \quad (8)$$

where $\eta = 1$ for fully polarized light.

To perform classical measurement of the Stokes parameters, a phase plate that introduces a known phase retardation between the two electric-field components, and an analyzer that preferentially transmits a single linearly polarized component have to be applied. A polarization measurement also requires independent rotation of both elements about the direction of light such that the polarization state is modified by the first element and subsequently analyzed by the second. In the standard measurement scheme, known as rotation-analyzer (polarizer) ellipsometry, the transmitted light intensity is recorded when the position of wave plate (polarizer) is constant, while the transmission axis of the linear polarizer (wave plate) is rotated around the propagation axis. Fitting a model function for the transmitted intensity, with the Stokes vector elements as parameters, to the experimental polarization curve [10,43–45], or determining the coefficients by Fourier transformation [46], yields the normalized Stokes parameters as well as the polarizing properties of the optical elements used. The ellipticity, orientation angle, and

degree of depolarization can then be obtained from standard relations.

In this work, we refer to the Mueller-matrix formalism, which describes the interaction of the input Stokes vector $\vec{S}_{\text{in}}$ with an optical element, to define a model for the transmitted intensity. The Stokes vector $\vec{S}_{\text{out}}$ of the light after passing through the setup is then given by

$$\vec{S}_{\text{out}} = R(\alpha)M_{\psi_a, \Delta_a}R(-\alpha)R(\beta)M_{\psi_p, \Delta_p}R(-\beta)\vec{S}_{\text{in}}, \quad (9)$$

where $R(\beta)M_{\psi_p, \Delta_p}R(-\beta)$ and $R(\alpha)M_{\psi_a, \Delta_a}R(-\alpha)$ describe the action of the rotating polarizer and rotating analyzer, respectively. The first element of the $\vec{S}_{\text{out}}$ Stokes vector, $S_{0,\text{out}}$, is proportional to the transmitted intensity I_{out} . The measured transmitted intensity together with the model function is then used to reconstruct the Stokes vector of the incident field.

Precise knowledge of the phase retardation introduced by each element of the setup, as well as their reflective properties, is crucial for a reliable evaluation. In the UV, visible, and near-IR spectral ranges, numerous well-characterized polarizing elements operating in transmission offer good opportunities for routine measurements. In the VUV, where no transparent materials exist, one must instead use multiple-reflection systems whose incidence angles and coatings are optimized to achieve effective polarizer or analyzer performance [45]. The major drawback of such multireflection systems is their very low overall transmission, which can be as little as a few percent.

Here, we experimentally apply a modified version of rotating-analyzer ellipsometry to determine the ellipticity, helicity, and degree of polarization of the generated high harmonics. For the polarimetric measurements, the MCP detector [Fig. 1(b)] located after the VUV spectrometer was removed, and a polarizer [Fig. 1(c)] was inserted into the beam path at the image plane of the toroidal mirror. The analyzer is based on a single reflection from a GaAs crystal with (001) orientation, which provides a highly suitable extinction ratio r_p/r_s in the spectral range of the generated harmonics and is mechanically more robust than unprotected metal surfaces. The GaAs plate was set at an incidence angle of approximately 40° , corresponding to the Brewster angle for radiation near the midpoint of the spectral range of interest.

The XUV radiation generated in the gas jet propagates toward the GaAs polarimeter through the XUV spectrometer, undergoing two reflections from the gold-coated surfaces of the toroidal mirror and the grating. Each of these reflections introduces not only an intensity modification determined by the extinction ratio, but also a phase shift between the p and s components. Mathematically, this action is described by a 4×4 Mueller matrix, which in its most general form is given by

$$M_{\psi, \Delta} = \begin{pmatrix} 1 & -\cos 2\psi & 0 & 0 \\ -\cos 2\psi & 1 & 0 & 0 \\ 0 & 0 & \sin 2\psi \cos \Delta & \sin 2\psi \sin \Delta \\ 0 & 0 & -\sin 2\psi \sin \Delta & \sin 2\psi \cos \Delta \end{pmatrix}, \quad (10)$$

where $\psi = \arctan(|r_p|/|r_s|)$ and Δ is the introduced phase shift. This matrix is also applied to the GaAs analyzer, together with the rotation Mueller matrix:

$$R(\alpha) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2\alpha & \sin 2\alpha & 0 \\ 0 & -\sin 2\alpha & \cos 2\alpha & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \quad (11)$$

where α is the azimuthal rotation angle of the analyzer. The Stokes vector after passing through the entire setup then is

$$\vec{S}_{\text{out}} = R(\alpha)M_{\psi_a, \Delta_a}R(-\alpha)M_{\psi_s, \Delta_s}\vec{S}_{\text{in}}, \quad (12)$$

where M_{ψ_s, Δ_s} describes the spectrometer components and $R(\alpha)M_{\psi_a, \Delta_a}R(-\alpha)$ represents the rotating analyzer. Since the MCP detector is not phase sensitive, we set the phase shift introduced by the analyzer to $\Delta_a = 0$. The MCP detector measures the total light intensity $I \equiv I(\alpha; \vec{S}_1, \vec{S}_2, \vec{S}_3)$, which corresponds to the first component of $\vec{S}_{\text{out}}$, yielding

$$\begin{aligned} I \propto & 1 - \vec{S}_1 \cos 2\psi_s \\ & + \cos 2\psi_a \cos 2\alpha (\vec{S}_1 - \cos 2\psi_s) \\ & + \sin 2\alpha \sin 2\psi_s (\vec{S}_2 \cos \Delta_s + \vec{S}_3 \sin \Delta_s). \end{aligned} \quad (13)$$

The experimental ellipticity ε and the ellipse orientation angle θ can also be expressed in terms of the components of the Stokes vector $\varepsilon = \tan[\arctan(S_3/\sqrt{S_1^2 + S_2^2})/2]$, $\theta = \arctan(S_2/S_1)/2$ [10].

Equation (13) contains the unknown parameters of the optical system ψ_s , Δ_s , and ψ_a . These can be determined by a least-squares fit to the measured data in a self-calibrating procedure, which enables simultaneous characterization of the incident light and determination of the polarizing properties of the optical elements used [44]. Alternatively, they can be obtained from the tabulated reflection coefficients of the corresponding materials. However, these values depend critically on the angle of incidence and on the optical quality of the reflecting surfaces. In this work, we determined the parameters ψ_a , ψ_s , and Δ_s , thereby experimentally calibrating our optical system using radiation with well-defined Stokes vectors. We exploit the property of HHG driven by a linearly polarized mid-IR laser beam to follow the polarization of the driving laser, whose polarization can be precisely controlled. The transmitted intensity was recorded for each harmonic as a function of the analyzer angle for four distinct linear polarization states of the high-harmonic radiation: p - and s -polarized light, $\vec{S}_{p/s} = (1, \pm 1, 0, 0)^T$ (T indicates the transpose), and polarization angles 45° [$\vec{S}_{45^\circ} = (1, 0, 1, 0)^T$] and -45° [$\vec{S}_{-45^\circ} = (1, 0, -1, 0)^T$].

The intensity of the transmitted beam was measured as a function of the analyzer's azimuthal angle over a full 2π rotation, with a step size of 5° . It is important to emphasize the necessity of performing measurements over the entire 2π range. The polarimetric curves recorded with the polarimeter must exhibit π symmetry, $I(\alpha) = I(\alpha + \pi)$ with, $\alpha \in [0, \pi]$. This symmetry provides a crucial indication of correct analyzer alignment, as its rotation axis must be strictly collinear with the propagation axis of the analyzed radiation. Any deviation from collinearity leads to a variation of the incidence angle α during rotation and results in asymmetric curves,

TABLE I. Parameters $|r_p|/|r_s| = \tan \psi$ and Δ_s evaluated for each individual harmonic. The indices a and s refer to the analyzer and the spectrometer, respectively.

	$\tan \psi_a$	$\tan \psi_s$	Δ_s (rad)
H17	0.418 ± 0.006	0.78 ± 0.04	2.19 ± 0.04
H19	0.320 ± 0.009	0.88 ± 0.03	2.31 ± 0.05
H21	0.374 ± 0.008	0.65 ± 0.02	2.38 ± 0.05
H23	0.392 ± 0.008	0.88 ± 0.03	2.29 ± 0.08

i.e., $I(\alpha) \neq I(\alpha + \pi)$. Consequently, performing only a π scan can cause poor convergence of the fitting procedure and significant errors in the evaluation of the Stokes components. The evaluated parameters of the setup are listed in Table I.

The present experimental geometry does not allow polarimetric measurements for harmonics H13 and H15, because of violation of the polarimeter collinearity condition at the edge of the grating's angular orientation. Using another grating with larger free dispersion range or rearrangement of the polarimeter position can help to meet the collinearity requirement for low order harmonics.

In the next step, we switched to generating high harmonics in the TCBC scheme using circularly polarized drivers. The harmonics were generated in Ar with the driver configuration corresponding to the optimal spectral asymmetry, as shown in Figs. 2 and 4. Typical polarimetric curves obtained under these conditions are shown in Fig. 12.

The curve-fitting procedure (e.g., *curvefit* package in Python) can yield a unique solution for the Stokes vector only if the degree of polarization η is assumed to be known. Because unpolarized and circularly polarized radiation cannot be distinguished using a single analyzer, the polarimetric curves can be fitted with different combinations of η and ε , and therefore do not provide a unique solution. To overcome this ambiguity, a phase retarder must be inserted into the beam path, and polarimetry must be performed for several compensator settings with well-defined phase shifts. A unique solution can then be obtained by recording at least two polarization curves. However, the limited space in the current setup does not allow the insertion of an XUV compensator, which typically consists of a stack of three or more mirrors. An additional drawback is the significant loss of transmitted radiation caused by the multiple partial reflections in such a device.

We have proposed an alternative technique [10] that exploits the polarization properties dictated by the symmetry of the driving fields in the TCBC approach for chiral HHG. In this scheme, the helicity of each harmonic can be reversed simply by interchanging the helicities of the driving fields. Under identical generation conditions for both helicities, it is reasonable to assume that changing the sign of the helicity affects only the sign of the Stokes parameter S_3 in Eq. (13).

Figure 12 presents the polarimetric curves of harmonic H21 recorded for positive and negative helicities. The same measurements were performed for harmonics H17, H19, and H23. Reliable measurements for harmonics of order higher than H23 were not achieved due to their low intensity. The polarimetric curves in Fig. 12 demonstrate that, after

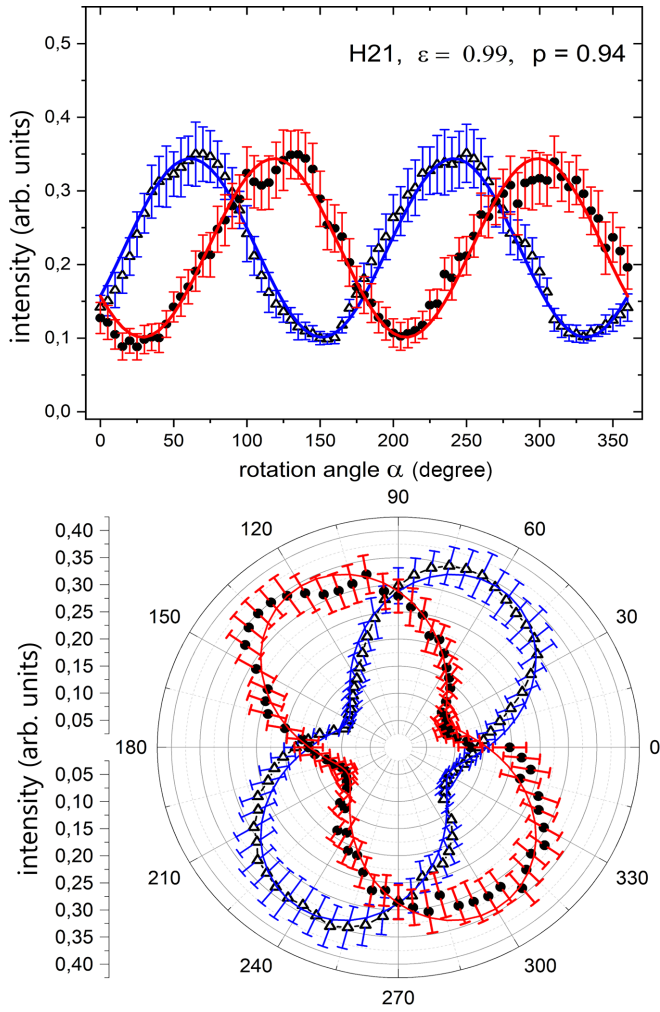


FIG. 12. Polarimetric curves for H21 recorded for positive (red line and solid symbols) and negative (blue line and open symbols) helicity. The lines represent fits using Eq. (13), while the symbols show the experimental data.

propagation through the spectrometer, the polarization at the analyzer position is no longer circular. By reversing the optical path and applying Eq. (12), one can infer the original state of polarization before the spectrometer.

We evaluated the Stokes parameters by fitting the model function coupled for both positive and negative helicity, i.e., $I_+(\alpha; \tilde{S}_1, \tilde{S}_2, +\tilde{S}_3)$ and $I_-(\alpha; \tilde{S}_1, \tilde{S}_2, -\tilde{S}_3)$ on basis of Eq. (13), to the combined in the same way experimental data for each harmonics. The chi-square value X^2 , which quantifies how well a given model function or parameter set reproduces the measured data, was calculated for the datasets.

The results of the Stokes-vector evaluation, together with the corresponding ellipticity and degree of polarization, are summarized in Tables II and III. The most intense harmonics, H17 and H21, whose helicities follow that of the IR driver and which determine both the spectral asymmetry and the energetic content of the pulse, are found to be nearly circularly polarized and exhibit a very high degree of polarization. Table III lists only the absolute values of ε , since the helicity can be either positive or negative depending on the specific combination of the driving fields. The X^2 values for H17, H19,

TABLE II. Components of the normalized Stokes vector and the corresponding likelihood estimates in the form of X^2 , retrieved from the evaluation of the polarimetry measurements.

	$\tilde{S}_1$	$\tilde{S}_2$	$\tilde{S}_3$	X^2
H17	0.041	0.026	-0.999	78 ± 2
H19	-0.072	-0.106	0.279	40 ± 2
H21	0.015	-0.014	-0.966	82 ± 2
H23	0.31	0.004	0.39	87 ± 25

and H23 are very close to the number of data points (72), providing confidence in the goodness of fit. In contrast, the weaker harmonics H19 and H23, which carry the opposite helicity, show a substantial unpolarized component despite their moderate ellipticity. The relatively small X^2 value for H19 can be attributed to the larger standard deviation of the measurements, caused by the reduced signal-to-noise ratio when the analyzer must operate at high gain to detect these strongly suppressed harmonics. Considering the dominant contribution of the $4q + 1$ harmonics, with their high ellipticity and high degree of polarization, and almost negligible contribution of the suppressed $4q - 1$ harmonics to the formation of the attosecond pulse train, we can conclude that highly chiral attosecond pulses ($\varepsilon > 0.85$) are successfully formed in the time domain. The resulting chirality is expected to be only slightly lower than the spectral chirality χ within the corresponding spectral range. Appropriate spectral filtering within the range of interest will further support the generation of circularly polarized attosecond pulses.

VII. ELLIPTICITY: THEORETICAL APPROACH

The large unpolarized content and the relatively low ellipticity of the high harmonics corotating with the 3ω driver, as obtained from polarimetry, are unexpected and require additional analysis and clarification. As established in Sec. II, the dynamical symmetry is violated for an arbitrarily elliptically polarized bichromatic field, leading to the formulation of the general selection rule given by Eq. (3). It should be noted that this selection rule was derived by considering spin angular momentum and energy conservation in the conversion of many pump photons into a single harmonic photon [23,34], taking into account the decomposition of the driving bicircular field into four circularly polarized components. Equation (3) simplifies for ellipticities $-1 \leq \varepsilon_r \leq 0$ and $\varepsilon_s = +1$, as shown in Eq. (4). Substituting $r = 1$ and $s = 3$ into Eq. (3) for the experimentally relevant case ($\varepsilon_r = -1$, $0 \leq$

TABLE III. Ellipticity ε , orientation angle θ , and degree of polarization η calculated for the harmonics generated using the TCBC approach.

	ε	θ	η
H17	0.92 ± 0.02	$76^\circ \pm 14^\circ$	1.0 ± 0.02
H19	0.64 ± 0.04	$62^\circ \pm 6^\circ$	0.30 ± 0.05
H21	0.98 ± 0.03	$-28^\circ \pm 17^\circ$	0.94 ± 0.02
H23	0.48 ± 0.03	$0^\circ \pm 2^\circ$	0.49 ± 0.02

TABLE IV. Possible combination for the numbers of exchanged photons from the two-color elliptical driving laser field for harmonics generated with TCBC approach.

	(p_-, q_+, q_-)				
H17-	(2, 3, 2)	(5, 4, 0)	(-1, 2, 4)	(8, 5, -2)	...
H17+	(-1, 3, 3)	(2, 4, 1)	(-4, 2, 5)	(5, 5, -1)	...
H19-	(1, 3, 3)	(4, 4, 1)	(-2, 2, 5)	(7, 5, -1)	...
H19+	(1, 4, 2)	(4, 5, 0)	(-2, 3, 4)	(7, 6, -2)	...
H21-	(0, 3, 4)	(3, 4, 2)	(6, 5, 0)	(9, 6, -2)	...
H21+	(0, 4, 3)	(3, 5, 1)	(6, 6, -1)	(9, 7, -3)	...
H23-	(-1, 3, 5)	(2, 4, 3)	(5, 5, 1)	(8, 6, -1)	...
H23+	(-1, 4, 4)	(2, 5, 2)	(5, 6, 0)	(8, 7, -2)	...

$\varepsilon_s \leq 1$) yields the following selection rule:

$$n_{\pm} = 2(2p_- + 3q_-) \pm 3, \quad (14)$$

where we used Eq. (2) for $p_+ = 0$, and where p_- and q_- are arbitrary integers corresponding to the number of exchanged photons with frequencies ω and 3ω , respectively, and helicity -1 . We note that the allowed harmonic orders $n_{\pm}$ remain odd. The applied decomposition represents the driving laser field as a combination of one circularly polarized beam at frequency ω and two circularly polarized beams at frequency 3ω , having different intensities and opposite helicities. The number of exchanged photons of a given frequency and helicity contributing to a specific harmonic order n is governed by the selection rule in Eq. (14) and can be expressed as a set (p_-, q_+, q_-) , where p_- denotes the number of exchanged ω photons with helicity -1 , and q_+ and q_- denote the numbers of exchanged 3ω photons with positive and negative helicity, respectively, subject to the condition $q_+ = p_- + q_- \pm 1$. For a given harmonic order n , numerous such sets exist for both positive (n_+) and negative (n_-) helicities. Table IV lists all possible combinations of exchanged photons for the two-color driving field. The bold-highlighted sets, (5, 4, 0) for H17-, (4, 5, 0) for H19+, (6, 5, 0) for H21-, and (5, 6, 0) for H23+, are the only allowed channels for $\varepsilon_3 = +1$, since this condition enforces $q_- = 0$. Consequently, for $\varepsilon_1 = -1$ and $\varepsilon_3 = +1$, H19 and H23 acquire ellipticity $+1$, while H17 and H21 acquire ellipticity -1 , in agreement with the selection rule (1), which yields $n_{\pm} = 4q \mp 1$.

For elliptically polarized driver radiation, additional generation channels listed in Table IV become accessible and can participate in the harmonic-generation process. The total field of the n th harmonic, $\mathcal{E}_n$, can be presented as a coherent sum of two components, $\mathcal{E}_{n+}$ and $\mathcal{E}_{n-}$, carrying helicities $+1$ and -1 , respectively. Their interference produces an elliptically polarized n th harmonic. Although the selection rule of Eq. (14) specifies which channels are allowed, determining the exact weight of each contributing channel remains challenging. Consequently, only the total harmonic intensity, summed over all allowed channels, can be reliably compared with experiment.

Figure 13 presents numerical results for the configuration $\varepsilon_1 = -1$ and $0 \leq \varepsilon_3 \leq 1$. The harmonic intensities exhibit distinct trends as ε_3 adjusted toward $\varepsilon_3=1$: Harmonics corotating with the ω driver increase in strength, whereas

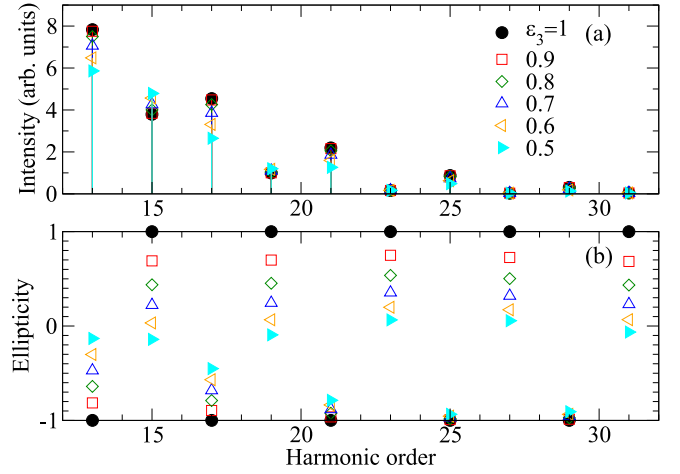


FIG. 13. XUV spectra of high harmonics generated in Ar for $I_{1200}/I_{400} = (4.5/1.2) \times 10^{14} \text{ W/cm}^2$ as a function of ε_3 . Panel (a) shows the simulated spectra, while panel (b) presents the corresponding harmonic ellipticities.

counter-rotating ones diminish. This behavior is consistent with experimental observations. A particularly notable feature is that for $\varepsilon_3 < 1$, the ellipticity of the harmonics corotating with the 3ω field, H15, H19, and H23, changes significantly, while the ellipticity of H13, H17, and H21 remains close to -1 .

To gain deeper insight into the experimental observations, numerical modeling was performed using the exact parameters of the bicircular field employed in the measurements. The simulation results are shown in Fig. 14. For the right-polarized harmonics ($\varepsilon_n < 0$) with orders $n_- = 4q + 1$ (H17, H21, H25, H29, ...), the contribution from the $m = -1$ component dominates. This leads to large ellipticities (circles), reaching nearly perfect circular polarization for H25, where $\varepsilon_{25} = -0.991$. In contrast, for the harmonics $n = 4q - 1$ (H15, H19, H23, H27, ...) the final polarization state is determined by a coherent superposition of the $m = -1$ and $m = +1$ components in the T -matrix. This mixing produces a pronounced minimum at H27, where $\varepsilon_{27} = 0.522$. The ellipticity of the left-polarized harmonics H19, H23, H27, and H31 (squares) remains close to 0.6. These results indicate that the coherent interplay between the $m = +1$ and $m = -1$ channels is the key factor shaping the final polarization state of the emitted harmonics. The ellipticities obtained from the numerical simulations for H19 and H23 (Table III) agree well with the values extracted from the polarimetric measurements. This agreement provides strong support for the necessity of a multielectron theoretical description to correctly interpret the experimental results [34]. The substantial unpolarized fraction observed for H19 and H23 can be attributed to the large number of open generation channels for these harmonics, whose interference with irregular phases (see Fig. 9) leads to depolarization.

VIII. CONCLUSIONS

We demonstrate the first successful implementation of the two-color bicircular scheme for generating circularly

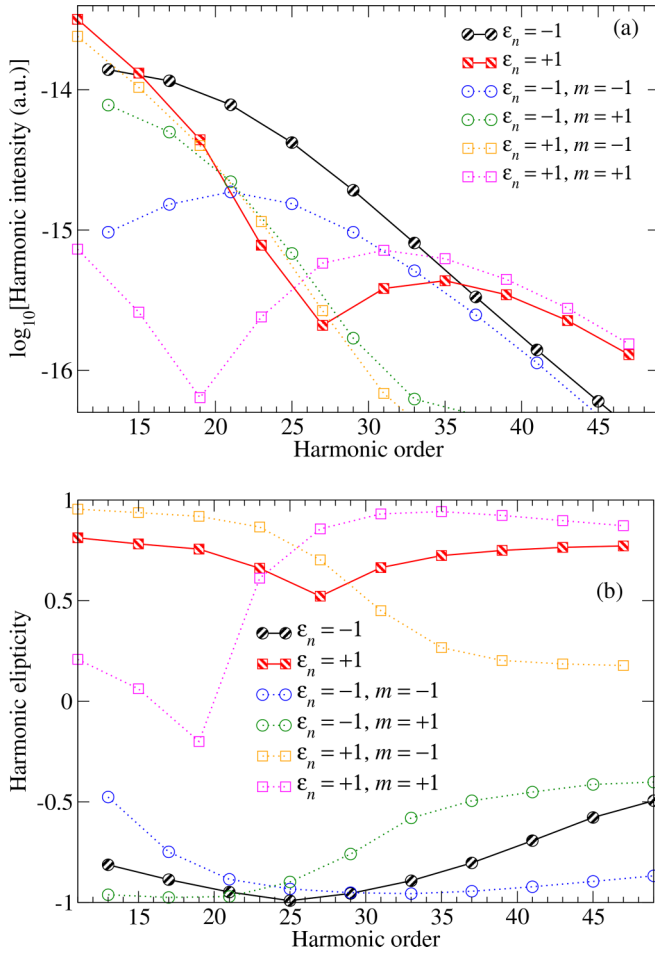


FIG. 14. Results of numerical modeling in Ar for a bicircular field with the following parameters: $\varepsilon_1 = -0.97$, $\varepsilon_3 = +0.93$, $I_{1200} = 3.4 \times 10^{14} \text{ W/cm}^2$, and $I_{400} = 2.0 \times 10^{14} \text{ W/cm}^2$. Panels (a) and (b) show the harmonic intensities and ellipticities, respectively, together with the partial contributions from the $m = \pm 1$ components.

polarized attosecond pulses using the $\omega + 3\omega$ (1200 nm + 400 nm) combination of the driving femtosecond laser fields. The generation process was simulated using an SFA-based numerical procedure with fixed intensity and an incoherent focal-averaging method. A key advantage of the $\omega + 3\omega$ configuration is the strong suppression of even-order harmonics, observed in both the simulated and experimental spectra, even for driving fields with reduced ellipticity. This suppression paves the way for enhanced chirality of the generated XUV

radiation. Apart from a slight deviation at low harmonic orders, the theoretical spectra accurately reproduce the experimentally observed trends in spectral asymmetry under conditions corresponding to different values of the intensity ratio $\gamma = I_{1200}/I_{400}$. A particularly compelling demonstration of the excellent agreement between theory and experiment is the predicted and observed reversal of dominance from harmonics of order $n = 4q - 1$ to those of order $n = 4q + 1$, occurring around H11–H13 for Ar and H17–H19 for Ne. The experimentally obtained spectral chirality of $\chi \approx 0.9$ ranks among the highest reported to date. Achieving such high chirality relies on maintaining an intensity ratio $3 < \gamma < 7$ between the components of the bicircular field, favoring the IR driver.

The predicted circular polarization of the dominant harmonics H17 and H21, with the helicity determined by the IR component and initially inferred solely from the spectral shape, was further confirmed by polarization characterization using XUV polarimetry to retrieve the full Stokes vector, as well as by numerical modeling. We identify additional generation channels that become active for elliptically polarized driving fields as the primary origin of the reduced ellipticity and lower degree of polarization observed for the $4q - 1$ harmonics H19 and H23.

Considering the dominant contribution of the $4q + 1$ harmonics, which exhibit high ellipticity and a high degree of polarization, and the negligible influence of the suppressed $4q - 1$ harmonics on the formation of the attosecond pulse train, we conclude that highly chiral attosecond pulses ($\varepsilon > 0.85$) are successfully generated in the time domain. The experimental results, supported by numerical simulations, provide a strong foundation for further ultimate proving helicity-dependent studies in chiral media, which we plan to pursue in the near future.

ACKNOWLEDGMENTS

N.Z. and U.B. acknowledge support from Deutsche Forschungsgemeinschaft the DFG QUTIF grant IV 152/6-1. D.B.M. acknowledge support from the Alexander von Humboldt Foundation.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

- [1] P. B. Corkum, Plasma perspective on strong field multiphoton ionization, *Phys. Rev. Lett.* **71**, 1994 (1993).
- [2] K. J. Schafer, B. Yang, L. F. DiMauro, and K. C. Kulander, Above threshold ionization beyond the high harmonic cutoff, *Phys. Rev. Lett.* **70**, 1599 (1993).
- [3] M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L'Huillier, and P. B. Corkum, Theory of high-harmonic generation by low-frequency laser fields, *Phys. Rev. A* **49**, 2117 (1994).

- [4] P. Salières, A. L'Huillier, and M. Lewenstein, Coherence control of high-order harmonics, *Phys. Rev. Lett.* **74**, 3776 (1995).
- [5] P. Balcou, P. Salières, A. L'Huillier, and M. Lewenstein, Generalized phase-matching conditions for high harmonics: The role of field-gradient forces, *Phys. Rev. A* **55**, 3204 (1997).
- [6] P. Salières, A. L'Huillier, P. Antoine, and M. Lewenstein, Study of the spatial and temporal coherence of high-order harmonics, *Adv. At. Mol. Phys.* **41**, 83 (1999).

- [7] P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Augé, P. Balcou, H. G. Muller, and P. Agostini, Observation of a train of attosecond pulses from high harmonic generation, *Science* **292**, 1689 (2001).
- [8] S. Li, L. Lu, S. Bhattacharyya, C. Pearce, K. Li, E. T. Nienhuis, G. Doumy, R. D. Schaller, S. Moeller, M.-F. Lin, *et al.*, Attosecond-pump attosecond-probe x-ray spectroscopy of liquid water, *Science* **383**, 1118 (2024).
- [9] Z. Guo, T. Driver, S. Beauvarlet, D. Cesar, J. Duris, P. L. Franz, O. Alexander, D. Bohler, C. Bostedt, V. Averbukh, *et al.*, Experimental demonstration of attosecond pump–probe spectroscopy with an x-ray free-electron laser, *Nat. Photonics* **18**, 691 (2024).
- [10] U. Bengs and N. Zhavoronkov, Elliptically polarized high-harmonic radiation for production of isolated attosecond pulses, *Sci. Rep.* **11**, 9570 (2021).
- [11] J. Long, X. Zhu, C. Zhai, W. Li, W. He, L. He, P. Lan, and P. Lu, Polarization control in high harmonic generation using molecular structures in nonaligned molecules, *Ultrafast Sci.* **5**, 0079 (2025).
- [12] D. B. Milošević and W. Becker, X-ray harmonic generation by orthogonally polarized two-color fields: Spectral shape and polarization, *Phys. Rev. A* **100**, 031401(R) (2019).
- [13] E. Bordo, O. Kfir, S. Zayko, O. Neufeld, A. Fleischer, C. Ropers, and O. Cohen, Interlocked attosecond pulse trains in slightly bi-elliptical high harmonic generation, *J. Phys. Photonics* **2**, 034005 (2020).
- [14] B. Ghomashi, S. Walker, and A. Becker, Enabling elliptically polarized high harmonic generation with short cross polarized laser pulses, *Sci. Rep.* **13**, 12843 (2023).
- [15] D. D. Hickstein, F. J. Dollar, P. Grychtol, J. L. Ellis, R. Knut, C. Hernández-García, D. Zusin, C. Gentry, J. M. Shaw, T. Fan, *et al.*, Non-collinear generation of angularly isolated circularly polarized high harmonics, *Nat. Photonics* **9**, 743 (2015).
- [16] H. Eichmann, A. Egbert, S. Nolte, C. Momma, B. Wellegehausen, W. Becker, S. Long, and J. K. McIver, Polarization-dependent high-order two-color mixing, *Phys. Rev. A* **51**, R3414 (1995).
- [17] D. B. Milošević, W. Becker, and R. Kopold, Generation of circularly polarized high-order harmonics by two-color coplanar field mixing, *Phys. Rev. A* **61**, 063403 (2000).
- [18] D. B. Milošević and W. Becker, Attosecond pulse trains with unusual nonlinear polarization, *Phys. Rev. A* **62**, 011403(R) (2000).
- [19] K. M. Dorney, J. L. Ellis, C. Hernández-García, D. D. Hickstein, C. A. Mancuso, N. Brooks, T. Fan, G. Fan, D. Zusin, C. Gentry, *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).
- [20] N. Zhavoronkov and M. Ivanov, Extended ellipticity control for attosecond pulses by high harmonic generation, *Opt. Lett.* **42**, 4720 (2017).
- [21] Á. Jiménez-Galán, N. Zhavoronkov, D. Ayuso, F. Morales, S. Patchkovskii, M. Schloz, E. Pisanty, O. Smirnova, and M. Ivanov, Control of attosecond light polarization in two-color bicircular fields, *Phys. Rev. A* **97**, 023409 (2018).
- [22] M. Zhu, X. Lai, L. Hua, and X. Liu, Quantum-orbit analysis of the generation of a high-ellipticity attosecond pulse with circular-elliptical bichromatic laser fields, *Phys. Rev. A* **108**, 043106 (2023).
- [23] D. B. Milošević, High-order harmonic generation by a bichromatic elliptically polarized field: Conservation of angular momentum, *J. Phys. B* **48**, 171001 (2015).
- [24] A. Fleischer, O. Kfir, T. Diskin, P. Sidorenko, and O. Cohen, Spin angular momentum and tunable polarization in high-harmonic generation, *Nat. Photonics* **8**, 543 (2014).
- [25] D. Habibović, W. Becker, and D. B. Milošević, Symmetries and selection rules of the spectra of photoelectrons and high-order harmonics generated by field-driven atoms and molecules, *Symmetry* **13**, 1566 (2021).
- [26] L. Barreau, K. Veyrinas, V. Gruson, S. J. Weber, T. Auguste, J.-F. Hergott, F. Lepetit, B. Carré, J.-C. Houver, D. Doweck, and P. Salières, Evidence of depolarization and ellipticity of high harmonics driven by ultrashort bichromatic circularly polarized fields, *Nat. Commun.* **9**, 4727 (2018).
- [27] D. B. Milošević and W. Becker, Improved strong-field approximation and quantum-orbit theory: Application to ionization by a bicircular laser field, *Phys. Rev. A* **93**, 063418 (2016).
- [28] D. S. Brambila, A. Husakou, M. Ivanov, and N. Zhavoronkov, On-target diagnosing of few-cycle pulses by high-order-harmonic generation, *Phys. Rev. A* **96**, 063825 (2017).
- [29] Á. Jiménez-Galán, N. Zhavoronkov, M. Schloz, F. Morales, and M. Ivanov, Time-resolved high harmonic spectroscopy of dynamical symmetry breaking in bi-circular laser fields: The role of Rydberg states, *Opt. Express* **25**, 22880 (2017).
- [30] This cutoff should not be confused with the classical high-energy cutoff, which is proportional to the ponderomotive energy [17], $U_p = U_{p1200} + U_{p400} = I_{1200}/(4\omega^2) + I_{400}/[4(3\omega)^2]$, so that the ω component contributes more strongly to this cutoff. Beyond this cutoff, which is not visible in our experimental results, the spectrum decreases exponentially.
- [31] T. Fan, P. Grychtol, R. Knut, C. Hernández-García, D. D. Hickstein, D. Zusin, C. Gentry, F. J. Dollar, C. A. Mancuso, C. W. Hogle, *et al.*, Bright circularly polarized soft X-ray high harmonics for X-ray magnetic circular dichroism, *Proc. Natl. Acad. Sci. USA* **112**, 14206 (2015).
- [32] A. Fleischer, P. Sidorenko, and O. Cohen, Generation of high-order harmonics with controllable elliptical polarization, *Opt. Lett.* **38**, 223 (2013).
- [33] D. B. Milošević, Control of the helicity of high-order harmonics generated by bicircular laser fields, *Phys. Rev. A* **98**, 033405 (2018).
- [34] D. B. Milošević, Circularly polarized high harmonics generated by a bicircular field from inert atomic gases in the p state: A tool for exploring chirality-sensitive processes, *Phys. Rev. A* **92**, 043827 (2015).
- [35] D. B. Milošević and W. Becker, Role of long quantum orbits in high-order harmonic generation, *Phys. Rev. A* **66**, 063417 (2002).
- [36] D. B. Milošević, Generation of elliptically polarized attosecond pulse trains, *Opt. Lett.* **40**, 2381 (2015).
- [37] D. B. Milošević, Quantum-orbit analysis of high-order harmonic generation by bicircular field, *J. Mod. Opt.* **66**, 47 (2019).
- [38] In general, the SFA theory is less reliable for harmonic-photon energies below the ionization potential. Nevertheless, the present results demonstrate that it still provides reasonable predictions even in this lower-energy regime.

- [39] P. Salières, B. Carré, L. L. Déroff, F. Grasbon, G. G. Paulus, H. Walther, R. Kopold, W. Becker, D. B. Milošević, A. Sanpera, and M. Lewenstein, Feynman's Path-Integral Approach for Intense-Laser-Atom Interactions, *Science* **292**, 902 (2001).
- [40] Historically, the multiindex (α, β, m) was introduced in 2002 [35], before the SFA theory of HHG was generalized to include the ground-state wave functions with arbitrary quantum numbers ℓ and m [34]. In order to avoid confusion of the magnetic quantum number m and the index m , we now denote this multiindex by (α, β, μ) , $\mu = 0, 1, 2, \dots$
- [41] R. M. A. Azzam and N. M. Bashara, *Ellipsometry and Polarized Light* (North-Holland Publishing Company, 1977).
- [42] R. M. A. Azzam, Stokes-vector and Mueller-matrix polarimetry [invited], *J. Opt. Soc. Am. A* **33**, 1396 (2016).
- [43] B. Schaefer, E. Collett, R. Smyth, D. Barrett, and B. Fraher, Measuring the Stokes polarization parameters, *Am. J. Phys.* **75**, 163 (2007).
- [44] F. Schäfers, H.-C. Mertins, A. Gaupp, W. Gudat, M. Mertin, I. Packe, F. Schmolla, S. D. Fonzo, G. Soullié, W. Jark, R. Walker, X. L. Cann, R. Nyholm, and M. Eriksson, Soft-x-ray polarimeter with multilayer optics: Complete analysis of the polarization state of light, *Appl. Opt.* **38**, 4074 (1999).
- [45] W. B. Westerveld, K. Becker, P. W. Zetner, J. J. Corr, and J. W. McConkey, Production and measurement of circular polarization in the VUV, *Appl. Opt.* **24**, 2256 (1985).
- [46] V. Wirthl, C. D. Panda, P. W. Hess, and G. Gabrielse, Simple self-calibrating polarimeter for measuring the Stokes parameters of light, *OSA Cont.* **4**, 2949 (2021).