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Schemes for generation of isolated attosecond pulses of pure circular polarization

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We propose and analyze two schemes capable of generating isolated attosecond pulses of pure circular polarization, based on results of numerical simulations. Both schemes utilize the generation of circularly polarized high-order harmonics by crossing two circularly polarized counter-rotating pulses in a noncollinear geometry. Our results show that in this set-up isolation of a single attosecond pulse can be achieved either by restricting the driver pulse duration to a few cycles or by temporally delaying the two crossed driver pulses. We further propose to compensate the temporal walkoff between the pulses across the focal spot and increasing the conversion efficiency by using angular spatial chirp to provide perfectly matched pulse fronts. The isolation of pure circularly polarized attosecond pulses, along with the opportunity to select their central energy and helicity in the non-colllinear technique, opens new perspectives to study ultrafast dynamics in chiral systems and magnetic materials.

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I. INTRODUCTION

Capture and control of ultrafast physical processes involving electron dynamics require the development of sources and measurement techniques operating on time scales of the order of attoseconds $(1 \text{ as} = 10^{-18} \text{ s})$ to femtoseconds (1 fs = 10^{-15} s). High-order harmonic generation (HHG) is an established table-top laboratory light source that produces coherent radiation at extreme ultraviolet (EUV) to soft X-ray wavelengths in bursts as short as a few tens of attoseconds [1-6]. For many years experimental research in HHG focused on the generation and application of linearly polarized harmonics and attosecond pulses. The physical mechanism at the microscopic level is well understood within the so-called three-step model [7, 8]: Following the release of an electron via tunnel ionization from the atom or molecule in an intense laser field, the electron is accelerated by the field and can be driven back to the parent ion. Upon recombination with the ion, the energy is released in form of high harmonic radiation.

This HHG technique with a one-color many-cycle linearly polarized driver pulse produces a train of several attosecond bursts [9]. Several schemes to isolate a single linearly polarized attosecond pulse have been developed and successfully implemented, such as reduction of driver pulse duration to a few cycles [10, 11], polarization gating schemes [12–14], combination of multicolor fields [15], spatio-temporal focusing [16], and ionization or time-gated phase-matching techniques [17–19].

Despite this substantial progress, the restriction in po-

larization of available attosecond pulses and pulse trains limits the prospects of ultrafast observations. Although techniques to produce isolated attosecond pulses with elliptical polarization have been proposed [20–23] to the best of our knowledge, so far there does not exist a practical proposal for a table-top laboratory set-up generating an isolated attosecond pulse of pure circular polarization. The availability of such pulses would be of importance for a new regime of applications across several areas of physics, ranging from generation of vortex patterns in photoelectron momentum distributions [24], via observation of ultrafast dynamics of photoelectron circular dichroism in chiral molecules [25] to ultrafast imaging of magnetic circular dichroism [26, 27]. In this paper we propose, based on results of numerical simulations, two schemes capable of overcoming this obstacle.

Although the physical principle has been proposed and used much earlier [28–30], controlling the polarization of the harmonics has become a center of interest more recently. While different techniques have been used to generate elliptically polarized harmonics [25, 31–34], the efficient generation of EUV [26, 35] and soft X-ray [27] beams of pure circular polarization was demonstrated by using bichromatic collinear beams with counter-rotating circular polarization, phase matched in a gas-filled waveguide. Another related technique, using non-collinear, counter-rotating beams [36], provides the additional capability of spatially separating circularly polarized high harmonics of different energy and helicity, however it is so far limited to the EUV region and lower flux.

The proposed schemes for the generation of isolated attosecond pulses of pure circular polarization utilize the non-collinear technique of generating circularly polarized high-order harmonics [36]. It consists of focusing two crossed counter-rotating pulses into a gas target (Fig.

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FIG. 1: Scheme of noncollinear circularly polarized high harmonic generation (NCP-HHG): Two counter-rotating circularly polarized femtosecond laser pulses are focused into a gas to produce both left and right circularly polarized harmonic beams. At the focal plane, the two circularly polarized beams sum to yield an electric field that exhibits linear polarization, which rotates as a function of the transverse position across the laser focus (inset). The locally linear nature of the polarization enables the application of schemes to isolate a single attosecond pulse, known from linearly polarized harmonic and attosecond pulse generation.

1). Though the technique does not depend on the specific wavelength of each pulse, here we consider circularly polarized pulses having the same wavelength and equal amplitude. A key element for the proposed isolation schemes is that the superposition of the electric fields of the two beams at the focal plane yields a linearly polarized electric field that rotates as a function of the transverse position (see inset in Fig. 1). Thus, locally at any point in the focus the same single-atom physics as for linearly polarized high harmonic generation applies. Consequently, strategies, previously established to isolate linearly polarized attosecond pulses, can be adopted to generate isolated circularly polarized HHG pulses in the non-collinear technique. Since furthermore each harmonic order emerges at a different angle and harmonic beams with right and left-circular polarization are separated in the far field, the capability of generating isolated attosecond pulses of pure circular polarization comes along with the opportunity of selecting the central energy and helicity of the light field.

The paper is organized as follows: In section II.A. we study the isolation process provided by restricting the pulse duration of the driver pulses to a single cycle. We feature the significance of the results in the noncollinear geometry by contrasting them to those obtained in the bichromatic collinear geometry (section II.B.) as well as by demonstrating the opportunities to select helicity and central energy of the isolated pulses (section II.C.). In section II.D, we show that the efficiency of the noncollinear high-order harmonic generation process and, hence, the flux of the isolated attosecond pulses can be increased by introducing angular chirp into the driving beams. In section III, we then analyze a second alternative method, in which a time delay between the pulses provides a polarization gating scheme that enables the generation of isolated circularly polarized pulses even when longer driving laser pulses are used. We end with a brief summary of the results.

II. ISOLATED CIRCULARLY POLARIZED ATTOSECOND PULSES DRIVEN BY SINGLE-CYCLE PULSES

In order to study the generation of high harmonics and attosecond pulses through the NCP-HHG scheme, we perform numerical simulations including propagation through the electromagnetic field propagator [37]. The single-atom dipole acceleration is computed using the SFA+, an extension of the strong field approximation [38] The gas in the interaction region is modeled as an argon gas at a pressure of 5 Torr. The signal at the detector is computed as the coherent addition of the HHG contributions of all the elementary sources, where the HHG light is assumed to propagate to the detector with a phase velocity equal to the speed of light c, and propagation effects of the fundamental field are taken into account.

A. Generation of isolated attosecond pulses of pure circular polarization in a noncollinear geometry

In Fig. 2 we present the attosecond pulses generated by two counter-rotating circularly polarized beams in a non-collinear one-color (left hand panels) geometry for long (upper row) and ultrashort (lower row) pulses. For the simulations the driving field at the focus is given by the superposition of two beams, E_+ and E_- :

$$\mathbf{E}_{\pm}(\mathbf{r},t) = E_0 A(t) \times U(x, y \cos \theta_{NC} \mp z \sin \theta_{NC}, \pm y \sin \theta_{NC} + z \cos \theta_{NC}) \times [\cos(2\pi c t/\lambda + \phi_0) \mathbf{e}_x \pm \sin(2\pi c t/\lambda + \phi_0) \mathbf{e}_y]$$
(1)

where the \pm sign reflects the geometrical and polarization differences between the two noncollinear beams. The wavelength of both beams is chosen to be $\lambda = 800$ nm, with a carrier-envelope phase of $\phi_0 = \pi/4$, and $\theta_{NC} = 32$ mrad is the crossing angle, which is chosen to match typical experimental conditions [36]. Each noncollinear driving beam is assumed to have a Gaussian spatial profile, U(x', y', z'), propagating in the z' direction, with a beam



FIG. 2: Comparison of circularly polarized attosecond pulses obtained through NCP-HHG and linearly polarized pulses driven by collinear two-color beams. Results are shown for relatively long driver (upper row) and ultrashort (lower row) driver pulses. Harmonic spectra (a,b) and circularly polarized attosecond pulses (c,d) for 800 nm counter-rotating noncollinear mixing with $\theta_{NC} = 32$ mrad and laser pulse durations $t_{\rm FWHM} = 5.8$ cycles (15.4 fs) and $t_{\rm FWHM} = 1.1$ cycles (2.9 fs), respectively. In panels (e,f) the linearly polarized attosecond pulses obtained using the collinear bi-chromatic (800 nm and 400 nm) scheme, for $t_{\rm FWHM,1} = t_{\rm FWHM,2} = 11.5$ fs, and $t_{\rm FWHM,1} = t_{\rm FWHM,2} = 2$ fs driver laser pulses, respectively.

waist of 80 μ m. The laser pulse envelope A(t) is modeled by a sin² function truncated at the first zeroes, with peak intensity of $E_0^2 = 8.75 \times 10^{13} \text{ W/cm}^2$ at the focus for each beam, and pulse durations of $t_{\rm FWHM} = 5.8$ cycles (15.4 fs, upper panels) and $t_{\rm FWHM} = 1.1$ cycles (2.9 fs, lower panels) full-width-at-half-maximum (FWHM) in intensity.

As shown before and observed in the experiments [36], for counter-rotating noncollinear mixing the resulting spectrum (Fig. 2(a)) consists of spatially separated harmonic beams with right and left circular polarization. For the longer driver pulse (a) well separated harmonics can be seen, while for the ultrashort driver pulse (b) a more continuous spectrum of harmonic radiation (on a linear scale) is observed. In Figs. 2(c) and (d) we present the attosecond waveforms which we obtained by Fourier transformation of the harmonic spectrum detected at 1.7 mrad. For the long driver pulse (c) we obtain a pulse train of two circularly polarized pulses per cycle, that in the case of a few-cycle driver pulse (d) is confined to an isolated pulse of attosecond radiation of circular polarization (with a couple of small side bursts). The pulse duration of the central burst is 360 as (at intensity FWHM), and the intensities of the side bursts are suppressed to about 2% of the central burst. Thus, as expected due to the superposition of the two beams to a local linearly polarized driving field with a rotating polarization direction as a function of the transverse position, attosecond pulses of circular polarization are generated upon each recombination event. Furthermore, as in the case of linearly polarized harmonics [10, 11], the reduction of the duration of the driver pulse leads to the isolation of a single burst.

B. Comparison of attosecond pulse generation in collinear and non-collinear geometry

In order to demonstrate the significance of this result obtained in the noncollinear geometry, we use the same strategy for bichromatic collinear beams with counterrotating circular polarization. As mentioned at the outset, the collinear technique leads to the efficient generation of circularly polarized harmonics beams as well [26, 27, 35]. However, as we will show now in that geometry the restriction to few-cycle driver pulses leads to the isolation of an attosecond pulse but of linear polarization.

Since our results indicate that in this geometry the isolation of a single burst requires the restriction of the driver pulse length to a regime in which S-matrix theories become unreliable due to nonadiabatic effects [40], we perform single-atom simulations by numerically solving the time-dependent Schrödinger equation in hydrogen, which has similar ionization potential as argon. In the simulations for the collinear bichromatic case the driving field is described as a superposition of two counterrotating circularly polarized laser pulses in the form of

$$\mathbf{E}_{C,i}(t) = A_i(t)E_{0,i}[\cos(2\pi ct/\lambda_i + \phi_0)\mathbf{e}_x \\ \pm \sin(2\pi ct/\lambda_i + \phi_0)\mathbf{e}_y]$$
(2)

where $\lambda_1 = 800$ nm, $\lambda_2 = 400$ nm and $E_{0,1}^2 = E_{0,2}^2 = 8.75 \times 10^{13}$ W/cm², $\phi_0 = \pi/2$. The pulse envelopes $A_i(t)$ are modeled, as in the case of noncollinear beams, via a sin² function and the envelope's pulse duration are $t_{\rm FWHM,1} = t_{\rm FWHM,2} = 11.5$ fs (upper panel) and $t_{\rm FWHM,1} = t_{\rm FWHM,2} = 2$ fs (lower panel).

The attosecond pulses generated in the collinear twocolor scheme show different polarization characteristics.



FIG. 3: Spatial selection of isolated circularly polarized attosecond pulses with different frequency. Shown are the HHG spectrum (first column), time-frequency analysis (second column) and attosecond pulse (third column) obtained at three different divergence angles: 1 mrad (a), 1.5 mrad (b), and 2 mrad (c), corresponding to the results presented in Fig. 2b.

As observed in previous works [23, 30], the resulting spectrum exhibits two families of alternative counter-rotating circularly polarized harmonics and, hence, the attosecond pulse train consists of three linearly polarized bursts per laser cycle, where the polarization rotates 120 degrees between each burst (Fig. 2(e)). Consequently, the reduction of the driver pulse duration leads to the isolation of a single pulse (Fig. 2(f)) of linear polarization instead of circular polarization as in the noncollinear scheme.

C. Selection of central energy of the isolated circularly polarized pulse in noncollinear geometry

Beyond the generation and isolation of circularly polarized attosecond pulses, the noncollinear scheme offers another feature, namely the spatial separation of attosecond pulses at different central energy. In the noncollinear HHG process, different harmonics are emitted with different angles, as shown in Figs. 2(a) and 2(b). As a consequence, it is straightforward to spatially select different parts of the harmonic spectrum.

This can be seen in Fig. 3, which shows the HHG spectrum (first column), time-frequency analysis (second column) and attosecond pulse (third column) obtained at three different divergence angles, 1 mrad (a), 1.5 mrad

(b), and 2 mrad (c), corresponding to the simulations presented in Fig. 2(b). Three different spectra centered at 37 eV, 32 eV and 26 eV are obtained. Note that at each angle still different spectral component overlap and, hence, isolated attosecond pulses with different central energy are generated. The overlap of the spectral components at a given detection angle, depends on the beam waist of the non-collinear beams, and on their crossing angle θ_{NC} . Smaller crossing angles and larger beam waists would result in broader spectral overlapping. On the other hand, the overlap can also be achieved by refocusing the harmonic light to the target. We note that such refocusing may alter the temporal structure of the attosecond pulse from that observed at any given angle (Fig. 3). In this case techniques for the compensation of the spatial chirp may be applicable, if necessary. Numerical studies of such scenarios are beyond the scope of the present analysis, which demonstrates the emission of isolated attosecond pulses with a rather broad spectrum at any given divergence angle in the noncollinear method itself only.

The time-frequency analysis in Fig. 3 (left column) exhibits a structure with positive slope, since the macroscopic addition of the single-atom results favors the socalled short quantum paths in the HHG process. Thus, the circularly polarized attosecond pulses are emitted with a positive chirp, as it is the case for HHG driven by a single-cycle linearly polarized pulse [39].

D. Compensation of transverse temporal walk-off with angular chirp

In the non-collinear geometry with conventional focusing, the pulse fronts (i.e., the intensity envelopes) of each beam are not parallel. Therefore, there is a temporal walk-off between each non-collinear laser pulse across the focal spot (see Fig. 4(a)). At the focal plane, this temporal walk-off is given by

$$\tau_{wo} = \frac{\sin \theta_{NC}}{c} x \tag{3}$$

where x is the transverse position and θ_{NC} is the crossing angle between the two beams. With temporal walkoff, the relative amplitudes of the two fields becomes a function of time. For two Gaussian pulses of counter-rotating polarization, equal amplitude and pulse duration $(1/e^2$ half-width τ), delayed by $t_d = \tau_{wo}$, we calculate the timevarying ellipticity of the resulting pulse:

$$\varepsilon(t) = \tanh\left(\frac{2t_d}{\tau^2}t\right)$$
 (4)

Thus, by superposing the two non-collinear laser pulses at the focal plane, the walk-off delay in the envelopes of each pulse restricts the spatial region over which polarization is linear. This effect will reduce the total HHG yield and the attosecond pulse intensity, since the HHG conversion efficiency drops quickly with increasing ellipticity [41].



FIG. 4: Compensation of transversal temporal walk-off using angularly chirped non-collinear beams. Shown are schematics of the wavefronts due the transverse walk-off (a) and after compensation using pulse-front-tilted beams, with opposite angular chirp rate (b). In (c) a comparison of the harmonic spectra at 1.5 mrad with (dashed red line) and without (light blue line) transverse walk-off effect, as well as after compensation with angular chirp (dark blue line) are shown. Laser parameters are given in the text.

In Fig. 4(c) we compare simulated HHG spectra, detected at an angle of 2 mrad, in which the walk-off effect is taken into account (light blue line) and, for the sake of comparison, neglected (dashed red line). The simulations parameters are as in Fig. 2(b), i.e., a laser pulse of $t_{FWHM} = 2.9$ fs with a crossing angle of $\theta_{NC} = 32$ mrad, but now with a Gaussian beam radius of 40 μ m. We have reduced the beam waist in order to compensate for the increase in computation time due to a higher resolution in the frequency domain needed for this kind of calculations. We can observe that in this case, the temporal walk-off reduces the HHG yield by a factor of 2 to 4 (c.f., dashed red and light blue lines). The suppression would further increase with increasing θ_{NC} and/or the beam waist. On the other hand, the effect is smaller for longer pulses.

In order to compensate for the temporal walk-off, we propose to imprint an opposite pulse front tilt in the noncollinear beams via an angular spatial chirp. To this end, we consider beams with transverse spatial chirp, where all the spectral components travel as beamlets in the same direction but with a transverse offset proportional to the frequency offset:

$$x_s(\omega) = \alpha(\omega - \omega_0) , \qquad (5)$$

where α is the transverse chirp rate. With a lens or curved mirror such a beam with transverse spatial chirp will produce a beam with pure angular chirp at the focal plane [42]. In that case, the beam angle θ_x changes with the frequency in terms of the angular chirp rate γ_0 as

$$\tan \theta_x(\omega - \omega_0) = \gamma_0(\omega - \omega_0), \qquad (6)$$

where $\gamma_0 = -\alpha/f$ and f is the lens focal length. This configuration, where the spectral components overlap with an angular sweep, is known as simultaneous space-time focusing [43]. Using the small angle approximation, $\tan \theta_x \approx \sin \theta_x$, the spectral phase of the pulse at the focal plane can be written as

$$\phi(x,\omega) = \omega \sin \theta_x(\omega) x/c .$$
(7)

By calculating the group delay via the first derivative of the phase, we obtain

$$\tau_g = \gamma_0 \frac{\omega_0}{c} x \tag{8}$$

that is, the group delay varies linearly across the focal spot, exhibiting a pulse front tilt of the beam [42]. The angular spatial chirp, that can be characterized by interferometric techniques [44], has been recently proposed as an element to improve the spectral and temporal resolution of HHG [45].

If the non-collinear beams are sculpted with opposing pulse front tilt so that the intensity envelopes match across the focal spot, perfect temporal overlap between the pulses can be achieved (see Fig. 4(b)). As a result, the electric field along the transverse direction will exhibit perfect linear polarization. Comparing Eqs. (2) and (8), the spatial chirp rate required for compensating the transverse temporal walk-off is

$$\gamma_0 = \frac{\sin \theta_{NC}}{\omega_0}.\tag{9}$$

Such an angular dispersion required for pulse front overlap can be achieved by using a diffraction grating as a beam splitter and imaging the grating to the interaction target [46, 47].

The dark blue line in Fig. 4(c) shows the HHG spectrum obtained when the transverse walk-off is included and compensated with opposite angular chirp in each of the non-collinear beams. In the corresponding simulations the spatial chirp rate, according to Eq. (9), is $\gamma_0 = 13.56$ as. We indeed observe that the compensation via the angular chirp leads to HHG yields similar to those obtained without the walk-off effect.

III. ISOLATED CIRCULARLY POLARIZED ATTOSECOND PULSES DRIVEN BY TIME-DELAYED LONG PULSES

In section II, we analyzed the generation of isolated circularly polarized attosecond pulses using near-singlecycle driving laser pulses. Those driver pulses are however difficult to obtain in the laboratory. In addition, effects such the transverse temporal walk-off, discussed in



FIG. 5: Time-gating mechanism for generating an isolated circularly polarized attosecond pulse with longer driving pulses. Ellipticity (a) and intensity (b) of the resulting laser pulse at the focus position as a function of the time delay t_d between the two non-collinear pulses. Attosecond pulses (left, linear scale, amplitude normalized to 1 in each panel) and HHG spectra (right, logarithmic scale) obtained in a NCP-HHG scheme for (c) $t_d = 0$, (d) $t_d = 0.5t_{FWHM}$, (e) $t_d = 0.75t_{FWHM}$, and (f) $t_d = t_{FWHM}$. Laser parameters are as in Fig. 2(b,d), except that the pulse duration is doubled to 5.8 fs.

section II.D., are more effective for short driving pulses. To this end, we have also analyzed schemes to obtain isolated circularly polarized attosecond pulses with longer driving pulses. These schemes can be considered as extension of the polarization gating technique, which has been successfully applied in linearly polarized HHG to obtain isolated linearly polarized attosecond pulses [12– 14]. For example, in Ref. [12] it is shown that two quarter waveplates can be used to produce a few-cycle pulse. In this case the polarization is linear at the center of the pulse only and elliptical at times away from the peak of the pulse. One way to apply this approach in the non-collinear method could be to create a pulse whose polarization is *circular* only at the pulse peak and use this in one or both beams.

A simpler approach, however, is to exploit the effect of a temporal delay t_d between the two noncollinear pulses, which we have discussed in the context of the walkoff delay in section II.D. Any delay results in a temporal evolution of the polarization of the resulting pulse at the focal plane (see Eq. (4)). By deliberately delaying the two pulses to each other, an effective polarization gate can be designed in which the polarization of the resulting pulse changes from circular to linear and back to circular. This is illustrated in Fig. 5, where we present the ellipticity (a) and intensity (b) of the resulting laser pulse at the focus position as a function of the time delay t_d between the two non-collinear pulses. Note that the temporal window over which linear polarization is obtained decreases as the time delay is increased. Since attosecond pulses are emitted more efficiently for linearly polarized drivers, the time-delay between the non-collinear beams gives the opportunity to control the number of attosecond pulses emitted within the train, and, therefore, to isolate a single circularly polarized attosecond pulse. However, the intensity at the center of the pulse reduces, as the time delay increases (see Fig. 5(b)), and therefore the cutoff energy of the resulting HHG yield as well as the efficiency of the yield are expected to be reduced.

In the other panels of Fig. 5 we present the attosecond pulses (linear scale) and HHG spectra (logarithmic scale) obtained using this scheme in the non-collinear geometry, where the pulse duration of the driving pulses is $t_{FWHM} = 2.2$ cycles (5.8 fs). In order to visualize the gating technique towards the isolation of the central cycle, the attosecond pulses yields have been normalized in each panel. Note that all laser parameters are the same as in Fig. 2(b,d), except the pulse duration, which is doubled. We did not include the transversal walk-off effect in these calculations, as it is expected to be smaller than for the ultrashort pulses studied in the last section and a near-perfect compensation can be achieved.

The results of the simulations indeed confirm, that as the time delay increases from $t_d = 0$ (c) to $t_d = t_{FWHM}$ (f), the side bursts in the train get suppressed relative to the main center cycle burst and, hence, the number of attosecond pulses within the train reduces towards the selection of an isolated circularly polarized attosecond pulse. While the intensity of the adjacent side pulses in Fig. 5(c) ($t_d = 0$) is 42% and 23% of that of the central pulse, in Fig. 5(f) ($t_d = t_{FWHM}$) it represents the 3% and 2% respectively. The pulse duration of the central burst in Fig. 5(f) is 300 as (at intensity FWHM). At the same time, we see that, as expected, the cutoff energy and the harmonic yield reduces, as compared to the case without any delay. Thus, the isolation of circularly polarized attosecond pulses is indeed possible, even with longer duration driving lasers, though there is a trade-off in conversion efficiency.

IV. SUMMARY

In summary, we have used the results of numerical simulations to propose and analyze different schemes of generating isolated circularly polarized laser pulses via high harmonic generation with non-collinear counter-rotating driver pulses of the same wavelength. It is shown that the reduction of the driver pulse duration reduces the number of circularly polarized pulses in the attosecond pulse train, similarly as previously observed in the case of linearly polarized pulses. Due to the emission of each harmonic at a different divergence angle in the noncollinear technique, the central energy of the isolated attosecond pulse and the handedness of the circular polarization can be selected in the far field. The loss of conversion efficiency due to the transversal walk-off in the case of fewcycle pulses can be compensated by matching the pulse front tilt in the non-collinear beams via an angular spatial chirp.

We have further shown that a time-delay between the two non-collinear pulses generates an effective polarization gate in the resulting pulse at the focal plane. This provides an alternative route to the generation of isolated circularly polarized attosecond pulses even with longer 7

duration driving pulses. This time-gating method for isolating attosecond pulses is anticipated to be more effective for longer wavelength driving pulses since in this case HHG is more sensitive to the ellipticity of the driver pulse.

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