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Published in:
Physical Review Letters

Document Version:
Publisher's PDF, also known as Version of record

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Download date:06. Dec. 2018
Generating Isolated Elliptically Polarized Attosecond Pulses Using Bichromatic Counterrotating Circularly Polarized Laser Fields

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(Received 29 April 2015; published 7 October 2015)

We theoretically demonstrate the possibility to generate both trains and isolated attosecond pulses with high ellipticity in a practical experimental setup. The scheme uses circularly polarized, counterrotating two-color driving pulses carried at the fundamental and its second harmonic. Using a model Ne atom, we numerically show that highly elliptic attosecond pulses are generated already at the single-atom level. Isolated pulses are produced by using few-cycle drivers with controlled time delay between them.

DOI: 10.1103/PhysRevLett.115.153001 PACS numbers: 32.80.Wr, 42.50.Tx, 42.65.Ky, 42.65.Re

High harmonic generation (HHG) in atoms and molecules is a highly nonlinear process that up-converts intense infrared laser field into the extreme ultraviolet (XUV) and soft x-ray radiation [1–4]. The emitted light can be used to track quantum dynamics underlying the nonlinear response [5–8], or as a tabletop source of bright, coherent, ultrashort pulses [9–12].

In the latter case, generation of circular or highly elliptic high harmonics and/or attosecond XUV pulses is very important. Such pulses would find numerous applications, e.g., in chiral-sensitive light-matter interactions such as chiral recognition via photoelectron circular dichroism [13–15], study of ultrafast chiral-specific dynamics in molecules [16,17], and x-ray magnetic circular dichroism spectroscopy [18–21], including time-resolved imaging of magnetic structures [18–22]. Tabletop sources of sub-100 fs, or even attosecond, chiral pulses would be a real breakthrough for laboratory-scale ultrafast studies. Not surprisingly, the search for schemes enabling the generation of short, coherent XUV pulses with tunable polarization is a very active area of research; see, e.g., Refs. [16,25–39].

Importantly, the control over polarization is desired not only for individual harmonics, where it has just been demonstrated [35,36], but also for individual attosecond pulses, both isolated and in a train, where a robust and practical scheme is still lacking. We show a way to solve this problem, proposing a practical scheme for the generation of highly elliptic attosecond pulses, both single and in a train.

An elegant solution to generating individual high harmonics with circular polarization has been found by Becker and co-workers [28,29,31,32]; see also [40] for strongly related ideas. It relies on combining a circularly polarized fundamental field with a counterrotating second harmonic. The resulting electric field peaks three times within one cycle of the fundamental, producing three ionization bursts. The electron promoted to the continuum near the peak of the instantaneous field can successfully revisit the parent ion within about a half-cycle, emitting an attosecond radiation burst [28,29].

This approach has now been very successfully used in Refs. [35,36], demonstrating generation of bright, phase-matched high harmonic radiation. Importantly, tuning the ellipticity of one of the fields allows one to tune the ellipticity of the generated harmonics from linear to circular [35]. While the theoretical interpretation of this control is an interesting question in its own right [33,35], the approach is very promising. However, until now the possibility of extending this scheme from controlling the polarization of individual harmonics to controlling the polarization of isolated attosecond pulses looked far from straightforward.

Indeed, the driving field dictates that the direction of electron return rotates by 120° three times per cycle. Consequently, recombination with an s state yields three linearly polarized attosecond bursts per cycle, with polarization rotating by 120° from burst to burst [29].

This can also be seen in the frequency domain. The harmonic lines are at energies \((n+1)\omega + 2n\omega = (3n+1)\omega\) and \(n\omega + (n+1)2\omega = (3n+2)\omega\). In a centrally symmetric medium, and for circularly polarized driving fields, the selection rules dictate that the \(\Omega = (3n+1)\omega\) line
The 2D model potential is taken from [43].

We show that the harmonics generated from orbitals with time-dependent Schrödinger equation (TDSE) for a 2D and the second harmonic. Pulses, by tuning the time delay between the fundamental isolated attosecond pulse, or a controllable train with 2 or 3 800 nm driver and its second harmonic, one can generate an right-circularly polarized harmonics can differ by an order of importance ways. Firstly, the height of the adjacent left- and rotating harmonic lines strongly depend on the orbital driving pulses become relatively short, e.g., 7

Converging was tested with respect to the absorbing potential, the time step, and the spatial grid. Note that HHG in bicircular fields is dominated by very short trajectories [28].

The initial wave functions were obtained using imaginary time propagation filtering out the ground state wave function to obtain p and p states. The p states are defined as p = p ± ip. The laser intensity was kept such as not to exceed 5% ionization and to avoid strong shifts and mixing of the degenerate atomic orbitals described in Ref. [43]. The spectra were obtained by performing the Fourier transform of the time-dependent dipole acceleration, evaluated at every 0.5 a.u.

The results are robust with the variation of the pulse length, the shape and length of its rising and falling edges, laser intensity, and wavelength: we performed calculations from λ = 600 nm up to λ = 1200 nm.

Figure 1(a) shows reference spectra obtained for the 1s initial state of the model potential with Ip of neon. It agrees well with previously published results [28,32,35,36]; the harmonics come in pairs (n + 1)ω + n2ω = (3n + 1)ω

The 2D model potential is taken from [43]:

\[
V(r) = -\frac{Z(r)}{\sqrt{r^2 + a}},
\]

where Z(r) = 1 + 9 exp(−r²) and a = 2.88172 to obtain the ionization potential of the Ne atom Ip = 0.793 a.u. for the 2p orbitals. The 1s state has an energy E1s = −2.952 a.u. and the 2s energy is E2s = −0.217 a.u.. For reference calculations we use 1s as the initial state but keep the same ionization potential taking Z(r) = 1 and a = 0.1195. The laser electric field is

\[
E(t) = E_{ir}(t)(\cos[\omega t] + \cos[2\omega t])\hat{x} + E_{ir}(t)(\sin[\omega t] − \sin[2\omega t])\hat{y},
\]

where f(t) is the trapezoidal envelope with 2 cycle rising and falling edges and 5 cycle plateau (in units of fundamental). The ω field rotates counterclockwise (+). The second harmonic rotates clockwise (−).

The TDSE is propagated on a 2D Cartesian grid using a Taylor-series propagator with expansion up to eighth order [44]. A complex absorbing potential,

\[
V_c(x) = \eta(x − x_0)^n,
\]

with η = 5 × 10^−4 and n = 3 is used to avoid nonphysical reflections from the boundary. Other simulation parameters are summarized in Table I.

Convergence was tested with respect to the absorbing potential, the time step, and the spatial grid. Note that HHG in bicircular fields is dominated by very short trajectories [28].

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Table I. Parameters of the calculations in atomic units unless stated otherwise.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laser frequency</td>
<td>0.05</td>
</tr>
<tr>
<td>Laser electric field</td>
<td>0.05</td>
</tr>
<tr>
<td>Grid step size</td>
<td>0.2</td>
</tr>
<tr>
<td>Time step size</td>
<td>0.005</td>
</tr>
<tr>
<td>Propagation time</td>
<td>1250</td>
</tr>
<tr>
<td>Maximal grid extent</td>
<td>±60</td>
</tr>
<tr>
<td>Absorbing boundary</td>
<td>±36</td>
</tr>
</tbody>
</table>
and $n\omega + (n + 1)2\omega = (3n + 2)\omega$ of similar heights. The left harmonic in the pair has the same polarization as the fundamental field; the right harmonic follows the $2\omega$ driver. The harmonics $3n\omega$ are parity forbidden.

Figures 1(b) and 1(c) show spectra for the $p_+$ and $p_-$ initial states. For the $p_+$ initial state, the harmonics that have the same polarization as the driving IR field are preferred. For the $p_-$ initial state, the harmonics with the same polarization as the $2\omega$ driver are stronger. There are additional spectral variations in the plateau region, different for $p_+$ and $p_-$ orbitals. There is also a qualitative difference between the below-threshold ($<I_p$) and above-threshold ($>I_p$) harmonics, showing that the evolution of the photoelectron in the continuum is critical for the observed propensity in the harmonic strengths.

Figure 1(d) shows the spectra obtained from adding the contributions from the $p_+$ and $p_-$ orbitals coherently, as required. In the plateau region, harmonics with the same polarization as the driving IR field dominate over those with opposite polarization.

The subcycle dynamics of the emission process was analyzed using the Gabor transform (GT) [45] of the time-dependent acceleration dipoles $a(t)$:

$$\text{GT}[\Omega, t_0] = \frac{1}{2\pi} \int dt a(t)e^{-i\Omega t} e^{-i(t-t_0)^2/2T^2},$$

(5)

where we have chosen $T = 1/3\omega$. The reference spectrograms for the $1s$ initial state in Figs. 2(a) and 2(b) show the time-dependent intensity [2(a)] and ellipticity [2(b)] for time-resolved spectra, in the regions where spectral amplitudes are significant. As expected, there are three radiation bursts per $\omega$ cycle with linear polarization, as predicted in Refs. [28,29,32].

Figures 2(c) and 2(d) show the same spectrogram for the $2p$ state, i.e., the coherent superposition of the radiation from $p_+$ and $p_-$ states. Although the signal strength in the spectrogram is similar to the $s$ orbital, the ellipticity of the emitted radiation is very different. Three distinct regions can be identified: (i) below threshold region, where the ellipticity is mostly negative, (ii) the middle region, where the ellipticity is high and positive, and (iii) near cutoff region where the emitted radiation is mostly linear. The energy region (ii) of the spectrogram coincides with the spectral window in Fig. 1(d) where the difference between clockwise and counterclockwise harmonics is the greatest.

![FIG. 1 (color online). Spectra for (a) 1s, (b) 2p+, (c) 2p− initial states, and (d) equal mixture of 2p+ and 2p− states. Colors mark harmonics corotating (red) and counterrotating (blue) with the $\omega$ field.](image1)

![FIG. 2 (color online). Time-resolved XUV emission intensity and ellipticity from (a),(b) 1s and (c),(d) 2p orbitals. Color in (b) and (d) indicates the ellipticity of the spectral components in the regions where the amplitude of the spectrogram is significant. The horizontal dashed lines mark the $I_p$.](image2)
Application of bicircular fields is naturally extended from
the generation of an attosecond pulse train to the generation
of an isolated attosecond pulse, using short driving pulses
and changing the time delay between them. Indeed, the
harmonic emission driven by circular fields is only possible
when the two counterrotating circular pulses overlap. Given
the high nonlinearity of the overall process (including
ionization), it will be limited to the temporal window where
the two driving pulses overlap with nearly equal and high
intensity. This idea is tested in Fig. 3, which shows time-
resolved spectrograms and ellipticity of the emitted light for
$\lambda = 800$ and 400 nm counterrotating drivers with full width
at half maximum duration of 4 fs and $\sin^2$ envelope with $I = 1.7 \times 10^{14}$ W/cm$^2$ peak intensity, for two time delays.
In the case of perfect overlap, three attosecond pulses are
generated. Delaying the low-frequency pulse by 2.6 fs (full
period of 800 nm field) yields two attosecond pulses with
strong ellipticity, that are well separated in energy. An
isolated pulse would be obtained by filtering the lower or
higher energy pulse. With shorter driving pulses, a single
isolated attosecond pulse will be generated.

What is the physical origin of the HHG sensitivity to
the angular momentum of the initial state? The energy
and angular momenta that the electron accumulates from
the laser field while propagating in the continuum are
transferred to the harmonic photon upon recombination.
The matrix elements associated with recombination are the
complex conjugate of the photoionization matrix elements. In
2D one photon ionization with the field corotating with the
initial state is much more likely than with counterrotating
field. This is a direct analogue of Fano-Bethe propensity rules
[46] and is also the case for Rydberg states corotating and
counterrotating with the field [47,48].

Consider the harmonic spectra from the $p_+$ orbital. The right-circularly polarized harmonics result from the
$(n+1)\omega + n2\omega$ pathway. The recombination step is
conjugated to photoionization from the $p_+$ state with a
corotating field, favored by the propensity rules. The
left-circularly polarized harmonics result from the $n\omega +
(n+1)2\omega$ pathway. The recombination step is conjugated
to photoionization from the $p_+$ state with a counterrotating
field, disfavored by the propensity rules. This explains the
relative heights of the harmonic pairs for the $p_+$ initial
state. The same analysis explains why harmonics corotating
with the $2\omega$ field are preferred for the $p_-$ initial
state.

But why is $p_+$ dominant over $p_-$? The answer lies in the
stronger effect of the lower-frequency (counterclockwise)
field on the continuum electron, which leads to higher
population of the continuum states with positive angular
momentum than the population with the negative angular
momentum. The more probable recombination from such
states is to the $p_+$ state, by emitting light with counterclock-
wise polarization.

The carrier-envelope phase stabilization controls the
orientation of the polarization ellipse of the attosecond pulse,
but not the pulse. Indeed, as long as the relative phase
between the two pulses, $\omega$ and $2\omega$, is locked, changing
the carrier-envelope phase will rotate the trefoil pattern of
the driving field and thus the polarization ellipse of the
attosecond pulse but will not alter its high ellipticity.
This property, in combination with the possibility of using
relatively routine durations of the two driving pulses, makes
the scheme extremely attractive for practical implementation.

Finally, we comment on the difference between the 2D
model presented here and a 3D system. Since the harmonic
dipole for the $m = 0$ orbital is negligible, the only expec-
ted difference is the additional spreading of the continuum elect-
ron wave packet in the direction perpendicular of the
place of polarization of the driving field. Such spreading
is identical for both $p_+$ and $p_-$ orbitals and thus will not
change the relative intensity of the corresponding harmon-
ics. Consequently, we expect that our results stand in the
full 3D case. In this context, we would also like to bring
the reader’s attention to the recent 3D strong field approxi-
mation calculations of HHG for a Ne atom in a two-color
bicircular field [49] where similar results of the intensity
of left- and right-circularly polarized harmonics from
$m = \pm 1$ orbitals have also been found.

We thank Emilio Pisanty, Felipe Morales, Wilhelm Becker,
and Dejan Milošević for valuable discussions. Financial
support from the FP7 Marie Curie ITN CORINF, the
EPSRC Programme Grant No. EP/I032517/1, and partially
from the U.S. Air Force Office of Scientific Research under
program No. FA9550-12-1-0482 is acknowledged.

The computer codes and data used to produce this Letter
can be downloaded from Ref. [50].