Illuminating Molecular Symmetries with Bicircular High-Order-Harmonic Generation

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We present a complete theory of bicircular high-order-harmonic emission from \( N \)-fold rotationally symmetric molecules. Using a rotating frame of reference we predict the complete structure of the high-order-harmonic spectra for arbitrary driving frequency ratios and show how molecular symmetries can be directly identified from the harmonic signal. Our findings reveal that a characteristic fingerprint of rotational molecular symmetries can be universally observed in the ultrafast response of molecules to strong bicircular fields.

High-order-harmonic generation (HHG) represents one of the primary gateways towards obtaining novel tabletop light sources with unique properties for a wide range of applications [1]. At the same time it holds the promise to revolutionize our understanding of fundamental dynamical processes in atoms and molecules, demonstrated for example by the ultrafast tracing of charge migration in iodoacetylene [2], as well as in condensed-matter systems, exemplified by the advent of extreme ultraviolet spectroscopy in solids [3]. While the generation of bright linearly polarized light through HHG is well-established [3], efforts to expand the toolbox of ultrafast light probes towards circular and elliptical polarization have subsequently attracted great interest, motivated by the vast potential for applications in, e.g., the study of circular dichroism in chiral molecules [5] or the direct measurement of quantum phases [6, 7]. The most promising approach, namely HHG of circularly polarized light by bicircular driving, has recently garnered much attention due to several groundbreaking experiments demonstrating tunable polarization through helicity-selective phase matching [8–11], the generation of isolated attosecond pulses [11], the extension into the X-ray regime [12] and even detailed three-dimensional tomography of the emitted high-order-harmonic fields [13]. Even though the generation of circularly polarized pulses via HHG was theoretically examined already in the 1990s [14–18] these experimental studies reinvigorated interest also from the theoretical side particularly regarding the question of selection rules [19, 20] and the role of molecular and orbital symmetries [21, 22]. Most notably, a recent article provided a detailed analysis on the correlation between symmetries and high-order-harmonic spectra in both atoms and molecules [23]. The primary focus of most studies has been in the analysis of the simplest bicircular HHG scheme which involves a circularly polarized driver with a fundamental frequency \( \omega \) and another driver with opposite circular polarization at \( 2\omega \). For atomic targets one observes in this setup harmonics of opposite circularly polarization at frequencies \( (3n + 1)\omega \) and \( (3n + 2)\omega \) \( (n \in \mathbb{N}) \) whereas no signal is observed for frequencies \( 3n\omega \). For molecular targets this pattern generally becomes more elaborate [22, 23].

In a previous work [24] we argued that bicircular HHG can be understood by using a rotating frame of reference. In the case of a spherically symmetric target the neighboring high-order-harmonic peaks in the laboratory frame can be understood to originate from a linearly polarized harmonic in the rotating frame. This explains, e.g., the similar emission strength of those two harmonics from \( s \) states in atomic targets, a fact also reported in [23]. While the orbital symmetry hence influences the relative strength, the molecular symmetry can completely lead to the appearance and disappearance of certain peaks in the spectrum. Although the connection between dynamical symmetries and HHG selection rules has been known for a long time [24–26], the imprint of the molecular symmetry for bicircular driving has only recently been discussed [22, 23]. Still, up to this point the focus was mostly on specific driving-field configurations under particular rotational symmetries. Notably, only setups where the driving field consist of frequencies with an integer multiple have been considered. Here, we present a model using the rotating-frame picture that makes this restriction unnecessary.

We begin by briefly reviewing the rotating-frame transformation for a field-free Hamiltonian \( H_0 \) under the influence of the electric field of two counter-rotating circularly polarized pulses with envelope \( F_0(t) \) and frequencies \( \omega_1, \omega_2 \) polarized in the \( xy \)-plane,

\[
H(t) = H_0 + F_0(t) \left[ x \cos(\omega t) + y \sin(\omega t) + x \cos(\omega_1 t) - y \sin(\omega_1 t) \right].
\]

Although we employ a single-active-electron picture it is straightforward to show that the following discussion holds even when multiple electrons are considered, see the appendix for details.

The unitary transformation \( U(t) = e^{-i\alpha L_z} \), with \( \alpha = (\omega_1 - \omega)/2 \) and \( L_z \) the operator of angular momentum corresponding to rotation around the \( z \) axis, leads to the Hamiltonian in the rotating frame

\[
H'(t) = H_0'(t) + \alpha L_z + 2F_0(t)x \cos(\tilde{\omega} t),
\]

where

\[
\tilde{\omega} = \frac{1}{2} \left( \omega_1 + \omega_2 \right),
\]

and

\[
F_0(t) = \frac{1}{2} \left( F_0(\omega t) + F_0(\omega_1 t) \right).
\]
where $\dot{\omega} = (\omega + \omega')/2$ and $H^0_0 (t) = U (t) H_0 U^\dagger (t)$ \cite{24}. Equation \ref{2} demonstrates that in a rotating frame the dynamics of two counter-rotating circularly polarized driving fields can be interpreted as that of a single linearly polarized driver with double the field strength at the mean frequency with an additional angular momentum term, which we call the Coriolis term, proportional to half the difference frequency. In the rotating frame the nuclei are rotating with angular frequency $\alpha$ in the $xy$-plane, indicated by the time dependence of $H^0_0(t)$.

The right-circularly polarized (RCP), respectively left-circularly polarized (LCP), signal in the laboratory frame $S^\text{lab} (\Omega)$ is obtained via the corresponding signal in the rotating frame shifted in frequency by $\alpha$ to the left, respectively to the right, i.e., \cite{24}

$$S^\text{lab} (\Omega + \alpha) = S^\text{rot} \left( \Omega \right), \quad S^\text{lab} (\Omega - \alpha) = S^\text{rot} \left( \Omega + \alpha \right). \tag{3}$$

These formulas are valid even in the absence of axial symmetry. In a non-axially symmetric setting, however, the linearly polarized driver in the rotating frame will now irradiate a rotating target. As such the simple selection rule leading to only odd multiplicles of the driving frequency in the rotating frame ceases to be valid.

Since the bicircular driving field is polarized in the $xy$-plane the HHG process is well-described in two dimensions. Moreover, we can simplify our discussion even further by focusing on the projection of the molecular potential in $x$-direction in the rotating frame, i.e., $V (x, t) \equiv V (x, y = 0, t)$. This is motivated by the fact that the driving field in the rotating frame is linearly polarized in the $x$-direction and the ground-state wave function is centered at the rotational center, i.e., $(y) = 0$. Thus, ionization events, which are the first step in HHG according to the three-step model \cite{28,30}, are centered around $y = 0$. Moreover, we showed in Ref. \cite{24} that the deflection from the Coriolis term is generally negligible even for moderately high values of $\alpha$ and only leads to a depression of the high-order-harmonic plateau but neither alters the symmetry nor the selection rules.

At the center of our model lies the observation that the molecular potential in the rotating frame inherits a dynamical symmetry if a static rotational symmetry in the laboratory frame is present,

$$V(x, t) = V \left( x, t + \frac{2\pi}{N\alpha} \right). \tag{4}$$

We choose $t = 0$ such that $V(x, t) = V(x, t) \forall t$ for the projected potential in $x$-direction. This allows to express the potential in a Fourier series as follows,

$$V(x, t) = V_0(x) + \sum_k V_k(x) \cos (N k t) \tag{5}$$

At this point it becomes important to distinguish between even and odd $N$. For even $N$ one observes that $V(x, t) = V(-x, t) \forall t$ for the projected potential in $x$-direction. This allows to conclude that in Eq. \ref{5} for even $N$ all $V_k(x)$ are even, too. For odd $N$ we instead have the relation $V(x, t) = V(-x, t + \frac{2\pi}{N\alpha})$ since after half a period the potential inverts in the $x$-direction, cf. Fig. 1 and inserting this relation in Eq. \ref{5} leads to

$$V_0(x) + \sum_k V_k(x) \cos (N k t)$$

$$= V_0(-x) + \sum_k (-1)^k V_k(-x) \cos (N k t).$$

\begin{table}[h]
\centering
\begin{tabular}{|c|c|c|c|c|}
\hline
$N$ odd & $m\omega$, $m$ odd & $\pm N\omega$ & $\pm 2N\omega$ & $\pm 2N\omega$ \text{+} \\
\hline
$N$ even & $m\omega$, $m$ odd & $\pm N\omega$ & $\pm 2N\omega$ & $\pm 2N\omega$ \text{+} \\
\hline
\end{tabular}
\caption{Possible combinations of actual and virtual driving (as defined in the main text) with corresponding parity indicated by [+] (even) and [-] (odd). The total parity is the product of the constituents’ parities. Signals with even total parity are forbidden (indicated by $\times$). Signals with high order $k$ in the virtual driving, cf. Eq. \ref{6} are generally expected to be less pronounced.}
\end{table}
TABLE II. Predicted leading-order signals in general and for the two particular setups analyzed in Ref. [22] in the laboratory frame. In the latter case we report the lines for easier comparison in terms of the frequency $\omega$ of the RCP driver, cf. Eq. (1). The two branches of the side lines, indicated by I and II in the table header, correspond to absorption, respectively emission, from the virtual driver, cf. the $\pm$ sign in Table I. Note the opposite convention for RCP and LCP compared to Ref. [22].

| $N$ | \(m\) even | \(m\) odd | \(m\) even | \(m\) odd |
|-----|------------|-----------|------------|-----------|
| \(2\) | \(\omega\) | \(\omega\) | \(\omega\) | \(\omega\) |
| \(3\) | \(\omega\) | \(\omega\) | \(\omega\) | \(\omega\) |
| \(4\) | \(\omega\) | \(\omega\) | \(\omega\) | \(\omega\) |

Evidently, \(V_k(x)\) is even for even \(k\) (including \(k = 0\)) whereas it is odd for odd \(k\). The Hamiltonian in the rotating frame thus reads

\[
H(t) = T + V_0(x) + \alpha L_z + 2F_0(t)x \cos(\tilde{\omega}t) \\
+ \sum_k V_k(x) \cos(Nkat)
\]  

(6)

with \(T\) denoting the kinetic energy operator and \(V_0(x)\) representing the even time-averaged part of the potential. The form of Eq. (6) allows to interpret the time-dependence of the molecular potential in the rotating frame as additional driving fields with frequencies \(N\alpha\) that couple spatially via the coefficient \(V_k(x)\). We call these fields “virtual” driving fields since they appear due to the transformation to the rotating frame and the resulting rotation of the nuclei therein. This is in contrast to the “actual” driving field which is a consequence of the bicircular driving in the laboratory frame. The virtual driving field has perturbative character since its strength is related to the strength of the Coulomb potential. Specifically, when the electron gathers its energy from the virtual driving field configurations \(\omega' = 2\omega\) and \(\omega' = 3\omega\). The lower part of Table I summarizes the expected main and side lines in these two settings according to our model, expressed in terms of multiples of the fundamental driving frequency \(\omega\). The polarization of particular peaks in the HHG spectrum is determined by the superposition of the contribution from the main and side lines. For \(\omega' = 2\omega\) all main and side lines with a given circular polarization coincide leading to alternating left- and right-circular polarization (RCP at \((3n + 1)\omega\) and LCP at \((3n + 2)\omega\), \(n \in \mathbb{N}\)). Conversely, for \(\omega' = 3\omega\) there exist secondary side line contributions with opposite polarization compared to the main lines at \((4n + 1)\omega\) and \((4n + 3)\omega\). Furthermore, there is both a left- and a right-circular contribution on the level of a first side line at \(4\omega\) and \((4n + 2)\omega\). In the former case we expect the total signal to be predominantly circularly polarized since the secondary side lines are expected to be much weaker than the main line. For the latter case the superposition between opposite circular polarization occurs for side lines of the same order, hence we expect...
FIG. 2. HHG signal obtained by numerical simulations for $N = 3$ (top) and $N = 4$ (bottom) (left for RCP signal and right for LCP signal). The circles indicate the expected position of the main line, the squares and triangles indicate the primary, respectively, secondary, side line.

that the superposition will be close to linearly polarized. These predictions are in perfect accordance with the theoretical analysis and numerics shown in Ref. [22]. We note, however, that in some settings superpositions between, e.g., main and primary side lines may occur and a precise prediction on the resulting polarization of the total signal would require a more in-depth analysis.

To illustrate our model’s high degree of predictability for general bicircular driving schemes we performed numerical simulations of the two-dimensional time-dependent Schrödinger equation using a single-active electron approximation on a set of model molecules which obey a discrete $N$-fold rotational symmetry in the $xy$-plane. They are described by a potential

$$V = \sum_{p=0}^{N-1} \frac{-Q/N}{\sqrt{\left[x - R \cos \left(\frac{2\pi p}{N}\right)\right]^2 + \left[y - R \sin \left(\frac{2\pi p}{N}\right)\right]^2 + a}},$$

which represents a set of $N$ atomic cores at a distance $R$ from the origin evenly distributed at polar angles $2\pi N$. We employ a smoothening parameter $a$ for all cores and smear out a total charge $Q$ homogeneously among them. Our calculations used the following parameter values (atomic units used throughout): $Q = 2, R = 4.01, a = 0.251$, the driving laser field is given by a trapezoidally shaped bicircular driver with $T_{\text{ramp}} = 250$ and $T_{\text{plateau}} = 1500$ as well as $F_0 = 0.04$ and $\tilde{\omega} = 0.0876$.

Figure 2 shows HHG spectra in the laboratory frame of RCP and LCP emission for the three-fold, respectively four-fold, cases. Evidently, the fingerprint of the corresponding molecular symmetries is well-pronounced and can be observed for all values of $\alpha$. The strong difference between odd and even $N$ is clearly revealed with the primary side lines originating at $\alpha = 0$ for even multiples of $\tilde{\omega}$ for odd $N$, cf. the top panels of Fig. 2 whereas for even
generalization to many-electron case

We show that all predictions from the main text remain valid when moving to the many-electron case. Our new starting point is the many-electron Hamiltonian

\[
\hat{H} (t) = \hat{H}_0 + F_0 (t) \left( X \cos (\omega_1 t) + Y \sin (\omega_1 t) + X \cos (\omega_2 t) - Y \sin (\omega_2 t) \right),
\]

where \(\hat{H}_0\) is the field-free Hamiltonian and \(X = \sum x_i\) and \(Y = \sum y_i\) are the sum over the Cartesian coordinates \(x_i, y_i\) of the \(i\)-th electron. The rotating-frame transformation is now performed with respect to all electrons, i.e.,

\[
\hat{U} (t) = e^{-i \alpha t L_z},
\]

with \(L_z = \sum_i L_z^{(i)} = \sum_i \left[ x_i p_y^{(i)} - y_i p_x^{(i)} \right]\) and \(p_x^{(i)}, p_y^{(i)}\) the Cartesian momenta of the \(i\)-th electron. Note that the operator \(L_z\) without superscript corresponds to the total angular momentum of all electrons whereas the angular momenta of the individual electrons are indicated by \(L_z^{(i)}\). Since all operators corresponding to different electrons commute we can also write this as

\[
\hat{U} (t) = \prod_i e^{-i \alpha t L_z^{(i)}},
\]

where the ordering in the product is arbitrary.

The Hamiltonian \(\hat{H}_0\) can be split now into one-particle (kinetic energy and electron-nuclei interactions) and two-particle (electron-electron interactions) contributions. The one-particle contributions transform exactly as discussed in the main text since only the factor \(e^{-i \alpha t L_z}\) corresponding to the specific electron at hand plays a role - this argument also extends to the operators \(X\) and \(Y\) in the bicircular driving. Furthermore the two-particle contributions are invariant since the rotation induced by the unitary transformation leaves all distances and angles between the electrons invariant. As a consequence we arrive at the rotated-frame Hamiltonian for the many-electron case,

\[
\hat{H}' (t) = \hat{H}_0' (t) + \alpha L_z + 2 F_0 (t) X \cos (\tilde{\omega} t),
\]

where \(\hat{H}_0' (t) = \hat{H}_0 (\{x_i (t), y_i (t), z_i (t)\})\), \(x_i (t) = x_i \cos (\alpha t) + y_i \sin (\alpha t)\), and \(y_i (t) = y_i \cos (\alpha t) - x_i \sin (\alpha t)\). This is completely analogous to the single-electron case.

Most notably, we can still define the projected potential in the rotating frame as the sum of the contributions by the individual electrons,

\[
\hat{V} (\{x_i\}, t) = \sum_i V_i (x_i, t),
\]

which follows the same symmetries as the individual contributions,

\[
V_i (x_i, t) = V_i \left( x_i, t + \frac{2 \pi}{N \alpha} \right)
\]

\[
\Rightarrow \hat{V} (\{x_i\}, t) = \hat{V} \left( \{x_i\}, t + \frac{2 \pi}{N \alpha} \right).
\]

From this point we can follow the same steps presented in the main text for a single electron and arrive at the

Appendix: Generalization to Many-Electron Case

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\[
\hat{H} (t) = \hat{H}_0 + F_0 (t) (X \cos (\omega_1 t) + Y \sin (\omega_1 t) + X \cos (\omega_2 t) - Y \sin (\omega_2 t)),
\]
Fourier-expanded Hamiltonian in the rotating frame for the many-electron case

\[ \hat{H}(t) = \hat{T} + V_{ee} + \sum_i V^{(i)}_0(x) + \alpha L_z \]

\[ + 2F_0(t)X(t) \cos(\omega t) + \sum_{i,k} V^{(i)}_k(x_i) \cos(Nk\omega t), \]

where the \( V^{(i)}_k \) obey the same symmetries as in the main text for all \( i \) and \( V_{ee} \) contains the electron-electron interaction which is of even parity since the potential energy of all electrons is invariant under any spatial transformation. In particular, it does not depend on the molecular symmetry at all. At this point it becomes clear that all symmetry arguments with respect to the high-harmonic spectra as well as the appearance of main and side lines remain completely intact even in a many-electron description.

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