

High-order harmonic generation with orbital angular momentum beams: Beyond-dipole corrections

Esra Ilke Albar^{1,*}, Valeriia P. Kosheleva¹, Heiko Appel¹, Angel Rubio^{1,2} and Franco P. Bonafé^{1,†}

¹*Max Planck Institute for the Structure and Dynamics of Matter, Center for Free Electron Laser Science, Luruper Chaussee 149, 22761 Hamburg, Germany*

²*Center for Computational Quantum Physics (CCQ), The Flatiron Institute, 162 Fifth Avenue, New York, New York 10010, USA*



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We study high harmonic generation with vortex beams beyond the dipole approximation. To do so, we employ the full minimal coupling approach to account for multipolar coupling without truncation and describe the full spatio-temporal properties of the electromagnetic field. This allows us to investigate the beyond-dipole deviations in electron trajectories and the harmonic spectra, where the influence of the orbital angular momentum contains both magnetic and quadrupolar effects. We show that when the system is driven by plane-wave light, nonlinear dipole oscillations occur along both the polarization and propagation directions, consistent with previous findings. In contrast, vortex beams also generate nonlinear dipole oscillations in the direction perpendicular to both the polarization and propagation of the incoming field. We identify the effects of the resulting symmetry breaking due to beyond-dipole corrections, which are especially visible in even harmonics.

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I. INTRODUCTION

The fact that light can carry orbital angular momentum (OAM) was first theoretically predicted by Allen *et al.* [1] and experimentally realized by He *et al.* [2]. Nowadays, OAM beams (vortex or twisted-light beams) are routinely produced using spiral phase plates [3], forked holograms [4], q-plates [5], metallic nanostructures [6–8], and many others (for more details, see reviews [9,10]). Due to OAM, twisted light has found applications in optical manipulation [11,12], quantum information [13,14], imaging and microscopy [15,16], super-resolution optical sensing [17], and many others. Numerous significant theoretical and experimental investigations have been conducted to gain a deeper understanding of the interaction between twisted light and matter, encompassing processes such as excitation [18–27], ionization [28–35], scattering [36–39], and various others. Recently, the study of high harmonic generation (HHG) with twisted light has gained momentum [40–51], as it offers a tabletop setup for generating OAM-carrying light beams.

The theoretical frameworks that study HHG have evolved through time to refine the description of light-matter interactions. As a first step, the HHG is understood as a three-step model [52]: (I) the electron is ionized from a parent atom, then (II) it moves in the laser field, and, finally, (III) it recombines with a parent atom and emits high harmonics. For optical

lasers with intensities around 5.0 V/nm, a typical electron motion range is much smaller than the wavelength of a driving laser. Therefore, the spatial profile of the laser field in step (II) is neglected, i.e., the electric dipole approximation is applied and the electron is assumed to be driven solely by an electric field. Within this approach, the dominant direction of the HHG emission process is determined by the incident field's amplitude and polarization direction. Nevertheless, various corrections to this picture ensued [53]. For instance, subsequent works have shown that the magnetic field can significantly influence the trajectories of high-speed electrons with the Lorentz force, resulting in a figure-eight electron motion which requires corrections to the electric dipole approximation [54–60]. This, in turn, leads to deviations of the trajectories in the propagation direction of the beam. However, the effects of the spatial structuring of light on the electron motion in the plane orthogonal to the propagation, as it occurs in twisted-light beams, are yet to be unraveled. In particular, twisted-light beams have a space-dependent intensity profile, which causes the usual dipole approximation to fail to describe the interaction between matter and these types of beams. This shortcoming of the electric dipole approximation has been shown both experimentally and theoretically in the linear regime [19,23]; however, the employment of OAM beams in the nonlinear regime requires further investigation. So far, HHG with OAM beams has been studied within the local dipole approximation [46,61], which merely considers the local values of the electric field, ignoring magnetic and higher-order terms in their light-matter coupling treatment.

In this work, we consider the coupling between light and matter beyond the dipole approximation, allowing us to account for the effects of field gradients and magnetic fields. To investigate these effects, we solve the quantum equation of motion for the electron of a hydrogen atom in real space and real time, and inspect the HHG spectrum resulting from

*Contact author: esra-ilke.albar@mpsd.mpg.de

†Contact author: franco.bonafe@mpsd.mpg.de

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different positions of the atom with respect to the beam center. We observe symmetry breaking due to the magnetic field and field gradients, which results in the appearance of even harmonics. Moreover, we resolve the electron trajectories and identify the effect of orbital angular momentum. Our analysis shows that the motion of the driven electron is not confined to the polarization or propagation axes only in the presence of optical vortices. Furthermore, we investigate the angular dependence of the emission of harmonics in the plane orthogonal to the laser propagation, where the off-axis (perpendicular to the laser polarization) component exhibits significant corrections beyond the dipole level. Finally, we show that the yield of such off-axis harmonics can be enhanced by tuning the OAM number of the incident beam.

The paper is organized as follows: In Sec. II, we present the details of the beyond-dipole approach used in this work and provide the expressions for the spectroscopic quantities analyzed thereafter. In Sec. III, we investigate the beyond-dipole effects in high harmonics yield for a case of Bessel beam and compare it to the case of HHG driven by a conventional plane-wave pulse. We also address the influence of orbital angular momentum on the HHG process. Finally, a summary and outlook are given in Sec. IV. The SI system of units is used throughout the paper, unless specified otherwise.

II. METHODS

In this work, we will study the HHG spectrum of the hydrogen atom calculated from the real-time dynamics of the system driven by different external beams. To solve the electron dynamics, we solve the time-dependent Schrödinger equation numerically,

$$i\hbar\partial_t\varphi(\mathbf{r}, t) = \mathcal{H}(t)\varphi(\mathbf{r}, t), \quad (1)$$

using the real-space, real-time octopus code [62]. Here, $\hbar$ is the reduced Planck constant and $\mathcal{H}(t)$ is the electronic Hamiltonian. In octopus, the quantum dynamics of arbitrary systems is implemented using a real-space basis, which is especially useful to describe strong-field phenomena, and has been successfully used to calculate HHG spectra of molecules [63], solids [64], and liquids [65]. Here, we employ a cylindrical simulation box with a radius of 1.32 nm and a length of 3.7 nm. The height of the cylinder is aligned with the polarization direction (x direction) of the laser applied to the system since the largest excursion is expected to take place in this direction. Absorbing boundaries of thickness 0.52 nm with complex absorbing potential are applied to the box to avoid spurious effects due to reflection of the electron [66]. The spacing is set to 0.016 nm, while the time step is 0.0036 fs. For our study, we keep the nucleus of the atom fixed. For time

evolution, we employ the approximated enforced time-reversal symmetry (AETRS) scheme [67]. The electronic Hamiltonian $\mathcal{H}(t)$ is given by

$$\mathcal{H}(t) = -\frac{\hbar^2\nabla^2}{2m_e} + V_{\text{nuc}}(\mathbf{r}, t) + \mathcal{H}_{\text{int}}(t). \quad (2)$$

Here, m_e is the mass of the electron, $V_{\text{nuc}}(\mathbf{r}, t)$ is the Coulomb potential of the nucleus, and $\mathcal{H}_{\text{int}}(t)$ is the interaction Hamiltonian. In this work, we will go beyond the commonly used multipolar expansion [68] and consider the full minimal coupling (FMC) Hamiltonian without truncations [69],

$$\mathcal{H}_{\text{int}}^{\text{f.m.c.}}(t) = -\frac{i\hbar|e|}{cm_e}\mathbf{A}(\mathbf{r}, t) \cdot \nabla + \frac{e^2}{2m_e c^2}\mathbf{A}^2(\mathbf{r}, t). \quad (3)$$

Here, e is the electron charge and c is the speed of light in vacuum. To discern the beyond-dipole effects, we also compare the results obtained with light-matter coupling in the electric dipole approximation in the velocity gauge,

$$\mathcal{H}_{\text{int}}^{\text{d.}}(t) = -\frac{i\hbar|e|}{cm_e}\mathbf{A}(\mathbf{r}_0, t) \cdot \nabla + \frac{e^2}{2m_e c^2}\mathbf{A}^2(\mathbf{r}_0, t), \quad (4)$$

where $\mathbf{r}_0$ is the coordinate of the center of charge of the system (which coincides with the center of mass in this case). In the present work, we restrict ourselves to the case of a Bessel beam to describe the vortex pulse. While we describe the spatial profile of the applied Bessel beam in a micrometer scale, which is much larger than the relevant scale of the electron dynamics [see Fig. 1(a) below], the electronic simulation box is shifted in space to the location of interest, and therefore the electronic Hamiltonian considers only the corresponding spatial portion of the beam that overlaps with the electronic simulation box.

Since circularly polarized beams do not generate high harmonic radiation in systems with inversion symmetry, we construct the Bessel-beam counterpart of linearly polarized plane-wave beams as [32,70,71]

$$A_{\text{tw}}^{3\text{C}}(\mathbf{r}, t) = \frac{1}{\sqrt{2}}[A_{m_{\gamma_1}, \lambda=+1, \theta_k}^{\text{tw}}(\mathbf{r}, t) + A_{m_{\gamma_2}, \lambda=-1, \theta_k}^{\text{tw}}(\mathbf{r}, t)], \quad (5)$$

where $m_{\gamma_1} - m_{\gamma_2} = 2$, m_{γ} is total angular momentum, $\lambda = \pm 1$ is the helicity, and $\theta_k = \arctan(\frac{\kappa}{k_z})$ is the so-called opening angle, with k_z and $\kappa = \sqrt{(\frac{\omega}{c})^2 - k_z^2}$ being the longitudinal and transversal components of the wave vector $\mathbf{k}$. Here,

$$A_{m_{\gamma}, \lambda, \theta_k}^{\text{tw}}(\mathbf{r}, t) = f(z)[A_{m_{\gamma}, \lambda, \theta_k, x}^{\text{tw}}(\mathbf{r}, t), A_{m_{\gamma}, \lambda, \theta_k, y}^{\text{tw}}(\mathbf{r}, t), A_{m_{\gamma}, \lambda, \theta_k, z}^{\text{tw}}(\mathbf{r}, t)]$$

is the vector potential in the Coulomb gauge describing a Bessel beam propagating in the z direction,

$$\begin{aligned} A_{m_{\gamma}, \lambda, \theta_k, x}^{\text{tw}} &= \frac{A_{\text{tw}}}{\sqrt{2}} \{ \cos[k_z z + \phi(m_{\gamma} + 1) - \omega_b t + \Phi_{\text{tw}}] d_{-1\lambda}^1(\theta_k) J_{m_{\gamma}+1}(\kappa r_{\perp}) \\ &\quad + \cos[k_z z + \phi(m_{\gamma} - 1) - \omega_b t + \Phi_{\text{tw}}] d_{1\lambda}^1(\theta_k) J_{m_{\gamma}-1}(\kappa r_{\perp}) \}, \\ A_{m_{\gamma}, \lambda, \theta_k, y}^{\text{tw}} &= \frac{A_{\text{tw}}}{\sqrt{2}} \{ \sin[k_z z + \phi(m_{\gamma} + 1) - \omega_b t + \Phi_{\text{tw}}] d_{-1\lambda}^1(\theta_k) J_{m_{\gamma}+1}(\kappa r_{\perp}) \\ &\quad - \sin[k_z z + \phi(m_{\gamma} - 1) - \omega_b t + \Phi_{\text{tw}}] d_{1\lambda}^1(\theta_k) J_{m_{\gamma}-1}(\kappa r_{\perp}) \}, \\ A_{m_{\gamma}, \lambda, \theta_k, z}^{\text{tw}} &= A_{\text{tw}} \sin(k_z z + m_{\gamma} \phi - \omega_b t + \Phi_{\text{tw}}) d_{0\lambda}^1(\theta_k) J_{m_{\gamma}}(\kappa r_{\perp}), \end{aligned} \quad (6)$$

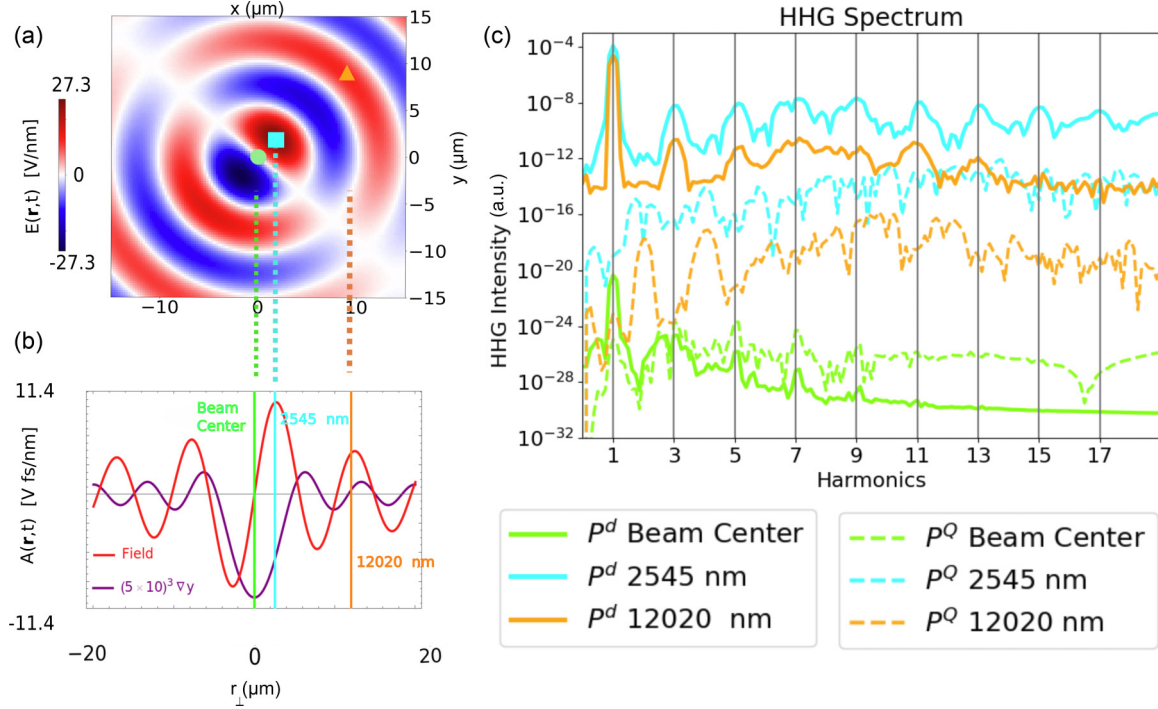


FIG. 1. (a) Three locations chosen for the impact parameter study are shown across the transversal plane depicting the electric field $E_{\text{tw}}(\mathbf{r}, t) = -\frac{\partial A_{\text{tw}}^{1\text{C}}(\mathbf{r}, t)}{\partial t}$ of the Bessel beam. The one-component Bessel beam $A_{\text{tw}}^{1\text{C}}$ with orbital angular momentum number $m = 1$ has a singularity at the middle of its cross section due to the cancellation of the fields, referred to as the beam center and marked in green. A hydrogen atom is first placed in this position, which results in the absence of driving electric and magnetic fields. However, the maximum of the field gradient occurs here, as seen in (b), where the amplitude $A_{\text{tw}}^{1\text{C}}$ and the spatial gradient in terms of the y position of the corresponding vector potential $A_{\text{tw}}^{1\text{C}}$ are shown as a function of $r_{\perp}$. The other locations are the hot spot located in the first ring of the Bessel beam ($r_0 = 2545$ nm away, noted by cyan color, with the highest field amplitude and nonzero gradient) and a point in the third ring ($r_0 = 12020$ nm away, marked in orange; field amplitude is still considerable and the gradient is zero). (c) The HHG spectra in full minimal coupling from all three locations. Solid lines represent the dipolar emission spectrum P^d [see Eq. (11)], while dashed lines refer to the quadrupolar P^Q one [see Eq. (12)]. The presented results are the calculated beyond-dipole approximation. Here and everywhere throughout the paper, the yields below $\approx 10^{-35}$ are considered to be numerically zero.

where Φ_{tw} is a constant phase, and A_0 and ω_b are the amplitude and central frequency of the beam, respectively. $J_m(\kappa r_{\perp})$ represents the Bessel function, $r_{\perp} = \sqrt{x^2 + y^2}$, $\phi = \arctan(y/x)$ is the azimuthal angle, and $d_{\mu\lambda}^1(\theta_k)$ is a small Wigner matrix given by [72]

$$d_{\mu\lambda}^1(\theta_k) = \begin{cases} \frac{\sin \theta_k}{\sqrt{2}} \lambda & \text{if } \mu = 0 \\ \frac{1 + \lambda \cos \theta_k}{2} & \text{if } \mu = 1 \\ \frac{1 - \lambda \cos \theta_k}{2} & \text{if } \mu = -1. \end{cases} \quad (7)$$

Here, $f(z)$ is the real-space envelope function along the propagation z axis,

$$f(z) = \sin^2 \left(\frac{\pi(z - R_0)}{w} \right) \theta(z - R_1) \theta(R_0 - z), \quad (8)$$

where $z = ct$, $\theta(z)$ is the Heaviside step function, R_0 and R_1 define the spatial extent of the envelope in real space, and $w = 8\lambda_w$ with $\lambda_w = 800$ nm being the central wavelength of the laser field. This choice ensures that the pulse contains eight optical cycles, corresponding to a duration of 21.3 fs.

We note that polarization of $A_{\text{tw}}^{3\text{C}}(\mathbf{r}, t)$ has, in general, a complex structure and therefore $A_{\text{tw}}^{3\text{C}}(\mathbf{r}, t)$ has three components (which are specified by index 3C). Physically, this

twisted-light field $A_{\text{tw}}^{3\text{C}}(\mathbf{r}, t)$ can be viewed as the analog of linearly polarized light since, in the paraxial limit (when the opening angle tends to zero, $\theta_k \rightarrow 0$), it reduces to a field with a single transverse component along the x axis,

$$A_{\text{tw}}^{1\text{C}}(\mathbf{r}, t) \approx A_{\text{tw}} f(z) J_m(\kappa r_{\perp}) \cos(k_z z + m\phi - \omega_b t + \Phi_{\text{tw}}) \mathbf{e}_x. \quad (9)$$

Here, $\mathbf{e}_x = (1, 0, 0)$ and we used the fact that in the paraxial approximation, $m_{\gamma_1} \approx m + \lambda = m + 1$ and $m_{\gamma_2} \approx m + \lambda = m - 1$, with m being the projection of OAM. Here we would like to note that the vector potential $A_{\text{tw}}^{1\text{C}}(\mathbf{r}, t)$ only approximately obeys the Coulomb gauge [73]. In Sec. III B, we discuss in detail observables which are strongly affected by the choice between $A_{\text{tw}}^{1\text{C}}(\mathbf{r}, t)$ and $A_{\text{tw}}^{3\text{C}}(\mathbf{r}, t)$, indicating the importance of carefully accounting for all three components of the vector potential. We note that in experiments, Bessel-Gauss beams with finite transverse extent are typically used. However, since in the present study a single atom is affected only by the local spatial profile of the beam, an ideal Bessel beam is sufficient to describe the spatial profile in the vicinity of the atom.

To understand the effects of OAM, we compare the results with the HHG calculation employing a plane-wave pulse with explicit space dependence, namely, the following:

$$A_{\text{pw}}(\mathbf{r}, t) = A_{\text{pw}} f(z) \cos(k_z z - \omega_b t) \mathbf{e}_x. \quad (10)$$

The plane wave is arranged to have the same field amplitude, wavelength (800 nm), phase, and envelope as the Bessel beam with $m = 1$ at the position $\mathbf{r} = (r_\perp, 45^\circ, 0)$ with $r_\perp = 2545$ nm (as this condition will be used throughout the article; see, for details, Appendix B). The resulting maximum electric field amplitude $E_{\text{pw}} = \omega_b A_{\text{pw}}$ is 2.72×10^{10} V/m.

The HHG yield is calculated from the dipolar emission P^d , i.e., the energy dissipated per unit frequency [74,75],

$$P^d = \sum_{i=x,y,z} P_i^d = \frac{\mu_0 \omega^4}{12\pi c} \sum_{i=x,y,z} \tilde{d}_i^2(\omega), \quad (11)$$

as well as quadrupolar contribution to the emission P^Q [74,75],

$$P^Q = \sum_{i,j=x,y,z} P_{ij}^Q = \frac{\mu_0 \omega^6}{1440\pi c^3} \sum_{i,j=x,y,z} \tilde{Q}_{ij}^2(\omega). \quad (12)$$

Here, μ_0 is the magnetic vacuum permeability, $\tilde{d}_i(\omega)$ is the Fourier transformation of the i th component of the time-dependent dipole response $d_i(t)$,

$$d_i(t) = |e| \langle \varphi(\mathbf{r}, t) | r_i | \varphi(\mathbf{r}, t) \rangle, \quad (13)$$

and $\tilde{Q}_{ij}(\omega)$ is the Fourier transform of the time-dependent quadrupole moments $Q_{ij}(t)$ related to directions i and j ,

$$Q_{ij}(t) = |e| \langle \varphi(\mathbf{r}, t) | (3r_i r_j - r^2 \delta_{ij}) | \varphi(\mathbf{r}, t) \rangle, \quad (14)$$

where δ_{ij} is the Kronecker delta.

III. RESULTS

A. Position dependence of the HHG spectra

As the electric field of the OAM beams has a spatially inhomogeneous structure, the HHG yield depends on the atom's position relative to the center of the beam. Therefore, we first investigate this dependence for the case of the Bessel beam $A_{\text{tw}}^{\text{IC}}$ (here and throughout the paper, we omit the explicit dependence of the vector potential on $\mathbf{r}$ and t for brevity) with $m = 1$. We consider three positions of the atom $\mathbf{r}_0 = (r_0, 45^\circ, 0)$, namely, beam center ($r_0 = 0$ nm), first ring (also called *hot spot*, $r_0 = 2545$ nm), and third ring ($r_0 = 12020$ nm), as depicted in Fig. 1(a). The path connecting these points was chosen so that the beyond-dipole effects have equal contributions of the field gradients in the x and y directions, as this simplifies the analysis of the results; see below.

For these positions, we have calculated the harmonic spectra from the electric dipole P^d and electric quadrupole P^Q terms. We have also calculated the magnetic dipole emission, which is found to be negligible (see the Supplemental Material, Fig. S2 [76]) and was not considered for this discussion. Note that we also adjust the vector potential phase to be the same for all three positions, to avoid spurious effects arising from phase differences.

As one can see from Fig. 1(b), the electric field of the beam described by $A_{\text{tw}}^{\text{IC}}$ in the beam center is zero, while the field gradient is maximum. This results in a quadrupolar P^Q emission being comparable to the dipolar P^d counterpart, as seen in Fig. 1(c). We note that since the spectra are calculated within the FMC approach, the dipole emission for the beam center is nonzero and is caused by the higher-order terms in the interaction Hamiltonian $\mathcal{H}_{\text{int}}^{\text{f.m.c.}}(t)$ [Eq. (3)]. However, due to the absence of field amplitude, the HHG yield at the beam center is very weak and with a low cutoff. On the other hand, the HHG yield for the atom located at the hot spot is stronger and has a cutoff reaching the 20th peak, in agreement with the three-step model prediction for the peak amplitude of the electric field, even though the peaks start to decrease after the 15th harmonic. While the dipolar emission follows the well-known selection rules and peaks only at odd harmonics, quadrupolar emission peaks at the even harmonics due to symmetry breaking, in agreement with previous findings [75]. Finally, at the position in the third intensity ring ($r_0 = 12020$ nm), the Bessel beam has a smaller amplitude in comparison to the hot spot and almost zero gradients [see Fig. 1(b)]. Less field amplitude naturally results in an earlier cutoff, which is found at the 11th harmonic, as well as weaker peaks. While the field gradient effects are minimal, the magnetic field effect causes quadrupolar yield at even harmonics in this case also. Quadrupolar emission increases with higher harmonics due to its ω^6 dependence [see Eq. (12)], reaching considerable yields at the hot spot, owing to the larger cutoff. We can infer that the quadrupolar emission (which is fully addressed only within beyond-dipole approaches) will become even more prominent at higher intensities which will entail higher harmonics. This yields a fresh perspective confirming the necessity of using beyond-electric-dipole approximation as the intensity increases [77].

Since the HHG spectrum at the hot spot is the strongest [see Fig. 1(c)] and there is a nonzero field gradient [see Fig. 1(b)], we choose this position of the atom for further consideration.

B. Beyond-dipole effects and components of the dipole emission

We now compare the spectra resulting from a Bessel beam $A_{\text{tw}}^{\text{IC}}$ and a plane wave to identify the OAM effects. In Fig. 2, we show frequency-resolved dipole component spectra (P_x^d , P_y^d , and P_z^d) induced by the plane wave A_{pw} and Bessel beam $A_{\text{tw}}^{\text{IC}}$, in the first and second column, respectively. When the coupling Hamiltonian is in dipole approximation (velocity gauge), the spectra of the Bessel beam and plane wave are indistinguishable (see the Supplemental Material, Sec. A [76]). This limitation of the electric dipole approximation stems from the inability to capture the spatial dependence of the beam, which is crucial for OAM beams. To go beyond this limitation, in Fig. 2 we show the results in full minimal coupling. The dipolar spectra reveal that the Bessel beam can excite all dipole components, while the plane wave only excites the dipole in the x and z directions. The dipole component in the y direction, i.e., the direction of propagation for both beams, is a clear result of beyond-dipole coupling, as it has been shown by computing the Lorentz force in the direction of propagation, which is proportional to the $\nabla_z A_{\text{tw/pw}}$ [56,57]. The P_z^d and P_y^d spectra, arising from beyond-dipole

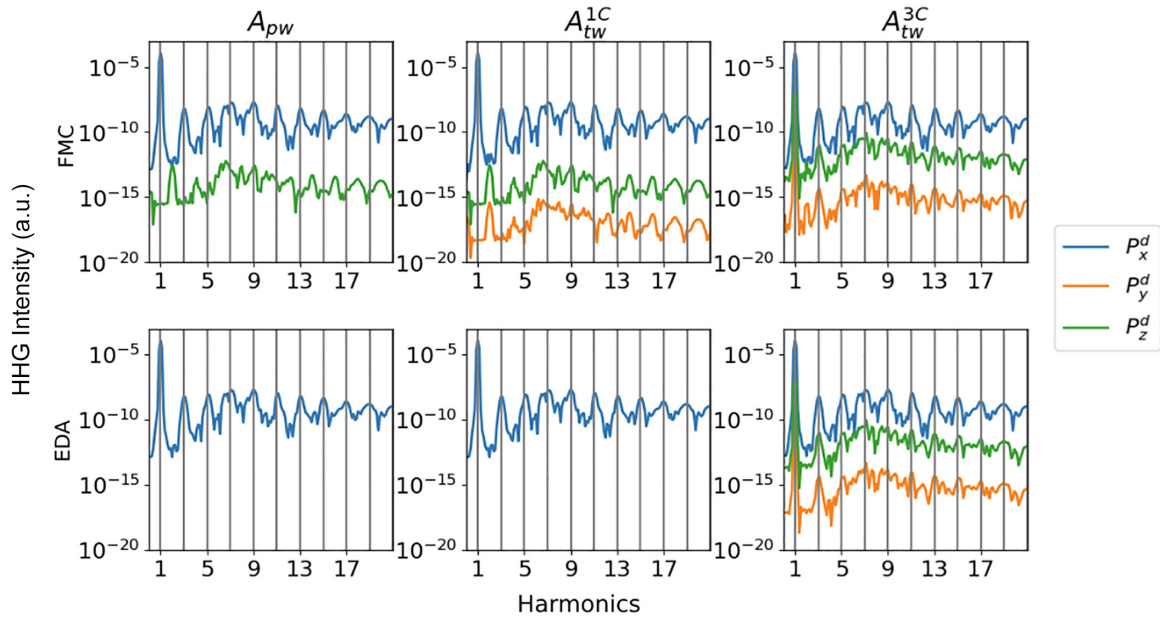


FIG. 2. Dipole component-resolved spectra for a plane-wave beam and Bessel beam for the case when the atom is located at $r_0 = 2545$ nm (hot spot). At this point, the field amplitude, frequency, phase, and envelope of the x component of the Bessel beam with all three components (A_{tw}^{3C} , the Bessel beam with one component, A_{tw}^{1C}) and plane wave match. The upper panels depict the dipolar emission from all three directions calculated with full minimal coupling (FMC), while the lower panels are reserved for the emission calculated within the electric dipole approximation (EDA).

corrections, both show larger amplitudes at the even harmonics, in contrast with the P_x^d spectrum that is dominated by the dipole interactions. The fact that spectra induced by the Bessel beam have contributions from components other than the beam polarization (x) direction shows the need for a beyond-dipole approach to resolve the spectrum thoroughly.

To understand the origin of the y and z contributions to the spectra (hereafter called *off-axis* contributions) excited by the linear Bessel beams A_{tw}^{1C} , we show the dipole moment dynamics $d_i(t)$ (13) in Fig. S6 of the Supplemental Material [76]. The dipole moment $d_i(t)$ shown in Eq. (13) yields the expectation value of the position of the electron along the Cartesian direction i . Therefore, it is referred to as the trajectory. An animation of the trajectory depicted in Fig. S6 of the Supplemental Material [76] can be retrieved from Ref. [78]. The origin of the off-axis spectral signals can be linked to the electron's trajectory as follows: As the electron travels further away from the ion, it accelerates and reaches a higher velocity. This results in a larger Lorentz force acting on the electron, which in turn causes deviations from a linear path constrained to the polarization axis of the incoming beam [54]. Hence, considering only the electric field amplitude (dipolar coupling) when calculating the electron's trajectory is insufficient [79,80].

As expected, the contributions in the z axis (arising from the longitudinal field gradients) for the linear Bessel beam A_{tw}^{1C} and plane-wave pulse are identical since the electric and magnetic field amplitudes, as well as the field gradients in the direction of propagation at the vicinity of the atom, of both fields are equal (see Fig. 2). In contrast, we observe that in addition to the magnetic-field-induced figure-eight motion in the xz plane, the OAM-carrying Bessel beam induces changes

in the electron trajectory in all directions due to the xy -plane field gradients (see Fig. S6 in the Supplemental Material [76]), which affects the angular dependence of the emission (see Sec. III C). The dynamics of the d_y dipole component can be understood as an effect of the $\nabla_y A_{tw/pw}$ field gradients, which are nonzero only for the structured beams. This means that by measuring the P_y component of the radiated emission, the effect of the transverse gradients could be understood. As can be seen in Fig. 2, specifically for the A_{tw}^{1C} case, the effects of transverse gradients (P_y) are much smaller than the ones from longitudinal gradients (P_z ; see the Supplemental Material, Fig. S5 [76]), which is known for paraxial beams. See Appendix A for an analytical expression of the field gradients of the Bessel beams.

So far, we have used the approximated expression (9) for the vector potential that disregards the y and z components. While these components are small for the opening angle used here, their effect can significantly contribute [19,23,68,81]. Thus, Ref. [19] demonstrated experimentally, and Ref. [23] explained theoretically, that even for paraxial beams, predictions neglecting the longitudinal field fail dramatically in the case of counterrotating spin and orbital angular momentum. To investigate the influence of these usually disregarded components, we compare the frequency-resolved dipole component spectra calculated for the case of A_{tw}^{3C} ; see Fig. 2, third column. For dipolar contribution P_x^d (the dominant contribution to the total emission P^d), both A_{tw}^{3C} and A_{tw}^{1C} yield the same result.

We also note that the first harmonic (the largest contribution) in both P_y^d and P_z^d emission spectra in the case of driving with A_{tw}^{1C} is absent because the x polarized field alone cannot efficiently drive dipoles in the respective directions. Since the

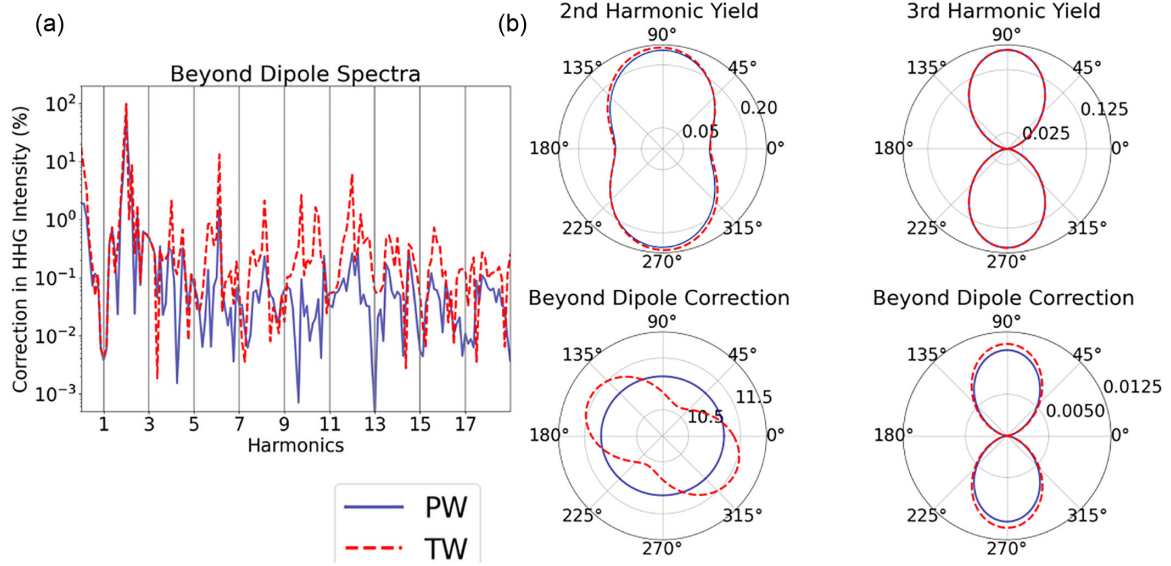


FIG. 3. (a) We calculate the spectra first using the full minimal coupling approach and then with the electric dipole approximation for two cases: a Bessel beam with three components, A_{tw}^{3C} , (OAM), and a plane wave (PW). We report the difference between the two spectra in percentage. Beyond-dipole corrections peak at even harmonics, demonstrating the necessity of beyond-dipole approaches to capture symmetry-breaking effects. (b) Angular emission spectra in the xy plane $P^d(\omega, \theta)$ for second and third harmonics. The panels on the left depict the results from the second harmonic, while the ones on the right show the results from the third harmonic. The upper panel from the left describes the angular distribution of total emission induced by a PW and Bessel beam. In contrast, the lower left one describes the full minimal coupling correction compared to the dipole approximation. Similarly, the right side shows total emission induced by a plane wave (PW) and Bessel beam for the third harmonic on the upper part, and the beyond-dipole correction on the lower panel. Results here are normalized by total emission induced by the plane wave $P_{pw}^d(\omega) = \int d\Omega P_{pw}^d(\omega, \theta)$, where $d\Omega = \sin\theta d\theta d\phi$.

Bessel beam described by A_{tw}^{3C} is more realistic and provides insight into dipole-level and beyond-dipole effects on all components of the emission, we will use A_{tw}^{3C} for the remainder of this work. We have also analyzed the quadrupolar emission comparing Bessel beams A_{tw}^{3C} and plane waves (see the Supplemental Material, Fig. S4 [76]). While the quadrupolar nature of the Bessel beam provides access to all quadrupolar components, the plane wave is missing the cross components $\tilde{Q}_{yz}(\omega)$ and $\tilde{Q}_{xy}(\omega)$ due to the absence of field gradients in the y direction. Also in the Supplemental Material (Fig. S3) [76], we have included a full view of two sets of dipolar emission spectra, where the components with negligible yield (numerically nonzero) can be seen for comparison.

C. Angular dependence of the beyond-dipole HHG emission

Now we study the beyond-dipole corrections caused by the structured fields in more detail. In Fig. 3(a), we show the normalized difference between the total HHG spectra calculated at the FMC level with respect to the EDA coupling, shown in Fig. 2. We show the results for the A_{tw}^{3C} Bessel beam (TW) and the plane-wave cases (PW). In both of them, the correction is expressed as a percentage. Notably, the beyond-dipole correction of the second harmonic peak reaches 100% both in the PW and TW cases. This correction stems from the magnetic field component in the z direction, which acts on the electron with the Lorentz force and causes a drift in propagation direction in both cases. This action is commonly referred to as the radiation pressure. However, for larger even harmonics (6th, 8th, 10th, and 12th harmonics, mainly), the

correction in the TW case reaches between 2% and 10%, and it is much larger than the PW difference. Since the corrections due to the gradients in the z direction are equal for both beams, these differences for larger even harmonics can be attributed to the gradients in the xy plane, i.e., to the spatial structure of the Bessel beam.

Next, we focus the analysis on the beyond-dipole corrections to the angular distribution of the resulting emission in the far field, $P^d(\omega, \theta)$. It is defined by $P^d(\omega) = \int d\Omega P^d(\omega, \theta)$, with $d\Omega = \sin\theta d\theta d\phi$ being the solid angle, and $P^d(\omega, \theta)$ can be calculated as follows:

$$P^d(\omega, \theta) = \frac{\mu_0 |\tilde{\mathbf{d}}(\omega)|^2 \omega^4}{32\pi^2 c} \sin^2 \theta, \quad (15)$$

where θ is the angle between the dipole moment $\tilde{\mathbf{d}}(\omega)$ and the direction of observation. For visualization, the expression is then normalized by the total emission driven by plane-wave pulse $P_{pw}^d(\omega) = \int d\Omega P_{pw}^d(\omega, \theta)$.

In Fig. 3(b), we present the dipole emission of the second and third harmonics in the top-left and top-right panels, respectively, resolved in the azimuthal angle in the xy plane (orthogonal to the propagation direction of the beams). Additionally, we present the angular-resolved beyond-dipole corrections in the xy plane in the bottom panels. The emission resulting from both the plane wave and the Bessel beam is seen in the upper-left panel, calculated in the full minimal coupling. The difference in the angular dependence of the normalized $P^d(\omega, \theta)$ for the Bessel beam and the plane wave is larger in the case of even harmonics, as evidenced by the second harmonic. As can be seen in the upper-left panel,

the amplitude of emission due to the OAM beam is tilted with respect to the plane wave.

By inspecting the difference of emission calculated within dipole approximation from that in the full minimal coupling approach (beyond-dipole correction, lower-left panel), it is clear that the angular difference is due to field gradient effects (off-axis motion), reaching up to 11.5% difference in amplitude and a slight tilt due to the P_y . A correction for the plane-wave case is still present due to the z gradients, and hence the correction has no angular dependence in the xy plane (see Fig. 3).

The corrections become much smaller for the odd harmonics, as seen in the case of the third harmonic angular spectrum (upper-right panel). Here, the dipolar contribution dominates heavily and renders the beyond-dipole correction small (of the order of 0.01%, as seen in the beyond-dipole correction, lower-right panel). Hence, the plane-wave correction in the second harmonic is around 100 times larger than that of the third harmonic.

Observing the large corrections in even harmonics is exciting for experimental realizations. Such corrections entailed by OAM beams have been reported for large odd harmonics before [82]. As seen in Fig. 1, quadrupolar effects dominate even harmonics; therefore, such harmonics are good candidates for the investigation of beyond-dipole contributions, as previously reported [75]. Added to the possibility of measuring even harmonics without dipolar background [60], we believe the considerable correction in the emission visible in the second harmonic yield given in Fig. 3(b) due to the deviations in the electron trajectory confirms that the second harmonic emission is a fitting candidate for an experimental observable, which provides a playground where the effect of OAM can be used to induce emission in all directions. While such a correction seen in Fig. 3 is not limited to an OAM-carrying Bessel beam, the correction for the plane wave is azimuthally symmetric, hinting that an angular deviation is inherent to incident OAM.

D. Dependence of the off-axis emission on the orbital angular momentum

Finally, to investigate the effect of the orbital angular momentum number, we construct A_{tw}^{3C} Bessel beams with higher m . We place the atom at the first intensity ring for each case of m , with the amplitude and phase of the beams matched in the x direction, so that the dipolar effects are identical for all the beams. The chosen parameters used for all beams are reported in Appendix B.

In Fig. 4, we show the y dipole component spectra P_y^d normalized by the $m = 1$ case, for values of m ranging from 1 to 21. As shown in Fig. 2, the P_y^d spectra provide unequivocal evidence of effects arising from in-plane spatial structuring of the beams. Here, it becomes clear that this quantity is also sensitive to the OAM number.

The top panel of Fig. 4 shows y dipole component spectra P_y^d as a function of OAM number m for the second harmonic, while the top panels show the same quantity for the third harmonics. While the second harmonic yield shows a monotonous increase with m , the third harmonic yield drops strongly for m from 1 to 3, and then increases. It is observed

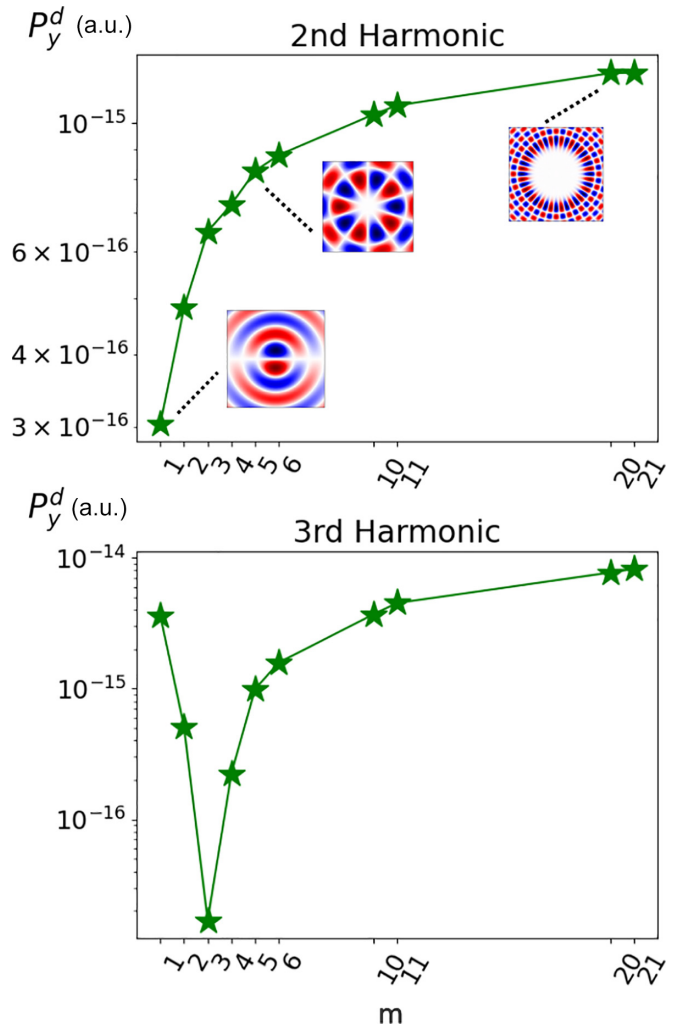


FIG. 4. We present the y dipole component spectra P_y^d for m ranging from 1 to 21 for the second and third harmonics. It is visible that P_y^d drastically depends on m . Here, the change of m affects mostly the second harmonic. Both harmonics show a saturation regime after $m = 10$. Here, the inset in the second harmonic graph displays the cross sections of beams with $m = 1$, $m = 5$, and $m = 20$. The presented results are calculated beyond the dipole approximation.

that a saturation regime is reached for larger values of m in both cases. This can be understood by the field gradient following the same trend for increasing values of m (see the Supplemental Material, Fig. S7 [76] where the y derivative of the electric field at the hot-spot position of each beam is plotted). In addition to the P_y^d dipole component, we calculated the angular emission spectra for different values of m for the second and third harmonic (see the Supplemental Material, Fig. S8 [76]). It can be seen that for the second harmonic, both the angle and intensity of the emission depend on the OAM number, while for the third harmonic, only the intensity increases. These findings show that the nonlinear response for the even harmonics depends on the OAM number of the beam, following a similar trend to the field gradients, which provides a path to include beyond-dipole effects in simulations of HHG from the gas-phase target.

TABLE I. Parameters used for the calculations of $A_{\text{tw}}^{1\text{C}}$ and $A_{\text{tw}}^{3\text{C}}$.

m	A_{tw} (a.u.)	r_0 (nm)	Φ_{tw}
1	0.0455	2545	1.4915
2	0.0543	4596	3.8481
3	0.0608	6081	6.2033
4	0.0661	7920	2.2779
5	0.0705	9334	4.6344
6	0.0745	10960	0.7059
10	0.0872	17324	3.8516
11	0.0898	18738	6.2082
20	0.1084	32527	2.2816
21	0.1102	33941	4.6332

IV. CONCLUSION

To conclude, we identify beyond-dipole effects in HHG in the presence of OAM beams. We predict that such corrections are particularly relevant for even harmonics, which appear due to symmetry breaking. We show that the dynamics, driven by the in-plane gradients of the structured fields, are well captured beyond the dipole approximation and lead to a tilt in the angular distribution of the emitted radiation for the second harmonic, a trend that can be generalized to even harmonics.

We also investigate the influence of the (often neglected) components of the vector potential for twisted beams and show that accounting for them is crucial when analyzing component-resolved spectra. Finally, we show that the harmonic yield in both off-axis and on-axis directions can be enhanced by tuning the OAM number of the Bessel beam, where the corrections of the even harmonics (as evidenced by the second harmonic yield) follow the trend of the electric field gradients. Future calculations of propagation in a medium, accounting for both microscopic and macroscopic effects, will provide further insight into experimental setups

for detecting and harnessing beyond-dipole effects in nonlinear spectroscopy with structured light.

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The authors declare no conflicts of interest relevant to the content of this article.

DATA AVAILABILITY

The data that support the findings of this article are openly available [83].

APPENDIX A: GRADIENT FROM VECTOR POTENTIAL

Here we give an explicit expression for the gradient of the linearly polarized Bessel beam $A_{\text{tw}}^{1\text{C}}(\mathbf{r}, t) = A_{\text{tw}}^{1\text{C}}(\mathbf{r}, t)\mathbf{e}_x$ from Eq. (9),

$$\nabla_y A_{\text{tw}}^{1\text{C}}(\mathbf{r}, t) = A_{\text{tw}} f(z) \left(\frac{\kappa y [J_{m-1}(r_{\perp} \kappa) - J_{m+1}(r_{\perp} \kappa)] \cos(m\phi + kz - \omega t)}{2r_{\perp}} - \frac{mx J_m(r_{\perp} \kappa) \sin(m\phi + kz - t\omega)}{x^2 + y^2} \right). \quad (\text{A1})$$

APPENDIX B: PARAMETERS FOR THE BESSEL BEAMS

For the HHG spectra driven by $A_{\text{tw}}^{1\text{C}}$, the parameters were chosen such that the field amplitude A_{tw} at the atomic position matches the plane-wave amplitude value $A_{\text{pw}} = 0.053$ a.u. at $t = 0$. The atom is placed at $\mathbf{r}_0 = (r_0, 45^\circ, 0)$ with r_0 adjusted for each OAM number m to locate the atom at the first intensity ring of the beam (hot spot). To match the total phase Φ_{pw}

of the plane wave at $t = 0$ and $z = 0$, we calculate Φ_{tw} from the condition

$$m\phi + \Phi_{\text{tw}} = 0, \quad (\text{B1})$$

where $\phi = 45^\circ$. For the case of $A_{\text{tw}}^{3\text{C}}$, we used the same set of parameters as for $A_{\text{tw}}^{1\text{C}}$, see Table I. For the case of $A_{\text{tw}}^{3\text{C}}$, we use the following formulas for the total angular momentum: $m_{\gamma_1} \approx m + 1$ and $m_{\gamma_2} \approx m - 1$.

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