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High harmonic generation with Laguerre–Gaussian beams

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Abstract

We summarize the development of high harmonic generation (HHG) with linearly polarized Laguerre–Gaussian (LG) beams and their superpositions to explain the non-perturbative aspects of HHG. Furthermore, we show that circularly polarized extreme ultraviolet vortices with well-defined orbital angular momentum (OAM) can be generated by HHG with bicircular LG beams. We introduce photon diagrams in order to explain how to calculate the OAM and the polarization of the generated harmonics by means of simultaneous conservation of spin angular momentum and OAM. Moreover, we show how the intensity ratio of the driving fields in HHG with bicircular LG beams further enhances the generation of circularly polarized twisted attosecond pulse trains.

Keywords: high-harmonic generation, optical vortices, strong field approximation, orbital angular momentum

(Some figures may appear in colour only in the online journal)

1. Introduction

Light beams can carry spin (SAM) and orbital angular momentum (OAM) [1]. While the SAM is known to refer to the polarization of the (light) beam, the OAM characterizes the spatial field distribution and the formation of helical wave fronts. Because of their helical phase fronts these beams are also often referred to as *twisted* beams. It was shown by Allen *et al* [2] that Laguerre–Gaussian (LG) beams carry both, SAM and OAM. The OAM of LG beams expresses itself through an azimuthal dependent phase $e^{i\ell\varphi}$, where ℓ is the OAM of the beam. The generation of these beams has opened a wide range of novel applications such as optical communication [3], particle trapping [4], microscopy [5], quantum optics [6] or the interaction with atoms and molecules [7, 8]. This interaction of twisted light with matter includes, in particular, high harmonic generation (HHG) [9–11], a nonlinear process in

strong laser fields ($I \sim 10^{14} \text{ W cm}^{-2}$) which was first observed in solids [12] and later also in rare gases [13, 14]. Nowadays, HHG is widely applied as a table top source of coherent extreme ultraviolet (XUV) radiation. The underlying physical process of HHG can be explained in terms of the three step model of strong field physics [15]: (i) a strong laser field irradiates an atom and suppresses the atomic potential, which makes it possible for an electron to be released by tunnel ionization. (ii) The released electron will be driven away from the parent ion due to the strong laser field. (iii) Since the laser field is oscillating, the electron can be driven back to its parent ion and recombine under the emission of a high energetic photon. However, if HHG is driven by beams with helical phase fronts, we need to consider the twofold nature of HHG, namely the microscopic interaction of the individual atoms with the intense laser field and as well the (macroscopic) superposition of the radiation emitted from different atoms at the detector, to determine the OAM and the divergence of the emitted harmonics. During recent years, the generation of XUV vortices by means of HHG became indeed an important research topic.

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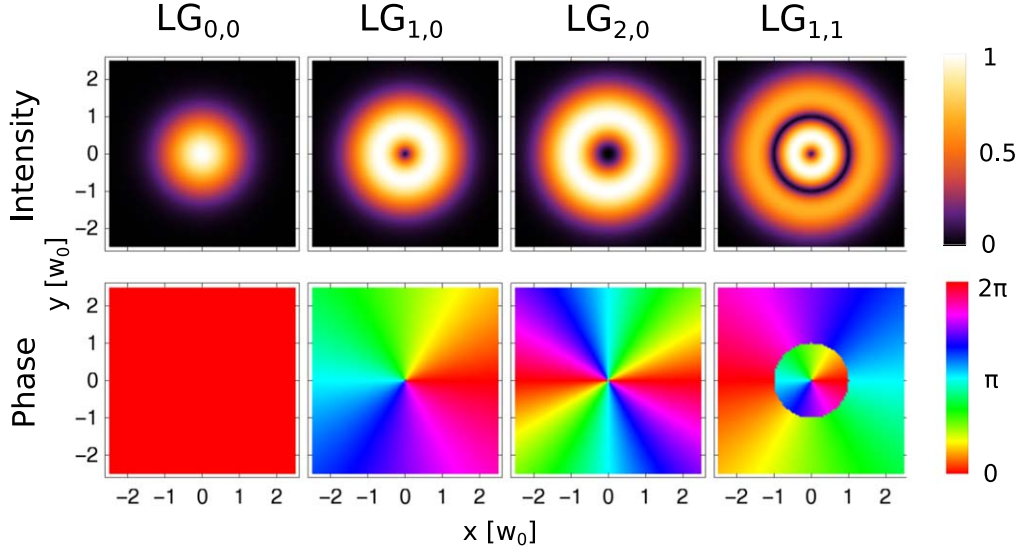


Figure 1. Transverse cross sections of the intensity and phase profiles of different LG beams at $z = 0$. (Left) Since the intensity maximum of a Gaussian beam is on the beam axis and since it has no OAM, the phase is independent on the azimuthal coordinate. Middle columns: LG beams with angular momentum have zero intensity at the beam axis. The OAM induces a phase shift of $\ell \cdot 2\pi$ along the azimuthal coordinate. (Right) If the radial node index is increased, the intensity cross section will exhibit additional rings.

Within this work, we shall first briefly outline the underlying theory to describe HHG with twisted light beams. We then summarize the most recent findings of HHG with linearly and circularly polarized beams and show how the OAM of the emitted harmonics can not only be manipulated but can be tailored quite precisely by polarization, intensity and OAM of the driving laser field.

Atomic units are used throughout the paper unless stated otherwise ($m_e = \hbar = e = 4\pi\epsilon_0 = 1$).

2. Theoretical model of HHG with LG beams

2.1. Description of LG beams

Before we consider HHG with LG beams, we will give a short derivation of their vector potential. Thereby, we consider the spatial part of a vector potential

$$\mathbf{A}(\mathbf{r}, t) = \mathbf{A}(\mathbf{r})e^{-i\omega t} \quad (1)$$

of a beam with frequency ω that propagates in z direction [16]

$$\mathbf{A}(\mathbf{r}) = \mathbf{e}_\lambda u(\mathbf{r})e^{ikz}, \quad (2)$$

with a polarization vector $\mathbf{e}_\lambda$, the spatial amplitude $u(\mathbf{r})$ and wave number $k = \alpha\omega$, and where α is the fine-structure constant. Generally electromagnetic waves are solutions to the Helmholtz equation

$$(\nabla^2 + k^2)\mathbf{A}(\mathbf{r}) = 0. \quad (3)$$

However, if the z derivative of the spatial amplitude $u(\mathbf{r})$ varies sufficiently slowly with z , such that

$$\left| \frac{\partial^2 u}{\partial z^2} \right| \ll \left| k \frac{\partial u}{\partial z} \right| \quad \text{and} \quad \left| \frac{\partial^2 u}{\partial z^2} \right| \ll |\nabla_\perp^2 u| \quad (4)$$

the Helmholtz equation (3) can be rewritten in its paraxial

approximation

$$\nabla_\perp^2 u(\mathbf{r}) + 2ik \frac{\partial u(\mathbf{r})}{\partial z} = 0. \quad (5)$$

A well-known set of solutions to the paraxial Helmholtz equation (5) are LG beams, whose amplitude can be expressed in cylindrical coordinates as:

$$LG_{\ell,p}(\rho, \varphi, z) := u(\mathbf{r}) = E_0 \frac{W_0}{W(z)} \left(\frac{\sqrt{2}\rho}{W(z)} \right)^{|\ell|} L_p^{|\ell|} \left[\frac{2\rho^2}{W^2(z)} \right] \times \exp \left(-\frac{\rho^2}{W^2(z)} \right) \exp \left(ik \frac{\rho^2}{2R(z)} + i\Phi_G(z) + i\ell\varphi \right). \quad (6)$$

Here W_0 is the beam waist, $W(z) = W_0 \sqrt{1 + \frac{z^2}{z_r^2}}$ is the beam width and $z_r = k \frac{W_0^2}{2}$ is the Rayleigh range. The Gouy phase $\Phi_G(z) = -(|\ell| + 2p + 1) \arctan \left(\frac{z}{z_r} \right)$ refers to a phase shift when passing through the focus of the beam, $L_p^{|\ell|}[x]$ are the associated Laguerre polynomials and $R(z) = z \left(1 + \frac{z_r^2}{z^2} \right)$ is the phase front radius. The OAM ℓ induces the azimuthal phase distribution. The modulus square of the amplitude is independent of the azimuthal angle φ , but depends on ρ , which leads to the formation of rings in the transverse intensity profile, see figure 1. The index p is associated with the number of radial nodes of the beam, or more explicitly, the number of rings in the transverse intensity profile is given by $p + 1$. Note that a $LG_{\ell,p}$ beam with $\ell = 0$ and $p = 0$ is just a Gaussian beam.

2.2. Lewenstein model for HHG

When an intense laser ($I \sim 10^{14} \text{ W cm}^{-2}$) irradiates an atomic gas target, it emits coherent radiation whose frequencies are

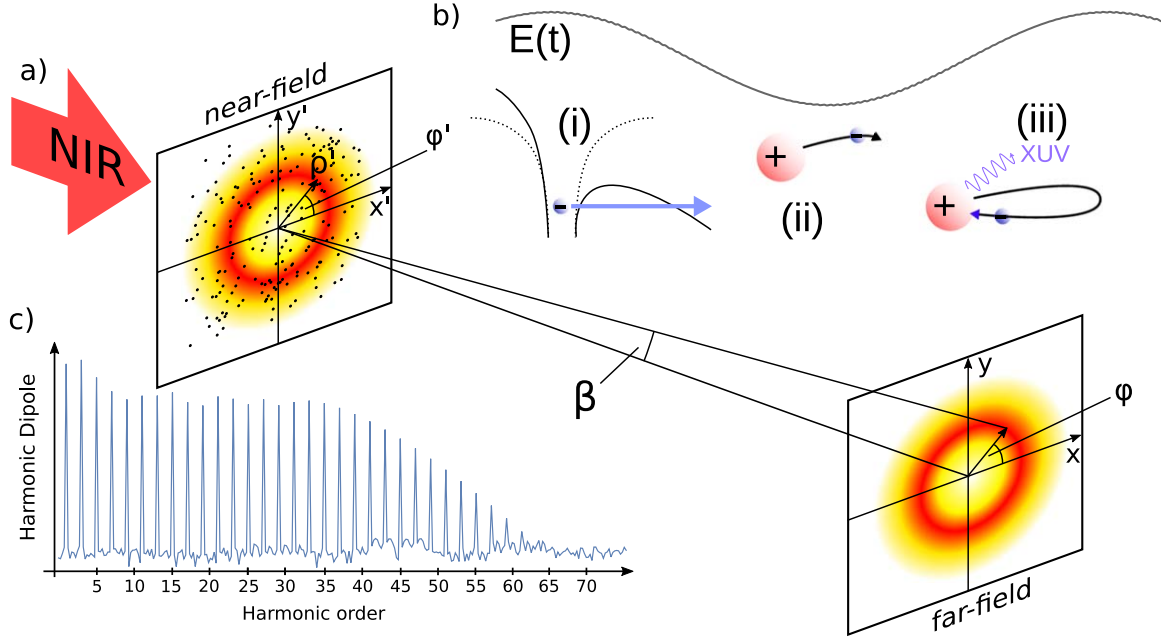


Figure 2. (a) Geometry of the Fraunhofer diffraction formula. The intense NIR laser interacts with the atomic gas target. High harmonics are generated in the near-field. The far-field arises as a superposition of all single atom responses to the NIR laser in the near-field. (b) Illustration of the three step model: (i) release of the electron due to tunnel ionization, (ii) propagation and acceleration of the free electron in the strong external laser field and finally (iii) recombination of the electron with the parent ion under the emission of a high energetic photon. (c) Typical spectrum of a high harmonic dipole in a linearly polarized laser field, where only the odd harmonic orders contribute to the spectrum.

multiples of the fundamental frequency of the laser, known as HHG. It can be theoretically described within the single electron approximation by means of the Lewenstein model [17] due to the interaction of an atom with a plane wave laser field. In this model, moreover, we assume that the harmonic field is emitted in terms of dipole radiation and that the electron is initially in the atomic ground state, denoted as $|0\rangle$ with ionization potential $I_p \gg \omega$. The time dependent Schrödinger equation

$$i\frac{\partial}{\partial t}|\Psi(\mathbf{r}, t)\rangle = \left[-\frac{1}{2}\nabla^2 + V(\mathbf{r}) - \mathbf{E}(t) \cdot \mathbf{r}\right]|\Psi(\mathbf{r}, t)\rangle \quad (7)$$

of an electron in an electric field $\mathbf{E}(t)$ can be solved analytically under the following assumptions: (i) the contributions of all atomic states, except of the ground and the continuum states are neglected in the time evolution, (ii) the electron is treated as a free particle in the continuum and does not feel any force from the potential of its parent ion $V(\mathbf{r})$ and (iii) the depletion of the ground state is neglected [17, 18]. With these assumptions, the wave function can be expanded as

$$|\Psi(\mathbf{r}, t)\rangle = e^{-i\mu_p t} \left[a(t)|0\rangle + \int d^3\mathbf{v} b(\mathbf{v}, t)|\mathbf{v}\rangle \right], \quad (8)$$

with the ground state amplitude $a(t)$ and the continuum state amplitudes $b(\mathbf{v}, t)$. The time dependent electric dipole moment can be written as

$$\begin{aligned} \mathbf{D}(t) = & i \int_{-\infty}^t dt' \int d^3\mathbf{p} \mathbf{d}^*(\mathbf{p} + \mathbf{A}(t)) \mathbf{E}(t') \cdot \mathbf{d}(\mathbf{p} + \mathbf{A}(t')) \\ & \times \exp \left[-i \int_{t'}^t dt'' \left(\frac{1}{2}(\mathbf{p} + \mathbf{A}(t'')) + I_p \right) \right] + \text{c.c.}, \end{aligned} \quad (9)$$

where $\mathbf{p} = \mathbf{v}(t) - \mathbf{A}(t)$ is the canonical momentum with the vector potential $\mathbf{A}(t) = -\int dt' \mathbf{E}(t')$. The dipole transition matrix element $\mathbf{d}(\mathbf{p}) = \langle \mathbf{p} | \mathbf{r} | 0 \rangle$ describes the transition from the ground state to the continuum (Volkov) state [19]. The expression for the time dependent dipole moment, equation (9), has a vivid physical interpretation in terms of the three step model [15], which is shown in figure 2(b). It describes the transition of the electron from a ground state to the continuum at time t' due to the laser field $\mathbf{E}(t')$. The phase that the electron waves accumulates from the ionization time t' to t , is the classical action of a free electron that propagates in the laser field. Finally, the dipole radiation for HHG originates from the recombination of the electron into its ground state at time t . The power spectrum of the emitted harmonic radiation is then given by the Fourier transform of the dipole acceleration $\frac{d^2\mathbf{D}(t)}{dt^2}$.

Figure 2(c) shows the typical spectrum of the emitted harmonics in a linearly polarized laser field. Each photon has a well defined helicity ± 1 . If an electron absorbs several photons, the helicities of the photons add up. Therefore, after the absorption of an even number of photons, it is not possible to emit the absorbed energy in terms of a single photon, because this single photon's helicity could not be ± 1 . This only can be achieved after the absorption of an odd number of photons. Therefore, only odd harmonics contribute to the spectrum, while all even harmonic orders are suppressed.

The Lewenstein model, in its original form, describes the interaction of a plane wave electric with the atom in the dipole approximation. The same approximation is still justified for twisted light beams, if, in addition, the azimuthal phase dependence due to the OAM ℓ is included. If an electron is

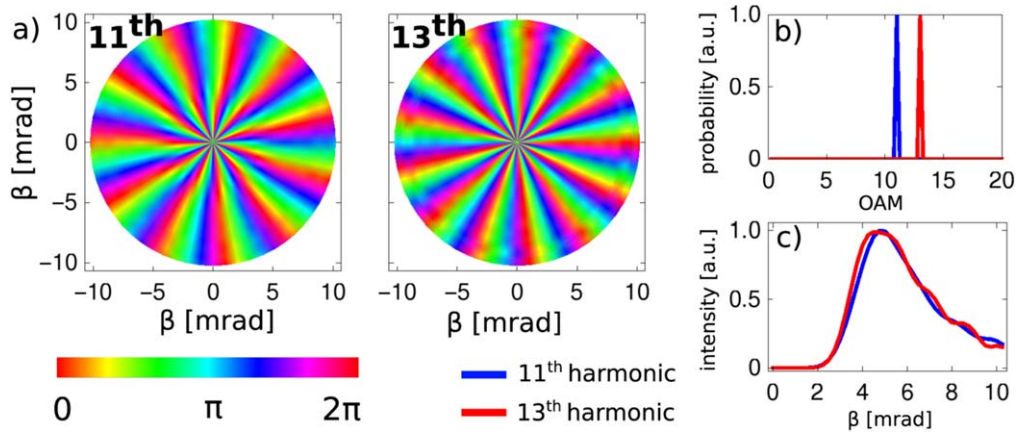


Figure 3. Results for HHG with a linearly polarized LG beam: (a) the phase profiles for the 11th and 13th harmonic at the detector. The OAM (number of phase shifts of 2π) equals the harmonic order. (b) OAM of the 11th (blue) and 13th (red) harmonic calculated by a Fourier transform along the azimuthal coordinate. (c) Intensity distributions of the 11th (blue) and 13th (red) harmonic in the far-field.

released from the atom, it typically travels only about 1–2 nm away from the atom, if it later recombines with the atom and therefore does not feel the spatial structure of the twisted light field. However, in order to describe HHG with twisted light beams it is required to sum up all the single atom responses from the interaction region of the beam at the detector. This is necessary to obtain the phase and intensity profiles of the emitted radiation and thus information about the OAM and divergence of the emitted radiation.

2.3. Fraunhofer diffraction

The Fraunhofer diffraction formula [20] can be applied to calculate the phase and intensity profiles of different harmonic orders in the far-field, respectively, at the detector. To make use of this formula, we model the atomic gas target as a thin layer, that can be displaced with respect to the focus plane of the beam. The complex amplitude of the q th harmonic in the far-field can be written as

$$A_q^{(\text{far})}(\beta, \varphi) = \int_0^\infty \int_0^{2\pi} \rho' d\rho' d\varphi' A_q^{(\text{near})}(\rho', \varphi', z') \times \exp\left(-i\frac{2\pi}{\lambda_q}\rho' \tan(\beta) \cos(\varphi - \varphi')\right), \quad (10)$$

where $\lambda_q = \lambda/q$ is the wavelength of the q th harmonic and $A_q^{(\text{near})}(\rho', \varphi', z')$ is the complex amplitude of the q th harmonic in the near-field, φ is the polar angle in the far field and β is the angle of divergence (see figure 2(a)).

3. HHG with linearly polarized LG beams

3.1. HHG with a single linearly polarized LG mode

3.1.1. Conservation of SAM and OAM. The development of the Ti:sapphire laser not only made Gaussian beams in the intensity regime of $\sim 10^{14}$ W cm $^{-2}$ accessible but also brought twisted light beams available to these intensities as well. From second harmonic generation it was known that the OAM of the second harmonic is twice the OAM of the fundamental

beam [21]. However, the first measurement of HHG with linearly polarized beam indicated, that the generated harmonics have the same OAM as the driving laser beam [22]. Zürich *et al* found a contradiction to their expectations from second-harmonic generation and to the Lewenstein model [17], which also predicts that the OAM of the q th harmonic equals $q \cdot \ell$, where ℓ is the OAM of the driving laser beam. They explained that the generated highly charged vortex may decay as it propagates to the detector. However, Garcia *et al* numerically simulated HHG in an atomic gas jet and propagated the generated radiation at each position in the gas jet to the detector [10]. Within their study, they proved that OAM of the harmonic vortices indeed scales with the harmonic order. Garipey *et al* supported these findings by measuring the OAM of several harmonics [9]. We can briefly summarize the rules for HHG with linearly polarized twisted light beams with frequency ω by

$$\text{LG}_{\ell,p}^{\omega} \longrightarrow \begin{cases} \omega_q = q\omega \\ \ell_{H_q} = q\ell, \end{cases} \quad (11)$$

where ω_q is the photon energy of the q th harmonic and ℓ_{H_q} and ℓ denote the OAM of the q th harmonic and the driving beam, respectively.

Figure 3(a) shows the phase of a 11th and 13th harmonic in the far-field. To obtain the OAM of a given harmonic order we can either count the phase shifts in multiples of 2π along the azimuthal coordinate or more exactly perform a Fourier transform along the azimuthal coordinate, which is shown in figure 3(b).

3.1.2. Divergence of the harmonic radiation. By making use of the Lewenstein model we can express the near-field amplitude of a q th harmonic as

$$A_q^{(\text{near})}(\rho', \varphi') = f(\rho') e^{i\ell_{H_q}\varphi'}, \quad (12)$$

where $f(\rho')$ includes the radial dependencies. If we insert equation (12) in (10), the integration over the azimuthal angle φ' can be carried out analytically [23] and the far-field

amplitude written as

$$A_q^{(\text{far})}(\beta, \varphi) = 2\pi i^{\ell_{Hq}} e^{i\ell_{Hq}\varphi} \int_0^\infty d\rho' \rho' f(\rho') J_{\ell_{Hq}}\left(\frac{2\pi}{\lambda_q} \beta \rho'\right), \quad (13)$$

where λ_q is the wavelength of the q th harmonic and $J_k(x)$ is a Bessel function of the first kind. The value β_{max} for which the far-field amplitude reaches its maximum can be seen as a measure of the divergence of each harmonic order, see figure 2(a). From equation (13) we can see, that the divergence of the harmonics increases with the OAM but decreases with the harmonic order. However, since the OAM scales with the harmonic order, all harmonics are emitted with similar divergence [10, 24]. Figure 3(c) shows the transverse intensity distributions for the 11th and 13th harmonic in the far-field depending on the angle of divergence. Since the intensity distributions of both harmonics reach their maximum at the same β , they are emitted with same divergence.

3.2. HHG with a superposition of two linearly polarized LG modes

In the previous subsection, we showed that the OAM of the generated harmonics increases with the harmonic order, if HHG is driven by a single linearly polarized LG mode. However, it is possible to further modify and control the OAM of the generated harmonics by using a superposition of two linearly polarized LG modes, so called mode mixing. Here, we present three possible setups of such mode mixing and explain how they affect the OAM of the generated harmonics.

1. Let us consider a superposition of two linearly polarized LG beams with the same frequency but different OAM. We use the same frequencies and OAM as in the original work by Rego *et al* [25] and write the superposition as

$$\text{LG}_{1,0}^{\omega \leftrightarrow} \oplus \text{LG}_{2,0}^{\omega \leftrightarrow}. \quad (14)$$

In order to generate a q th harmonic the electron has to absorb q photons. However, since both beams have the same photon energy, there are several photon pathways that contribute to the q th harmonic. The electron can absorb f.i. q photons from the first or q photons from the second beam. Moreover, it is also possible that the electron absorbs photons from both beams. If the electron absorbs n photons from the first beam, it has to absorb $q - n$ photons from the second beam, in order to generate a q th harmonic. If the photons are added perturbatively to the atoms, this gives rise to $q + 1$ possible different combinations of photons from both beams. We illustrate some of the possible pathways for the simple example $q = 5$ in figure 4(a). However, in contrast to the perturbatively possible pathways of the photons, it is also possible that the 5th harmonic carries OAM of $\ell_{H_5} = 4$, which shows that the generation of a 5th harmonic order can also be a photon process of 7th. Here, the electron may absorb an additional photon with $\ell = 1$ and emit a photon with $\ell = 2$, which shows that HHG is indeed a non-perturbative process. Similarly, if the electron absorbs six photons with $\ell = 2$ and

emits one photon with $\ell = 1$ the 5th harmonic carries an OAM of $\ell_{H_5} = 11$.

This argumentation can be easily extended to higher photon orders (9th, 11th). However, the probability for the HHG along pathways of higher photon orders decreases with the number of photons that are involved in the generation. A cutoff law has been derived in [25].

2. In the scenario above each generated harmonic carries multiple values of OAM which will become apparent as a superposition of several LG modes with different OAM at the detector. However, it is possible to spatially separate the generated harmonics at the detector according to their OAM with a non-collinear geometry of the two driving beams. Here, we consider the non-collinear superposition of a strong Gaussian beam with a weaker LG beam, where both beams have the same frequency

$$\text{LG}_{0,0}^{\omega \leftrightarrow} \oplus \text{LG}_{1,0}^{\omega \leftrightarrow}. \quad (15)$$

Non-collinear means that the beams are superimposed under a small angle of $\sim 1^\circ$ – 2° . In the original work, the intensity ratio of the two beams was approximately 100:1 [26]. Figure 4(b) shows some possible pathways for the example of a fifth harmonic. It is more likely for the electron to absorb photons from the Gaussian beam (zero OAM), since this beam is more intense. However, the electron can also absorb photons from the weaker LG beam, which will add OAM to the generated harmonic. Conservation of linear momentum gives rise to XUV radiation into direction of $\mathbf{k}_q = n_1 \mathbf{k}_1 + n_2 \mathbf{k}_2$, where $\mathbf{k}_{1,2}$ and $n_{1,2}$ are the wave vectors and the number of absorbed photons from the first and the second driving beam, respectively. Therefore, harmonics generated by different combinations of photons from the two driving beams will be spatially separated at the detector.

3. Finally, we discuss the non-collinear superposition of a strong Gaussian with a weaker LG mode that has two times the frequency of the Gaussian beam:

$$\text{LG}_{0,0}^{\omega \leftrightarrow} \oplus \text{LG}_{1,0}^{2\omega \leftrightarrow}. \quad (16)$$

Gauthier *et al* [27] used an intensity ratio of approximately 5:1 in their experiment. Figure 4(c) shows some possible photon pathways for the fifth and sixth harmonic. Here, for example, there are two possible pathways for a fifth harmonic. The left path shows the absorption of five photons with frequency ω and the right one the absorption of one photon with frequency ω and two photons with frequency 2ω . Both of these pathways lead to different OAM in the fifth harmonic, which will be spatially separated at the detector because of the non-collinear experimental setup. Note that absorption of three ω and one 2ω photons does not contribute to the fifth harmonic, since the total number of involved photons has to be an odd number. In contrast to the first two scenarios, this superposition of beams gives rise to a sixth harmonic as well, as shown in the right diagram in figure 4(c).

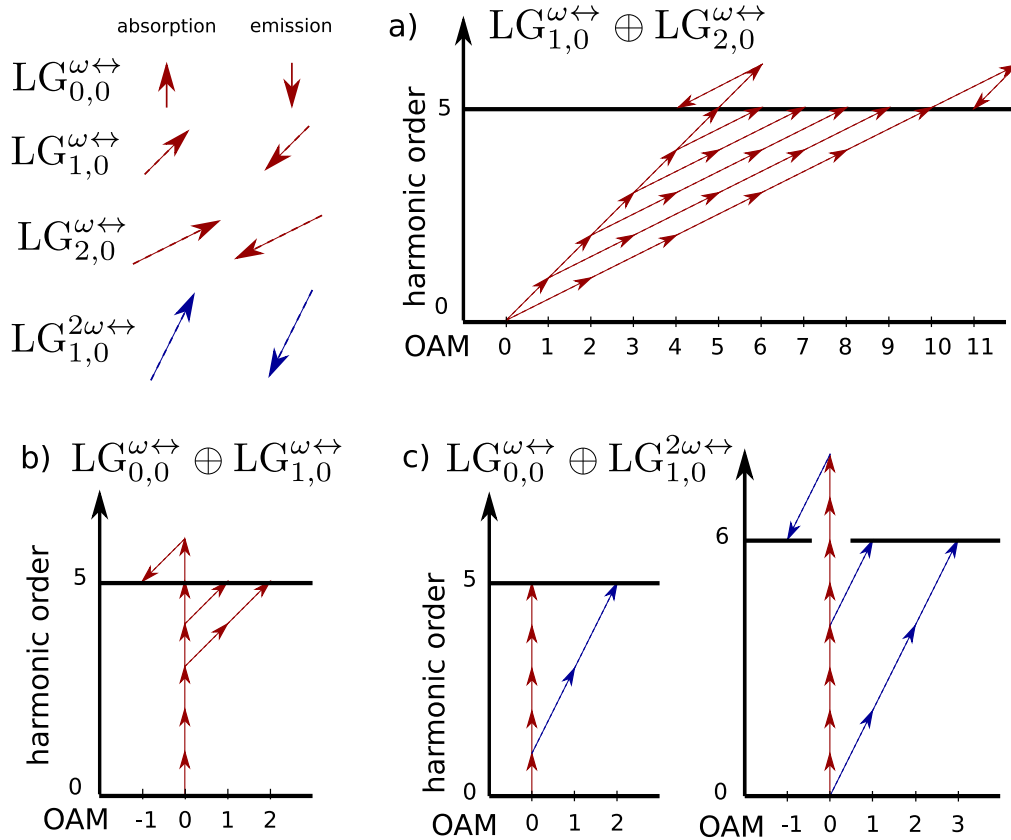


Figure 4. Photon diagrams for three different scenarios of HHG with superpositions of two linearly polarized (Laguerre-)Gaussian beams to control and manipulate the OAM of the generated harmonics. Different arrows as shown in the left-upper corner represent absorption and emission of photons from different beams, respectively. (a) Superposition of two linearly polarized LG beams with the same photon energy but different OAM as proposed in [25]. This scheme gives rise to multiple pathways for the generation of each harmonic order and therefore each harmonic carries several values of OAM. (b) Non-collinear superposition of a strong Gaussian beam with a weaker LG beam gives rise to the spatially separated XUV-vortices with a well defined OAM [26]. (c) Non-collinear superposition of a strong Gaussian with its second harmonic that carries a well defined OAM. Similar to (b), we get spatially separated XUV-vortices that carry a well defined OAM as well. In addition, within this superposition it is also possible to generate even harmonic orders [27].

4. HHG with circularly polarized LG beams

4.1. HHG with a single circularly polarized LG mode

All harmonics that are generated by linearly polarized driving beams are also linearly polarized as discussed in section 3. Similarly, one may expect to generate circularly polarized harmonics with circularly polarized driving beams. However HHG in circularly polarized beams is strongly suppressed, which can be explained with classical physics and with conservation of SAM.

From a semi-classical viewpoint, the HHG arises from the recombination of the released electron with the parent ion after the electron was accelerated by the driving laser field. In circularly polarized laser fields, however, the classical trajectories of the electrons will not come back to the parent ion, therefore HHG is strongly suppressed in circularly polarized fields. The suppression of HHG in circularly polarized laser fields can be understood in terms of the conservation of SAM. Since each photon has spin 1 and a projection of the spin onto the direction of propagation axis, called helicity, of ± 1 , which is related to the polarization of light. Thus, in a circularly polarized beam, all photons have the same helicity. If an

electron absorbs more than one photon from a circularly polarized beam, their helicities add up. However, the helicity of the emitted harmonic photons can only be ± 1 . Therefore it is not possible to emit a high harmonic after the absorption of several photons, where all photons have the same helicity.

4.2. HHG with a superposition of two counter rotating circularly polarized LG modes

4.2.1. Description of the electric field. While HHG is strongly suppressed in circularly polarized fields, circularly polarized harmonics can be generated by applying a superposition of a circularly polarized beam with its counter rotating second harmonic, which is also known as a bicircular beam. Figure 5 shows how the Lissajous figure of the electric field of a bicircular field is constructed from the superposition of two counter rotating fields.

4.2.2. Conservation of SAM and OAM. In HHG with linearly polarized beams, all even harmonics are suppressed and only the odd harmonic orders contribute to the emitted spectrum. For HHG with bicircular beams, in contrast, we find that every third harmonic is suppressed, while the contributing

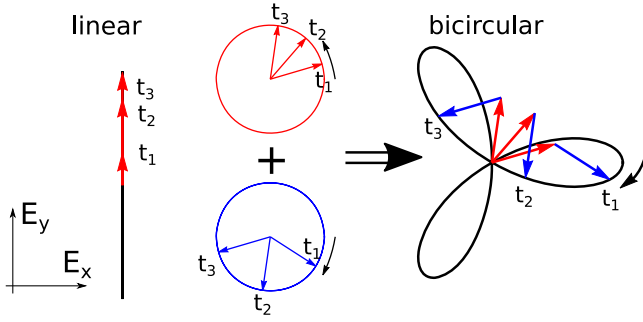


Figure 5. Parametric plots of the electric field vector. (Left) The electric field vector in a linearly polarized beam just oscillates in one dimension. The red arrows indicate the electric field vectors at three different times. (Middle) Circularly polarized fields (red) and its counter rotating second harmonic (blue). The arrows indicate the electric field vectors at the same times as for the linearly polarized field. (Right) Bicircular field as a superposition of a circularly polarized beam and its counter rotating second harmonic, which gives rise to the Lissajous figure for the electric field.

harmonics exhibit alternating helicities. This can be explained with the following selection rules [28–30]

$$LG_{0,0}^{\omega \odot} \oplus LG_{0,0}^{2\omega \ominus} \longrightarrow \begin{cases} \omega_q = q\omega = m\omega + n2\omega \\ m - n = \pm 1. \end{cases} \quad (17)$$

Here, the harmonic order is given by the total energy of all absorbed photons while the number of absorbed ω and 2ω photons must differ by one, because of the conservation of SAM. Note, there is only one possible pathway, i.e. one combination of ω and 2ω photons for each contributing harmonic, which gives rise to the polarization of the harmonics. In particular, if the electron absorbs one more photon from the ω beam than from the 2ω beam, the generated harmonic will exhibit the same polarization as the ω beam.

Obviously, the generated harmonics will carry zero OAM, if the OAM of the incident beams is zero. However, if we add OAM to the incident LG modes, we additionally have to consider of OAM of the generated harmonics. Since LG beams are paraxial beams, both, the SAM and OAM, will be conserved simultaneously and we can extend the selection rules from equation (17) to [11]:

$$LG_{\ell,0}^{\omega \odot} \oplus LG_{\ell,0}^{2\omega \ominus} \longrightarrow \begin{cases} \omega_q = q\omega = m\omega + n2\omega \\ m - n = \pm 1 \\ \ell_{H_q} = m\ell_1 + n\ell_2. \end{cases} \quad (18)$$

For HHG with bicircular beams, all generated harmonics carry a well defined OAM, since there is only a single pathway for each contributing harmonic. As seen from the selection rules in equation (18), the OAM of each harmonic can be directly accessed by the OAM of the incident beams. In particular, it is possible to tailor the OAM of the generated harmonics by the OAM of the incident beams such that every harmonic can carry an arbitrary OAM [11]. Figure 6(a) shows the photon diagrams for the generation of the simple cases of

a 5th and 7th harmonic order from a

$$LG_{1,0}^{\omega \odot} \oplus LG_{1,0}^{2\omega \ominus} \quad (19)$$

superposition. Here, for instance, the 5th harmonic exhibits the polarization of the 2ω beam and carries an OAM of $\ell_{H_5} = 3$ due to its generation out of three photons, each with an OAM of $\ell = 1$.

Figure 7(a) shows the phase distributions in the far field for the 11th and 13th harmonic that were generated by the superposition of LG beams from equation (19). The OAM of the generated harmonics can be obtained, if we count the number of phase shifts of 2π along the azimuth or by azimuthal Fourier transform, which is shown in figure 7(b).

Moreover, it is possible that all generated harmonics carry the same modulus of OAM, if both incident beams carry the same of OAM but with opposite signs, as shown in figure 6(b) for a

$$LG_{1,0}^{\omega \odot} \oplus LG_{-1,0}^{2\omega \ominus} \quad (20)$$

superposition.

4.2.3. Divergence of the harmonic radiation. Similar to HHG with linearly polarized beams, we can apply the Fraunhofer diffraction formula in order to calculate the intensity in the far-field. This means that if the OAM increases with the harmonic order, the harmonics will be emitted with similar divergence. Figure 7(c) shows, for example, that the divergence of an 11th harmonic with OAM $\ell_{H_{11}} = 7$ and a 13th harmonic with OAM $\ell_{H_{13}} = 9$ are similar to each other. However, since it is possible to tailor the OAM of the harmonics precisely by the OAM of the driving beams and since the divergence of the harmonics is affected by the OAM, the divergence of the harmonics can be tailored as well.

Figure 8 shows the intensity distributions for several harmonics from the plateau (10th–17th). Here, we compare three different superpositions of LG beams: equations (19) and (20) and a

$$LG_{2,0}^{\omega \odot} \oplus LG_{-1,0}^{2\omega \ominus}, \quad (21)$$

superposition, which was used by Dorney *et al* [31].

Figure 8(a) displays, similar to figure 7(c), that the harmonics are emitted at similar divergence. In figure 8(b), the divergence decreases for each harmonic, because all harmonics will be emitted with the same modulus of OAM. Figure 8(c) shows that left (red) and right (blue) circularly polarized harmonics have different divergence, which leads to a spatial separation of left and right circularly polarized harmonics at the detector. As a result, we obtain right circularly polarized twisted attosecond pulse trains (APT) for low angles of divergence ($\beta \approx 1$ mrad) and left circularly polarized twisted APT for $\beta \approx 2$ mrad.

4.2.4. Relative intensity ratio and polarization control. Until the present, the relative intensity ratio between the ω and 2ω beams in bicircular beams has been utilized as an easily accessible parameter in experiments in order to control various strong-field with plane-wave beams processes such as

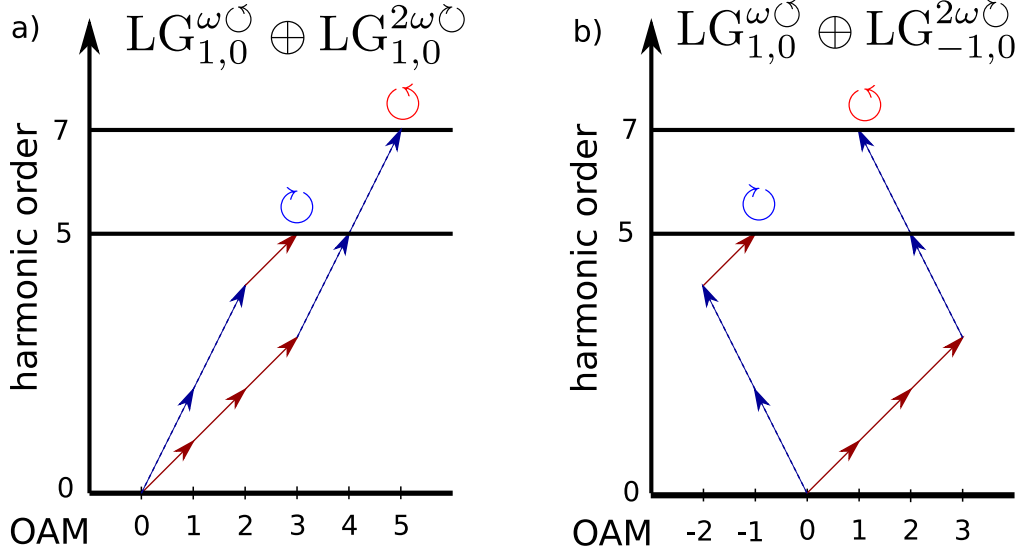


Figure 6. Photon diagrams of HHG with with bicircular twisted beams. (a) Superposition of two LG beams with the same OAM. For each harmonic order, there is only one possible pathway, which gives rise to XUV vortices with well defined OAM. Each harmonic is circularly polarized. (b) Superpositions of two LG beams with opposite OAM. Since the number of photons absorbed from the ω -beam always differs from the number of photons absorbed from the 2ω -beam by one, the modulus of the OAM of each XUV vortex equals one.

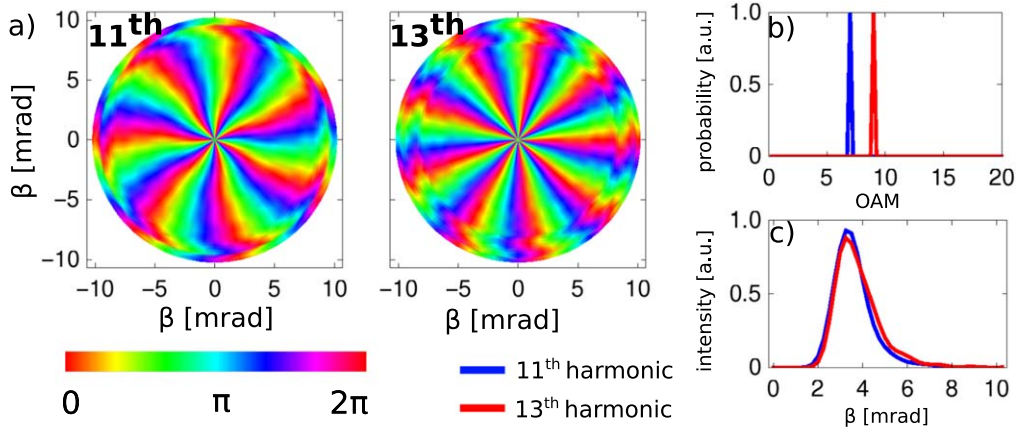


Figure 7. Results for HHG with a bicircular LG beam from equation (19). (a) The phase profiles for the 11th and 13th harmonic at the detector. The 11th harmonic carries OAM of $\ell_{H11} = 5$ and the 13th harmonic of $\ell_{H13} = 9$. (b) OAM of the 11th (blue) and 13th (red) harmonic calculated by a Fourier transform along the azimuthal coordinate. (c) Intensity distributions of the 11th (blue) and 13th (red) harmonic in the far-field.

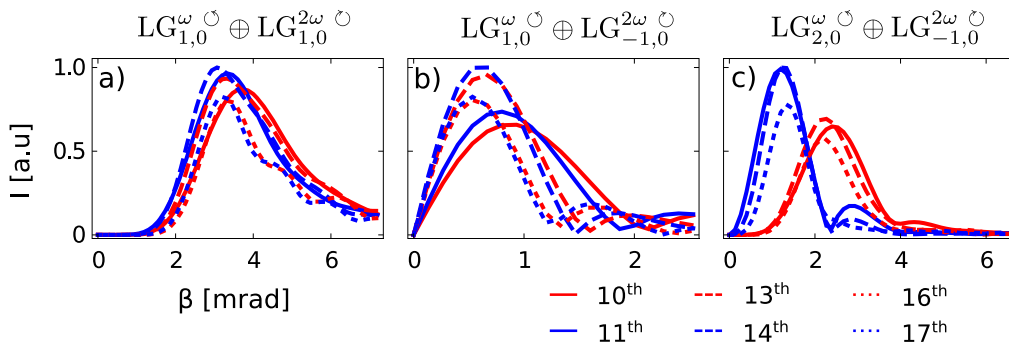


Figure 8. Intensity distributions of generated harmonics for three different incident bicircular LG beams. Different colors indicate different polarization of the emitted harmonics. The intensity distributions show that the divergence of the harmonics can be controlled by the OAM of the driving beam. (a) All harmonics, left (red) and right (blue) circularly polarized harmonics, are emitted with similar divergence. (b) The divergence decreases as the harmonic order increases, since all harmonics have the same modulus of OAM. (c) Left and right circularly polarized harmonics can be spatially separated at the detector, by proper choosing of the OAM of the incident beams [31].

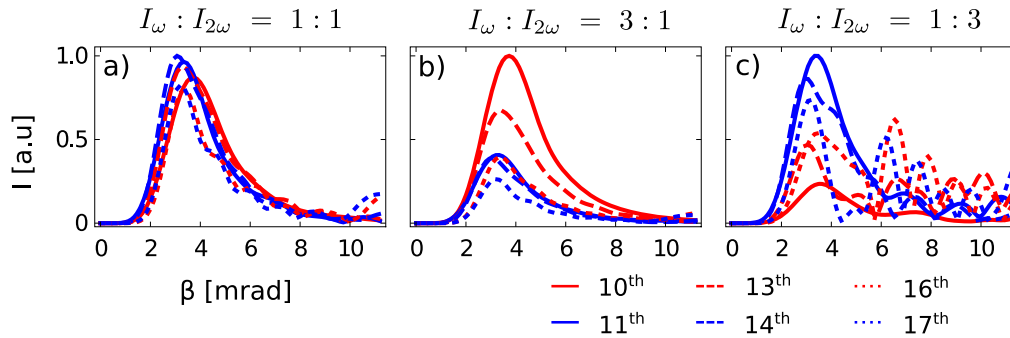


Figure 9. Intensity distributions of generated harmonics for three different intensity ratios of the driving beams. Different colors indicate different polarization of the emitted harmonics. For all calculations, we used a $\text{LG}_{1,0}^{\omega} \oplus \text{LG}_{1,0}^{2\omega}$ superposition. (a) Same plot as figure 8(a). Intensity distribution for (b) a dominating ω -beam and (c) a dominating 2ω -beam.

above threshold ionization [32], non-sequential double ionization [33, 34] and also HHG [30, 35].

For HHG, in particular, it was shown that it is possible to enhance the generation either left or right circularly polarized harmonics by adjusting the relative intensity ratio of the ω and 2ω beam. In this section, we show that the same scheme can be applied to bicircular LG beams. Figure 9 displays the intensity distributions in the far field for three different intensity ratios of the incident ω and 2ω beams in a $\text{LG}_{1,0}^{\omega} \oplus \text{LG}_{1,0}^{2\omega}$ superposition. Figure 9(a) shows the same distributions as figure 8(a). If both incident beams have the same intensity, left and right circularly polarized harmonics are emitted with similar intensities. Despite the circular polarization of each harmonic, however, the generated APT are still linearly polarized, since neighbored harmonics have opposite helicities. If all harmonics of a given helicity could be entirely suppressed, the generated attosecond pulses trains will exhibit a pure circular polarization. In figure 9(b) the incident ω beam is dominant and, thus, harmonics with the same polarization as the ω beam are emitted preferably. In contrast, figure 9(c) shows the intensity distributions when the 2ω beam dominates. Interestingly, the latter case not only leads to the preferred emission of harmonics with the same polarization as the 2ω beam, but also to the formation of several maxima in the far-field intensity distribution. Here, the harmonics are not pure $\text{LG}_{\ell,0}$ modes, but superpositions of modes with different radial quantum numbers; see the right column of figure (1). The appearance of higher order radial modes has already been observed in HHG with linearly polarized LG beams caused by a displacement of the atomic target with respect to the focus plane [36]. A detailed analysis of the radial intensity profile in HHG with bicircular LG beams will be made in forthcoming works.

5. Outlook and conclusions

In this article, we summarized various advancements in the HHG with LG beams. In particular, we discussed the HHG driven by linearly polarized, superpositions of linearly polarized and finally bicircular polarized LG beams. Moreover, we presented these results in terms of intuitive photon diagrams, and discussed possibilities to modify and control

the OAM of the generated harmonics. We also showed that the generated harmonics can also be spatially separated at the detector according to their OAM by non-collinear HHG with linearly polarized LG beams. Whereas, for HHG with bicircular LG beams, we showed that the OAM of the harmonics can be precisely tailored by the OAM of the driving beams, which also makes it possible to spatially separate left and right circularly polarized harmonics at the detector.

Until the present, most HHG studies just applied dipole amplitudes that were generated from parameterized, and often hydrogenic single-electron wave functions. It will be interesting to explore how the spectra of the emitted harmonics are affected if realistic orbital functions from many-electron and easily accessible structure codes are applied [37]. Another open question is the HHG with ultra short bicircular driving pulses, to generate isolated attosecond pulses. Here, the spectrum of emitted harmonics will not exhibit the discrete $3n \pm 1$ contributions, but it will smear out. In HHG with bicircular beams, this gives rise to the emission of the forbidden $3n$ harmonics and leads to depolarization of the harmonics [38].

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