Supplementary Materials for

Necklace-structured high-harmonic generation for low-divergence, soft x-ray harmonic combs with tunable line spacing

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Supplementary Text
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In this Supplemental material we include information for: (i) Role of the amplitude ratio and dipole phase on the brightness of the on-axis emission; (ii) Phase delay between the lobes of the driving necklace; (iii) Theoretical demonstration of the divergence scaling of the on-axis emission; (iv) Experimental characterization of the driving beam.

**Role of the amplitude ratio and dipole phase of the drivers to enhance the brightness of the on-axis emission**

The relative amplitude or intensity between the two driving fields is a key feature to enhance the brightness of the on-axis high-order harmonic generation (HHG) driven by the opposite, non-degenerate orbital angular momentum (OAM) driving combination. As reported in Ref (40), when driving HHG with two vortex beams, with different topological charges \( \ell_1, \ell_2 \) and overlapped in time and space, each harmonic presents several OAM contributions. The perturbative photon composition law implies that the weight of each OAM contribution to the \( q \)-th order harmonic is given by the binomial probability distribution associated with the photon number combinations of composing \( q - n \) photons from mode \( \ell_1 \) and \( n \) photons from mode \( \ell_2 \). Thus, the amplitude of the polarization of the medium corresponding to the different composition channels results from the product of the amplitudes of each driver, \( U_i \), to the power of the number of photons of each mode (see Supplemental Material of Ref. (21)). As a consequence, the amplitude ratio between the two driving pulses can be used to shift the center the OAM distribution between \( q \ell_1 \), and \( q \ell_2 \). Thus, in order to enhance the on-axis emission \((\ell_q=0)\), the amplitude ratio should satisfy \( U_1=(|\ell_2|/|\ell_1|)U_2 \). For example, in the case of \( \ell_1=1 \) and \( \ell_2=-2 \), for an amplitude ratio of \( U_1=2U_2 \), (intensity ratio of
$I_1 = 4I_2$) the OAM distribution of all harmonics will be centered at $\ell_q = 0$, and thus the on-axis emission would be the brightest.

However, non-perturbative HHG contains also an OAM signature connected to the intensity dependent harmonic dipole phase, whenever the driving field presents an intensity gradient along the azimuthal coordinate (40). As a result, the harmonic OAM distribution depends not only on the amplitude ratio, but also on the intensity gradient through the dipole phase. Therefore, the amplitude ratio that yields the brightest on-axis emission depends on the harmonic order.

**Figure S1 | Role of the amplitude ratio and dipole phase on the brightness of the on-axis emission.** Simulated on-axis HHG yield driven in He when considering different driving OAM combinations: a) $\ell_1 = 1$, $\ell_2 = -2$, b) $\ell_1 = 2$, $\ell_2 = -3$, c) $\ell_1 = 3$, $\ell_2 = -4$ and d) $\ell_1 = 4$, $\ell_2 = -5$. The amplitudes of the driving pulses are chosen to obtain a maximum peak intensity at focus at the radii of maximum $6.9 \times 10^{14}$ W/cm$^2$, with amplitude ratios of $U_1/U_2 = 0.75 |\ell_2|/|\ell_1|$ (red circles), $U_1/U_2 = |\ell_2|/|\ell_1|$ (pink squares), and $U_1/U_2 = 1.25 |\ell_2|/|\ell_1|$ (blue diamonds). The legend indicating the different amplitude ratios is placed at the top-right of the figure. Note that the harmonic yield is normalized for each OAM combination, but it is not normalized between different amplitude ratios.
In Figure S1 we present the simulated on-axis HHG yield obtained in He for driving OAM combinations of a) \( \ell_1=1, \ell_2=-2 \), b) \( \ell_1=2, \ell_2=-3 \), c) \( \ell_1=3, \ell_2=-4 \) and d) \( \ell_1=4, \ell_2=-5 \). The amplitudes of the driving pulses are chosen to obtain a maximum peak intensity at focus at the radii of maximum \( 6.9 \times 10^{14} \text{ W/cm}^2 \). Three different amplitude ratios have been chosen: \( U_1/U_2=0.75|\ell_2|/|\ell_1| \) (red circles), \( U_1/U_2=|\ell_1|/|\ell_1| \) (pink squares), and \( U_1/U_2=1.25|\ell_2|/|\ell_1| \) (blue diamonds). If the generation process were perturbative, the \( U_1/U_2=|\ell_2|/|\ell_1| \) (red dots) case would maximize the on-axis signal for all harmonic orders. However, as it can be observed in all panels from a) to d), the amplitude ratio that optimizes the on-axis harmonic yield varies, especially for the high-order harmonics.

In the numerical simulations presented in the main text, we chose the intensity ratios \( I_1/I_2=1.5|\ell_2|/|\ell_1| \) for all cases presented in Ar at 800 nm. For HHG driven in He at 800 nm, the intensity ratios are \( I_1/I_2=4 \) for the OAM combination \( \ell_1=1, \ell_2=-2 \); \( I_1/I_2=2.25 \) for \( \ell_1=2, \ell_2=-3 \); \( I_1/I_2=2 \) for \( \ell_1=3, \ell_2=-4 \); and \( I_1/I_2=1.25 \) for \( \ell_1=4, \ell_2=-5 \). Finally, for HHG driven in He at 2 \( \mu \)m, the intensity ratios selected are \( I_1/I_2=4 \) for the OAM combination \( \ell_1=1, \ell_2=-2 \); \( I_1/I_2=2 \) for \( \ell_1=2, \ell_2=-3 \); \( I_1/I_2=2 \) for \( \ell_1=3, \ell_2=-4 \); and \( I_1/I_2=1.88 \) for \( \ell_1=4, \ell_2=-5 \). Note that have used the simple relationship between the intensity and amplitude ratios \( I_1/I_2=(U_1/U_2)^2 \).

**Temporal cadence and relative phase of the harmonic emission from the necklace lobes.**

The necklace-driven high-order harmonics detected on-axis are emitted as a sequence of \( \Delta q=\Delta \omega/\omega_0 \) bursts per cycle of the driving pulse (see Eq. (3) in the main text). This is a result expected from the Fourier transform of the generated harmonic comb, which is a direct consequence of the coherent macroscopic emission of the necklace-driven harmonics. The different lobes in the necklace driving field possess different phases, which implies that the harmonics are emitted at different instants in each lobe. Since the on-axis emission is detected at the same distance from all lobes, the minimum phase shift between the lobes determines the time delay between consecutive on-axis HHG bursts.

Let us consider the necklace driving field composed by two OAM components \( \ell_1 = |\ell_1| \) and \( \ell_2 = -|\ell_2| \), with \( |\ell_1| < |\ell_2| \). To understand the azimuthal field structure, let us first consider that the two OAM beams overlap at their radius of maximum intensity so the field at that radius can be written as,

\[
U(\theta) = U_1 e^{i|\ell_1|\theta} + U_2 e^{-i|\ell_2|\theta}, \tag{S1}
\]

where \( \theta \) is the azimuthal coordinate and \( U_1 \) represents the amplitude of each OAM component. From \( |U(\theta)|^2 \) we find that there are \( N = |\ell_1| + |\ell_2| \) lobes in the necklace, located at \( \theta_n = n \ 2\pi/N \), \( n \) ranging from 0 to \( N-1 \). By writing the field at the radius of maximum intensity as \( U(\theta) = U_1 e^{i|\ell_1|\theta} \left( 1 + (U_2/U_1)e^{-iN\theta} \right) \), we can calculate the phase of the driving field at the position of maximum intensity of each lobe:
\[ \phi(\theta_n) = n \frac{2\pi |\ell_1|}{|\ell_1| + |\ell_2|}. \quad n = 0, 1, \ldots, N - 1 \]  

Note that this phase is equivalent to \( \phi(\theta_n) = n \frac{2\pi}{|\ell_1| + |\ell_2|} \).

By inspecting the above expression one can find particular OAM combinations in which there are lobes that exhibit the same phase, and, thus, their emission is constructively superimposed in time in the on-axis signal. Therefore, to find the number of harmonic bursts emitted, it is necessary to determine the number of distinguishable lobes of the driving field. It must be a fraction of the number of lobes, \( N_d = \frac{(|\ell_2| + |\ell_1|)}{K} \), \( K \) being a natural number that indicates the number of repetitions of each phase value or, in other words, the number of lobes that are indistinguishable for each OAM combination. \( K \) can be calculated as the number of azimuthal angles in which the \( \ell_1 \)-component is in phase with the \( \ell_2 \)-component, which is the greatest common divisor (g.c.d.) between \( |\ell_1| \) and \( |\ell_2| \): \( K = g.c.d(|\ell_1|, |\ell_2|) \). From the mathematical relation \( g.c.d(a, b) = \frac{a-b}{\text{l.c.m}(a, b)} \), being l.c.m. the least common multiple, we can express \( K = \frac{|\ell_1||\ell_2|}{\text{l.c.m}(|\ell_1|, |\ell_2|)} \) for convenience.

Following the same notation as in the main text—where we introduced \( \xi_1 = \text{lcm}/|\ell_1| \) and \( \xi_2 = \text{lcm}/|\ell_2| \), \( \text{lcm} \) being the least common multiple of \( |\ell_1| \) and \( |\ell_2| \)—, the number of distinguishable lobes is \( N_d = \frac{(|\ell_2| + |\ell_1|)}{|\ell_2||\ell_1|} \text{lcm} = \xi_1 + \xi_2 \).

Let us now calculate the minimum phase shift between two of these distinguishable lobes, \( \Delta \phi_{\min} \). First, it is necessary to recall, that the HHG emission occurs twice per cycle as a consequence of the symmetry of the driving field and atomic target, which means that the fundamental phase shift between the emission of two harmonic bursts in each lobe is \( \omega_0 T_0 / 2 = \pi \), where \( \omega_0 \) is the driving field frequency and \( T_0 \) its corresponding oscillation period. As a consequence of this periodicity, we are interested in the modulo \( \pi \) of the lobe phases. Second, note that the variation of the driving field phase at the peak intensity of the lobes is constant along the azimuthal angle (see Eq. [S2]). Therefore, since the number of distinguishable lobes is \( N_d \) and their phases must range linearly from 0 to \( \pi \), the minimum phase shift between different lobes is

\[ \Delta \phi_{\min} = \frac{\pi}{N_d} = \frac{\pi}{\xi_1 + \xi_2}. \]

Finally, from the minimum phase shift—which may occur between non-consecutive lobes—, we can extract the time delay between successive emissions, \( \Delta \tau \):

\[ \Delta \phi_{\min} = \omega_0 \Delta \tau \rightarrow \Delta \tau = \frac{\Delta \phi_{\min}}{\omega_0} = \frac{T}{2(\xi_1 + \xi_2)}. \]

Therefore, the number of bursts per cycle are \( 2(\xi_1 + \xi_2) \), which is equal to the line spacing of the harmonic comb \( \Delta \omega/\omega_0 \) (Eq. (3) in the main text).
Divergence of the on-axis HHG emission driven by two opposite non-degenerate OAM driving beams

To estimate the divergence of the harmonic beams emitted on axis we calculate their far-field distribution. Assuming that the qth-harmonic presents on-axis signal, we consider that its spatial distribution at the gas target (the harmonic near-field distribution) can be approximated as a thin ring (which would correspond to the ring of maximum intensity) with an azimuthal dependence related to the OAM content of that harmonic:

$$A_q(\rho', \phi', z = 0) = A_0 \delta(\rho' - R) \sum_{\ell=-\infty}^{\infty} c_{\ell} e^{i \ell \phi'}, \quad [S4]$$

where $R$ is the radius of the ring and $c_{\ell} \neq 0$ only for the OAM values that contribute to the harmonic emission. If we introduce this near-field distribution in the Fraunhofer integral formula, we end up with the following expression:

$$U_q(\rho, \phi, z) \propto A_0 R \int_0^{2\pi} \left( \sum_{\ell=-\infty}^{\infty} c_{\ell} e^{i \ell \phi'} \right) e^{-i \frac{2\pi}{\lambda_q} R \cos(\phi - \phi')} d\phi' \quad [S5]$$

Making use of the Jacobi-Anger identity (56), we expand the exponential of the trigonometric function in basis of their harmonics obtaining two different contributions to the far-field spatial distribution:

$$U_q(\rho, \phi, z) = U_q^{(0)}(\rho, \phi, z) + U_q^{(1)}(\rho, \phi, z),$$

where:

$$U_q^{(0)}(\rho, \phi, z) \propto A_0 R \int_0^{2\pi} \left( \sum_{\ell=-\infty}^{\infty} c_{\ell} e^{i \ell \phi'} \right) e^{-i \frac{2\pi}{\lambda_q} R \cos(\phi - \phi')} d\phi' \quad [S6]$$

$$U_q^{(1)}(\rho, \phi, z) \propto A_0 2R \sum_{n=1}^{\infty} (-i)^n \int_0^{2\pi} \left( \sum_{\ell=-\infty}^{\infty} c_{\ell} e^{i \ell \phi'} \right) \cos(n(\phi - \phi')) d\phi' \quad [S7]$$

The first contribution, $U_q^{(0)}(\rho, \phi, z)$, is the only one that exhibits signal on-axis and, as shown in Eq. [S6], and it is related to the zero OAM contribution of the q-th order harmonic, $c_{\ell=0}$. This result is expected, as the q-th order harmonic exhibits on-axis signal only if it possesses $\ell=0$ content. We can now calculate its divergence distribution as:

$$U_q^{(0)}(\beta) \sim \int_0^{2\pi} \frac{2\pi R}{\lambda_q} \beta, \quad [S8]$$

where $\beta \sim \rho/z$. Moreover, we can calculate the divergence width of the qth-harmonic. The Full Width at Half Maximum (FWHM) divergence angle of the intensity distribution is:

$$\Delta \beta_q^{FWHM} = 2.25 \frac{\lambda_q}{2\pi R} = 2.25 \frac{\lambda_0}{2\pi q R} \quad [S9]$$

This estimation agrees very well with the results obtained in the experiments and simulations presented in the main text, demonstrating that the divergence angle of the harmonics emitted on-axis decreases with the harmonic order. Moreover, the estimation shows that the larger the harmonic ring is, the sharper the comb of harmonics emitted on-axis becomes. To test the dependence of the divergence of the harmonics emitted on axis vs. the size of the ring, we have
performed simulations changing the radius of the necklace ring of the driving beam and, therefore, the size of the ring of the EUV/SXR phased antenna array. Figure S2 shows the FWHM divergence of the harmonics emitted on axis for the case of a driving beam composed of $\ell_1=2$ and $\ell_2=-3$ vortex beams, considering different ring radii, $R$, (28.3 $\mu$m, 21.2 $\mu$m and 14.1 $\mu$m). We have also added in the figure the estimation indicated by Eq. [S9] showing a very good agreement with those obtained in the full-quantum simulations, which demonstrates the decrease of the divergence when increasing the size of the vortices used as driving beam.

![Figure S2 | Divergence of the OAM-driven high harmonic combs for different driving beam waists.](image)

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**Experimental characterization of the necklace driving beam composed of two opposite non-degenerate OAM fields**

The necklace beams described in this work can be generated by the interference of two vortex beams each carrying pure, opposite and non-degenerate orbital angular momenta. Experimentally, we form a three-lobe necklace driver for HHG by interferometrically combining two ultrafast pulses with OAM $\ell_1 = 1$ and $\ell_2 = -2$. In order to stably produce the predicted low divergence, on-axis high harmonic comb, we take care to ensure high spatiotemporal quality of the component
pulses, as well as excellent relative temporal and spatial stability.

Using a partial reflection from a wedge inserted at the output of the interferometer, we observe the combined driver using a visible CCD (Mightex BTE-B050-U) mounted on a translation stage. The intensity of the combined field can thus be observed at various $z$-positions, shown in Fig. S3a ($z = 0$mm) and Fig. S3b ($z = 9$mm), showing that the necklace structure persists in and out of the focal plane. The component pulses are synchronized in time by maximizing the interference contrast in these images, and confirmed to have identical pulse durations ($\tau_{FWHM} = 57$ fs) by individual FROG measurements (Fig. S3c) – here, the relative amplitudes are plotted to reflect the intensity ratio of the component pulses, which is set to optimize the on-axis HHG emission.

A series of seven images are acquired at various $z$-positions on either side of and including the focal plane. These images form an overconstrained data set corresponding to a unique solution for the complex electric field (phase and amplitude) of the beam. This solution is obtained by feeding the images into a Gerchberg-Saxton phase-retrieval algorithm (57-59). Beginning with a random guess for the spatial phase profile, the algorithm uses numerical Fresnel propagation in order to enforce consistency with the measured images at each $z$-position. By iteratively cycling through the acquired image stack, the algorithm converges to the correct phase within 20 iterations. For the present combination ($\ell = 1$, $\ell = -2$), the reconstruction shows that each of the lobes in the necklace has approximately constant phase in the focal plane, with the expected relative phase shift between neighbors of $2\pi/3$ (Fig. S3d).

We integrate the amplitude profile of the beam at focus about the azimuth in order to measure the radius of maximum intensity $R \approx 32$ μm (Fig. S3e), which can be used to calculate the expected divergence of the on-axis HHG comb. We then compare this to the azimuthal Fourier transform of the retrieved complex electric field, which gives the OAM distribution as a function of radius. We see that at the necklace radius $R$, the two OAM channels have an amplitude ratio of $|E_1|/|E_2| \approx 1.72$, consistent with the theory prediction for optimized on-axis emission.

We note that in addition to confirming the expected phase structure and OAM content of the necklace beam, the success of the phase reconstruction also indicates the high degree of stability of our interferometer. The phase-retrieval algorithm used here is suited for measuring the phase of a quasi-monochromatic, spatially and temporally coherent beam. In order to apply it for the superposition of multiple pulses, a high degree of mutual coherence is necessary. Beyond the necessary conditions of identical wavelength and polarization, significant spatial or temporal jitter between the component pulses causes the acquired images to morph and/or smear. The images would then be inconsistent with a naturally diffracting field with the expected properties, and cause the reconstruction to fail. Hence, by applying the GS technique to the total (combined) electric field, we confirm the quality of both the necklace beam and the interferometer setup used to form it.
Figure S3 | Experimental Characterization of Dual-Vortex Necklace Driving Beam. Images are acquired of the combined 800nm (ℓ = 1, ℓ = −2) driver at several points in (a) and out of (b) the laser focus using a visible CCD. The amplitude of the ℓ = 1 pulse is increased relative to that of the ℓ = −2 pulse in order to optimize the on-axis emission in the generated harmonics. The pulses are set to be temporally overlapped by maximizing the interference contrast in the CCD images, and confirmed to have identical pulse durations and structure via individual FROG measurements (c). The CCD images of the driver are used in a Gerchberg-Saxton phase-retrieval algorithm to measure the complex electric field of the combined beam (d), where brightness and color correspond to amplitude and phase, respectively. Angular integration of the necklace-structured electric field amplitude (e) gives the radius of maximum intensity $R \approx 32 \, \mu m$, while an azimuthal Fourier transform of the complex electric field (f) gives the OAM content, including the relative strength and approximately matched radial positions.
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