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Low-Divergence, Soft X-Ray Harmonic Combs with Tunable Line Spacing from Necklace-Structured Driving Lasers

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High-harmonic generation (HHG) is among the most extreme nonlinear optical processes to date. In HHG, some of the driving laser beam properties are imprinted on the dynamics of the radiating electron, and, in turn, on the emitted extreme-ultraviolet/soft X-ray light. As a consequence, it is now possible to coherently manipulate the HHG pulse duration (attosecond waveforms), the photon energy (UV to soft X-rays [1]), polarization and orbital angular momentum (OAM) [2,3], among others. However, precise coherent control over the frequency content—a key property to perform high-harmonic spectroscopy—has not been achieved yet. Also, equally important to the frequency content is the transverse confinement of the HHG radiation [4,5] is a key aspect for applications in imaging and scatterometry.

Here we demonstrate that by harnessing OAM conservation in the HHG process, we can generate a transverse necklace-shaped spatial phased array of harmonic emitters that allow us to tune the line spacing and divergence of the emitted harmonic combs [6]. Our theoretical and experimental results show that the on-axis HHG emission (1) is composed of harmonics whose frequency separation can be controlled through the OAM of the driving field, extending towards the soft X-rays; and (2) presents extremely low divergence, well below that obtained when using Gaussian driving beams, which further decreases with the harmonic order (Fig. 1). This work provides a new degree of freedom for the design of harmonic combs—particularly in the soft X-ray regime, where very limited options are available. This control will enable the use of structured X-ray light for probing and imaging the fastest correlated charge and spin dynamics in molecules, nanoparticles and materials.

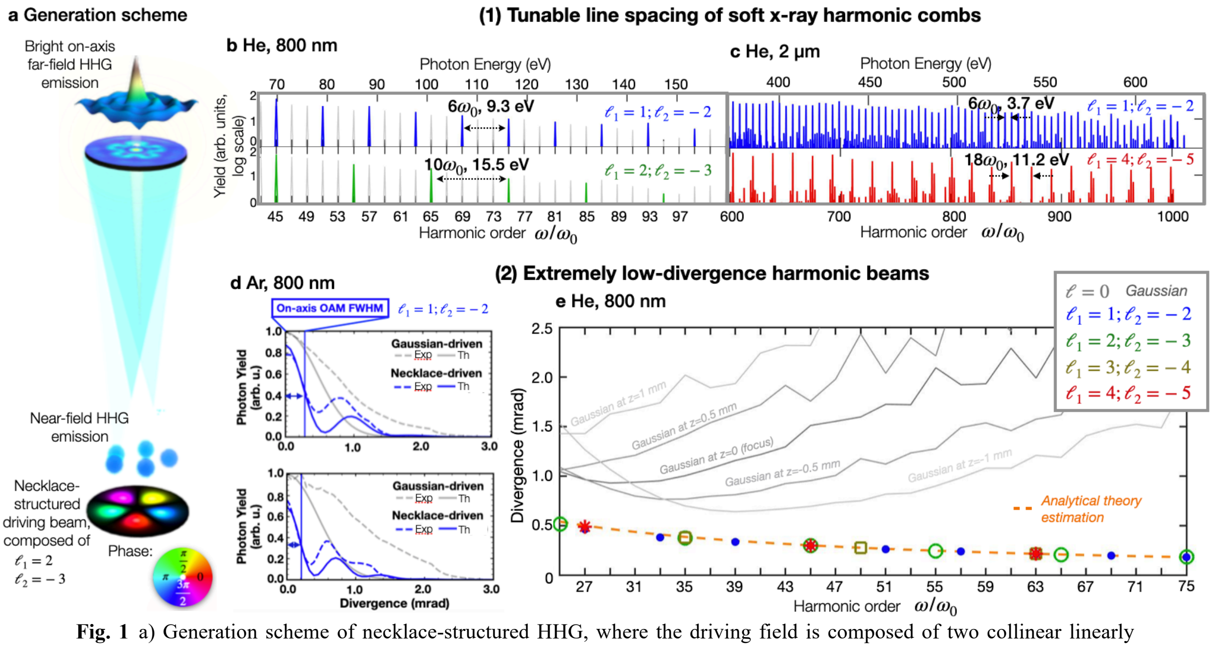

Fig. 1 a) Generation scheme of necklace-structured HHG, where the driving field is composed of two collinear linearly polarized vortex beams with opposite—and different—OAM content (ℓ_1, ℓ_2) and same frequency (ω_0). Panels b) and c) show the tunability of the on-axis HHG spectra driven in He for 800 nm and 2 μm drivers, respectively. The line spacing can be tuned through the OAM content of the driving fields, up to the soft X-rays. Panels d) and e) show the extremely low divergence of the on-axis harmonic beams, if driven in Ar (where theory vs experiment is presented for the 25th harmonic) and He. The divergence is only lower than that obtained with Gaussian driving fields, and it also decreases with frequency.

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