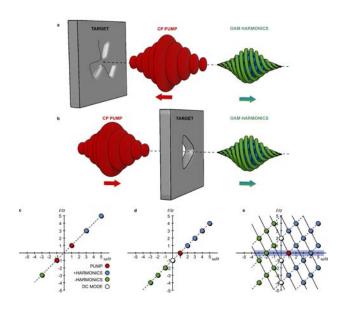
Theory and Computation

Laser harmonic generation with independent control of frequency and orbital angular momentum

The non-linear optical process of laser harmonic generation (HG) enables the creation of high-quality pulses of UV or even X-ray radiation, which have many potential uses at the frontiers of experimental science, ranging from lensless microscopy to ultrafast metrology and chiral science. Although many of the promising applications are enabled by generating harmonic modes with orbital angular momentum (OAM), independent control of the harmonic frequency and OAM level remains elusive. Here we show, through a theoretical approach, validated with 3D simulations, how unique 2-D harmonic progressions can be obtained, with both frequency and OAM level tuned independently, from tailored structured targets in both reflective and transmissive configurations. Through preferential selection of a subset of harmonic modes with a specific OAM value, a controlled frequency comb of circularly polarised harmonics can be produced. Our approach to describe HG, which simplifies both the theoretical predictions and the analysis of the harmonic spectrum, is directly applicable across the full range of HG mechanisms and can be readily applied to investigations of OAM harmonics in other processes, such as OAM cascades in Raman amplification, or the analysis of harmonic progressions in nonlinear optics.



Schematic illustrating the generation of higher-order harmonics: shown are the LCP $(\sigma=1, blue)$ and RCP $(\sigma=-1, green)$ harmonics of an incoming CP pump pulse (red) in a reflection and b transmissive configurations, from a threefold structured target. c-e Expected 2D harmonic spectrum from: c a flat target driven by an LP pump pulse with $\ell=1$; da circular aperture or dent target driven by a CP pump pulse of $\ell = 0$; and, e a threefold reflective or transmissive target with a CP pump pulse of $\ell = 0$. The blue shaded area indicates the potential for a frequency comb at $\ell = 0$.

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