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Multi-vortex high-harmonic beams from graphene's anisotropy

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High-order harmonic generation (HHG) is a unique tool to produce ultrafast structured laser beams in the extreme ultraviolet (EUV) spectral regime. The deep understanding of this process in gaseous media has triggered the emergence of a wide variety of interaction schemes to control the spatiotemporal properties of the emitted beams, resulting from both spin and orbital angular momentum conservation (SAM and OAM, respectively) [1]. By contrast, HHG in solids has begun to attract the interest of the community more recently. Crystals offer rich scenarios in HHG, where the coupling of the electromagnetic field with the target's structure introduces new phenomena, such as the matter-Talbot effect [2], or the recently observed anisotropic HHG from single-layer graphene [3].

Here, we study HHG in single-layer graphene driven by a mid-infrared vector beam (VB) at normal incidence (see Fig. 1, left panel). The exploration of the coupling of graphene's symmetries with the SAM and OAM of the driving beam requires a macroscopic picture of the interaction. In unstructured media, like gases, HHG preserves the geometry of a cylindrical VB driver [4], which is composed by two OAM modes, $l = \pm 1$. In such case, the simultaneous conservation of SAM and parity leads to the photon composition rules for the qth-order harmonic: $q = n_1 + n_2$ odd and $|n_2 - n_1| = 1$, where $n_1$and n_2 are the number of photons of the two modes. Hence, the OAM of the harmonic beam is restricted to $l = \pm 1$, constituting the cylindrical harmonic VB. In contrast, our theoretical simulations show that graphene's anisotropy [5] imprints two distinct properties into the generated harmonics: (1) a spin-dependent angular diffraction pattern and, (2) high OAM components. Strikingly, the fundamental symmetry associated with the simultaneous spin-orbit conservation forces the splitting of the harmonic beam into a set of EUV vortices with $|l| = 1$ and well-defined helicity. The right panel of Fig. 1 shows the intensity and phase distributions of the multi-vortex formation for the 9th harmonic, for the left circular (a and b) and right circular (c and d) components. Our study highlights the non-trivial interplay between the structural symmetries of the target and the driving beam in HHG, as well as its understanding in terms of both spin-dependent diffraction and the fundamental law of simultaneous conservation of SAM and parity. This opens exciting new routes for the generation of spatially complex EUV structured beams and for a deeper understanding of the ultrafast electron dynamics in solid systems.

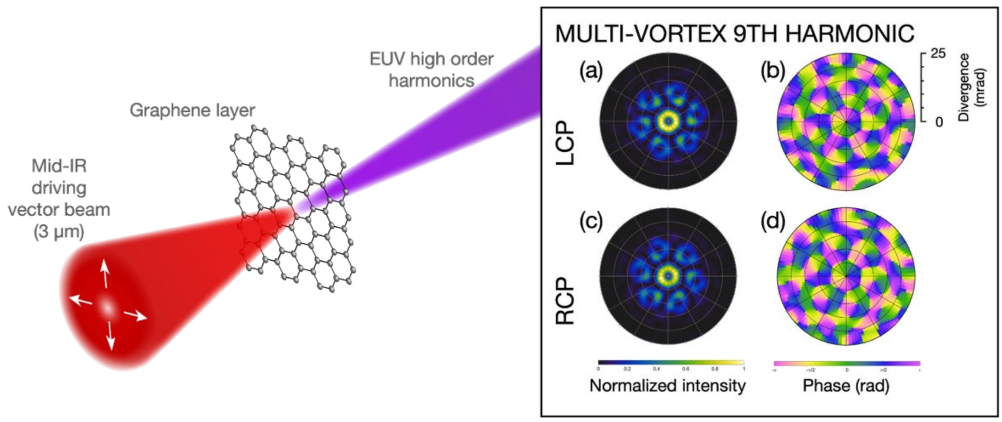

Figure 1. Multi-vortex high-order harmonics in single-layer graphene from a mid-infrared vector beam. The right panel shows the intensity and phase distribution of the 9th harmonic (33 nm).

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