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Multi-beam vortex generation induced by the non-linear optical anisotropy of graphene.

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Abstract. We analyse the high harmonic emission from single-layer graphene driven by infrared vector beams. We demonstrate that graphene's anisotropy offers a privileged scenario to explore non-trivial light spin-orbit couplings, which substantially extends the possibilities for the generation of high-harmonic structured beams currently studied in atomic and molecular targets. In our case, graphene's crystal symmetry introduces a spin-dependent diffraction pattern that, coupled with the fundamental conservation of the driver's topological phase, leads to the splitting of the harmonic field in a multi-beam structure, composed of spatially diverging vortices. Our work demonstrates that anisotropic targets are extraordinary tools to sculpt complex structured short-wavelength beams.

1 Introduction.

High-order harmonic generation (HHG) is well established as the main pathway to the production of high-frequency coherent radiation. The extreme non-linear, non-perturbative, response of atoms and molecules under intense driving leads to the emission of high harmonics. This process can be described using a strikingly simple mechanism [1]: electrons are tunnel ionized from the target elements, then evolve as quasi-free particles under the field excitation, following a trajectory redirected to the parent ion. Harmonics are radiated during the electron recollision with the ion, liberating its kinetic energy in the form of high-frequency radiation.

Recently the intense-field community has raised interest in the study of HHG in crystalline solids. This new playground offers evident advantages, as an increased harmonic conversion efficiency, together with promising scenarios, where crystal symmetries play a role during the harmonic conversion. Among them, for instance, HHG has been postulated as a realization of an ultrafast nanoscale temporal mater Talbot-Lau interferometer [2]. Driven in normal incidence, HHG in solids can be understood from a similar mechanism to that in atoms or molecules, now in terms of recolliding electron-hole trajectories in the reciprocal space [3].

Recent studies have demonstrated graphene as a very particular target for HHG. The presence of point degenerations in the energy dispersion, i.e. Dirac points, modifies the first step of HHG, as tunnel excitation is replaced by electron-hole pair creation during the non-adiabatic crossing of electron trajectories near the singular points [4]. Also, graphene confers HHG with a strong anisotropy [5,6]. Therefore, it is a natural playground to explore polarization-dependent modifications in HHG. In particular, the possibility of non-trivial structuring of harmonic vector beams, i.e. short-wavelength beams with an inhomogeneous distribution of polarization in the transverse plane.

2 Geometry.

We consider graphene single layer (SLG) driven at normal incidence by a 3-micron radial vector beam with waist 90 μm (see Fig. 1).

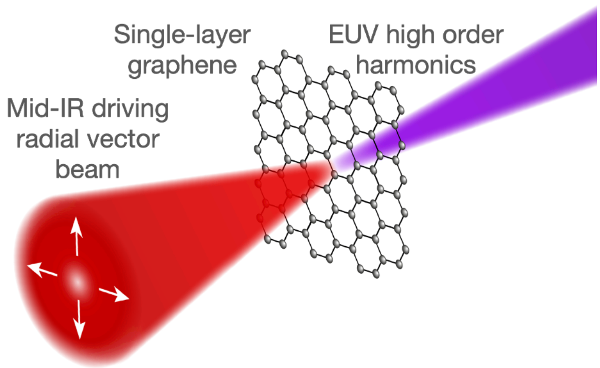

Fig. 1. Interaction geometry. A radial vector beam drives HHG in a graphene layer.

The vector beam is represented by a superposition of two Laguerre-Gauss vortices with topological charges $\ell_{1,2} = \pm 1$ and opposite polarization state $\sigma_{1,2} = \mp 1$, so that the net angular momentum of each beam component $k = 1,2$ is $j_k = \ell_k + \sigma_k = 0$. This condition is translated to the invariance of the field's geometric phase in each of the

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two beam components when circling around the vortex ring, since the azimuthal phase introduced by the topological charge compensates the geometrical phase acquired by a circularly polarized beam when rotating around the axis [7]. It has been demonstrated [8] that the phase invariance is preserved in HHG therefore, in isotropic media, harmonics driven by vector beams are generated as vector beams. As we report here, graphene's response being strongly anisotropic introduces a polarization-dependency in HHG that, for vector beam drivers, turns into an azimuthal modification of the harmonic emission and, therefore, the introduction high OAM charges. This modifies substantially the isotropic scenario.

3 Harmonic near-field distribution.

To compute HHG from SLG we follow [9]. First, we discretize the target layer into elemental surface elements, small enough to consider the driver's profile along them as constant. HHG from each element is computed in the nearest-neighbour tight-binding approximation, and propagated to the far field using the integral solution of Maxwell's equations. The total field is then computed as the addition of the elemental contributions.

Figure 2a shows the harmonic intensity profile at the target, for the total beam and its left circular polarized component, demonstrating a rotationally shifted necklace pattern depending on the polarization. This shifted near field configuration translates to a shifted rotation in the far-field pattern, therefore demonstrating graphene's spin-dependent diffraction.

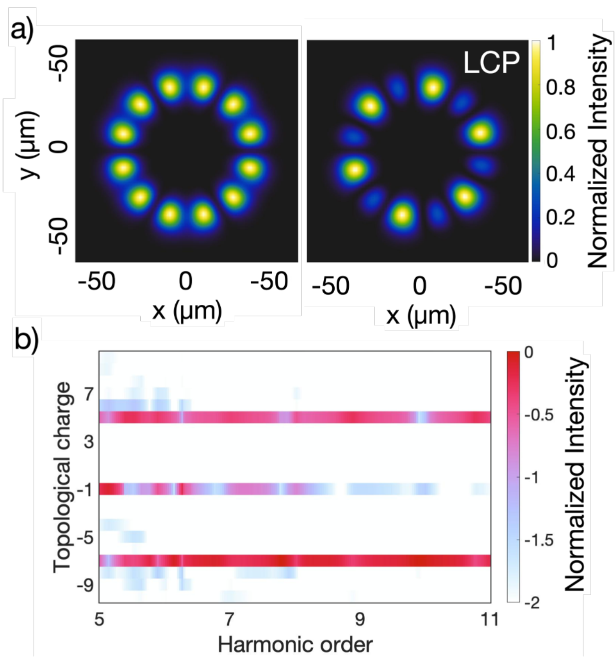

Fig. 2. (a) 9[th] harmonic near field intensity profile for (left) the total vector beam and (right) the left circular polarized component.

Figure 2b shows the angular momentum content of the near field harmonic. Most strikingly, harmonics are generated with high vortex charges, therefore apparently breaking the conservation rule for the total angular momentum $j_k = 0$ and, consequently, the preservation of the topological phase invariance that is found in HHG from isotropic charges.

4 Harmonic far-field distribution.

The analysis of the harmonic far-field distribution unveils the structural details of the harmonic field and resolves the apparent breaking of the topological phase invariance in HHG. We plot in Figure 3 the far-field intensity distributions of the 9[th] harmonic. Note that the harmonic field is composed of thirteen independent beams. The central beam is a ring structure corresponding to a radial vector beam, i.e. the composition of two vortex beams with opposite helicities. On the other hand, the twelve diverging beams correspond to vortex beams, with different azimuthal distributions depending on their polarization. Each of these field beams has a net angular momentum $j_k = 0$ and, therefore preserves the topological phase invariance of the driving beam.

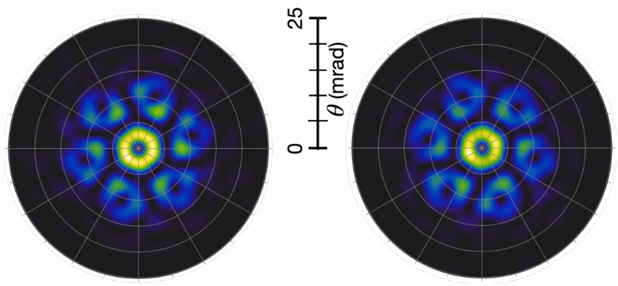

Fig. 3. Far field intensity distributions of the left (left) and right (right) polarized components of the 9th harmonic beam in terms of the azimuthal and divergence angles.

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