Ellipticity dependence of higher-order harmonics in solids: unraveling the interplay between intraband and interband dynamics

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Recently, we introduced an ab-initio time-dependent density-functional theory (TDDFT) framework that allows us to investigate the coupled interplay between the interband and intraband mechanisms of high-harmonic generation (HHG) from solids [1] without making a-priori model assumptions or strong approximations. Here, using HHG experiments on bulk silicon samples combined with TDDFT simulations, we study the complex physics underlying anisotropic harmonic emission, as reported by You et al. [2] for the strongly anisotropic ellipticity dependence of the 19th harmonic (HH19) generated in bulk MgO. In [2], the observed anisotropy was explained with real-space trajectories in a 2D one-band model including scattering from neighboring atomic sites. Our TDDFT simulations [3] and HHG experiments reveal that the various higher-harmonic orders generated in solids exhibit qualitatively different sensitivity to driver-pulse ellipticity $\varepsilon$ (not displayed here), resulting from a different response of intraband and interband dynamics [3], in contradiction with the model proposed in [2]. In fact, band-structure and joint-density-of-states (JDOS) effects become important [1].

![Figure 1](image_url)

**Figure 1.** Measured ellipticity dependence of HHG at normal incidence from Si: a quarter-wave plate (QWP) allows tuning the polarization of the driver pulses from linear (dotted blue vertical lines) to elliptic to circular polarization. The panels show ellipticity plots for different rotation angles of the [100]-cut 10-µm crystals as indicated. Excitation with ~120-fs 2.08-µm driver pulses with $I_0 = 10^{12}$ W/cm$^2$ in matter.

Fig. 1 shows experimental ellipticity profiles from Si. The driver-pulse polarization was varied from linear to elliptic to circular by a combination of a half- and a quarter-wave plate (QWP), to ensure that the axes of the polarization ellipse are not rotated. The HHG spectra were not corrected for the response of detection. As our TDDFT simulations predicted, the harmonics exhibit different behaviors: HH7 and HH9 have distinct sidelobes (see center panel in Fig. 1), the peaks of HH9 show an oscillatory behavior versus sample rotation. The computed ellipticity profiles for MgO exhibit an even stronger anisotropy compared to Si [3].

Our TDDFT simulations also reveal that non-zero driver-pulse ellipticity permits harmonic cutoff extension [Fig. 2(a)] and circularly polarized higher harmonics [Fig. 2(b),(c)] from a single-color driver pulse, whose helicity can be controlled by the driver-pulse helicity. This might open up a wealth of exciting new possibilities, e.g., for spectroscopy on magnetic materials.

![Figure 2](image_url)

**Figure 2.** (a) Calculated HHG spectra from MgO for linear ($\varepsilon = 0$) and elliptical ($\varepsilon = 0.65$) polarization for excitation with 25-fs 1333-nm pulses, $I_0 = 3 \times 10^{12}$ W/cm$^2$ in matter. In both cases, the major axis of the polarization ellipse is along $\Gamma\overline{X}$. The dashed lines indicate the position of the cutoff energy. (b) Evolution of the time-derivative of the electronic current in Si, bandpass filtered around the 11th harmonic for left-handed circular polarization. (d) Comparison of the left-handed and right-handed driven 11th harmonic. The $x$-component (major axis) is found to be identical for the two cases, whereas the $y$-components have opposite phase, demonstrating that the helicity of the 11th harmonic can be controlled by the driver-pulse helicity. Excitation in (b),(c) with 25-fs 3000-nm pulses with peak intensity in matter of $I_0 = 10^{12}$ W/cm$^2$.