# Circularly polarized high-order harmonics from solids driven by single-color infrared pulses

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**Abstract.** Intraband and interband dynamics generating high-order harmonics in solids exhibit qualitatively different responses to driver-pulse ellipticity, enabling ellipticity-based harmonic cutoff extension. *Ab-initio* calculations and experiments demonstrate generation of circularly polarized harmonics from single-color pulses.

**Keywords:** High-harmonic generation, solids, TDDFT, interband and intraband dynamics, elliptic and circular polarization.

Recently, we introduced an *ab-initio* time-dependent density-functional theory (TDDFT) framework [1] that allows us to investigate the coupled interplay between the intraband and interband mechanisms of high-order harmonic generation (HHG) from solids 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 harmonic emission, that can lead, e.g., to a strongly *anisotropic* ellipticity dependence, as was reported in [2] for bulk MgO. In [2], the observed anisotropy was interpreted with *real-space* trajectories in a 2D one-band model including scattering from neighboring atomic sites. Later, the possibility of maximal harmonic yield at finite ellipticity $\varepsilon$ was proposed in the regime of semi-metallization [3] and observed in graphene [4]. This regime, however, occurs at much higher intensities than used in [2]. Here, using HHG experiments and TDDFT simulations, we study anisotropic harmonic emission and demonstrate generation of circularly polarized high-order harmonics. Our TDDFT simulations [1, 5] and HHG experiments [6] (see Fig. 1) reveal that the various higher-harmonic orders generated in solids exhibit qualitatively different sensitivity to the driver pulse's ellipticity $\varepsilon$, resulting from a different response of intraband and interband dynamics [5], in

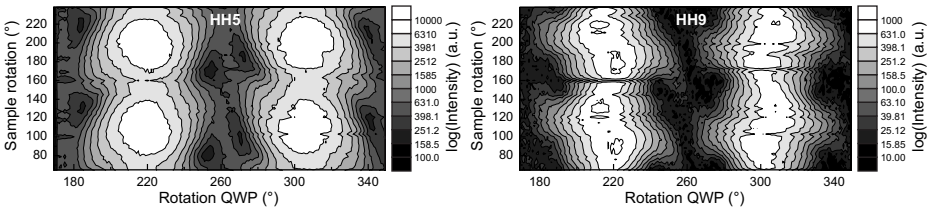


**Fig. 1.** Measured ellipticity dependence of harmonics HH5 (below band gap) and HH9 (above gap) from [100]-cut 10-$\mu$m-thick silicon: a quarter-wave plate (QWP) allows tuning the driver pulse's polarization from linear (214°) to elliptic to circular (259°), and back to orthogonal linear (304°). Excitation with ∼120-fs 2.08-$\mu$m pulses with peak intensity $I_0 = 0.56\,\mathrm{TW/cm^2}$ in matter. Note the oscillation of anisotropy of HH9.

contradiction with the model proposed in [2]. In fact, band-structure and joint-density-of-states (JDOS) effects become important to understand the observed behavior [1, 5].

Figure 1 reports measured ellipticity profiles from bulk silicon. 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 harmonics HH5 and HH9 exhibit different behaviors: when increasing the sample rotation angle, HH9 exhibits an oscillatory anisotropic behavior and sidelobes for certain angles, in contrast to HH5.

By exploiting the ellipticity as new control knob, we demonstrate the possibility of steering the electron wavepacket in *momentum space* ($\boldsymbol{k}$ space), and our theory predicts that the HHG cutoff can be strongly modified and even extended for non-zero ellipticity (in Fig. 2 by 30%) [5]. This increase of the HHG cutoff, which can not occurr in HHG from gases, is even more impressive, considering the fact that the maximum electric field at finite ellipticity is a factor $(1+\varepsilon^2)^{-1/2}$ (=0.84 for $\varepsilon$=0.65) smaller than the field strength for linear polarization.

Our TDDFT simulations in [5] predicted that odd *circularly* polarized harmonics with alternating helicities can be generated from single-color driver pulses in these crystals, which is consistent with group-theoretical considerations for

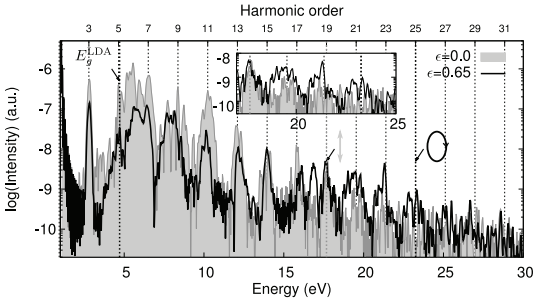


**Fig. 2.** Calculated HHG spectra from MgO for linear ($\varepsilon = 0$) and elliptical ($\varepsilon = 0.65$) polarization for excitation with 25-fs 1333-nm pulses, $I_0 = 3\,\mathrm{TW/cm^2}$ in matter. In both cases, the major axis of the polarization ellipse is along $\overline{\Gamma K}$. The thick dashed black and grey lines (at harmonic orders 19 and 25) indicate the positions of the harmonic cutoff for both cases. Adapted from [5].

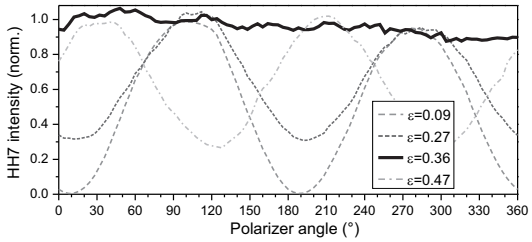


**Fig. 3.** Experimental evidence for circular polarization of HH7 from Si: dependence of the HH7 intensity on polarizer angle for various driver ellipticities $\varepsilon$ (as indicated). For $\varepsilon = 0.36$ (thick black curve), HH7 is circularly polarized.

circularly polarized driver fields [7], as Si and MgO both have point group $O_h$ ($m3m$). In our experiment shown in Fig. 3, we indeed observe a circularly polarized harmonic HH7 (for $\varepsilon$=0.36), here in Si. This might open up exciting new possibilities for the spectroscopy on materials exhibiting strong spin-orbit coupling or magnetic properties.

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