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.

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 strongly *anisotropic* ellipticity dependence of the 19th harmonic (HH19) generated in bulk MgO [2]. In [2], the observed anisotropy was interpreted with *real-space* trajectories in a 2D one-band model including scattering from neighboring atomic sites. Our TDDFT simulations[3,4] and HHG experiments[4] (see Fig. 1) reveal that the various higher-harmonic orders generated in solids exhibit *qualitatively* different sensitivity to the driver pulse's ellipticity ε, 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 crucial to understand the behavior[1,3].

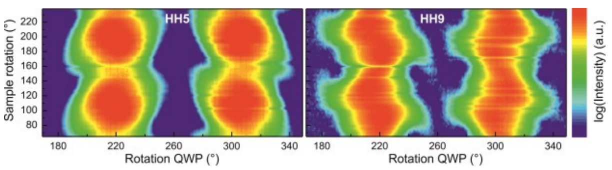

Fig. 1. Measured ellipticity dependence of harmonics HH5 (below band gap) and HH9 (above gap) from [100]-cut 10-μm-thick silicon: a quarter-wave plate (QWP) allows tuning the driver pulse's polarization from linear (214°) to elliptic to circular (259°), and back to orthogonal linear (304°). Excitation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

By exploiting the ellipticity as new control knob, we demonstrate the possibility of steering the electron wavepacket in *momentum 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%). This increase of the HHG cutoff 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 ε = 0.65) smaller than the field strength for linear polarization.

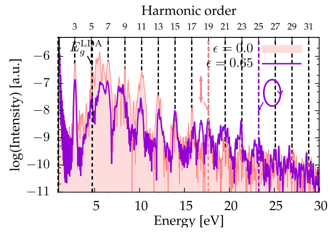

Fig. 2. Calculated HHG spectra from MgO for linear (ε = 0) and elliptical (ε = 0.65) polarization for excitation with 25-fs 1333-nm pulses, I_0=3 TW/cm^2 in matter. In both cases, the major axis of the polarization ellipse is along ($\overline{\Gamma K}$). The colored dashed lines indicate the positions of the harmonic cutoff for both cases.

As predicted in [3], we experimentally observe for the first time a *circularly* polarized harmonic (see HH7 in Fig. 3) generated by a *single-color driver pulse* in a bulk crystalline solid. This might open up exciting new possibilities for the spectroscopy on magnetic materials.

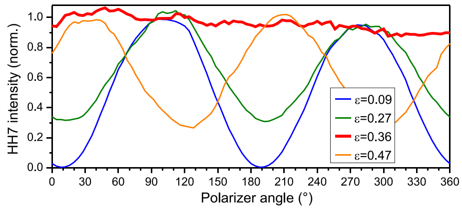

Fig. 3. Experimental evidence for circular polarization of HH7 from Si: dependence of the HH7 intensity on polarizer angle for various driver ellipticities ε (as indicated). For ε=0.36 (red), HH7 is circularly polarized.

REFERENCES

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