Symmetry and Polarization of High-Order Harmonic Generation from Solids

Shima Gholam-Mirzaei[1], Shicheng Jiang[2], Erin Crites[1], John E. Beetar[1], Ruifeng Lu[2], C. D. Lin[3], and Michael Chini[1,4]

[1]Department of Physics, University of Central Florida, Orlando, Florida 32816, USA
[2]Department of Applied Physics, Nanjing University of Science and Technology, Nanjing 210094, P. R. China
[3]J. R. Macdonald Laboratory, Department of Physics, Kansas State University, Manhattan, Kansas 66506, USA
[4]CREOL, the College of Optics and Photonics, University of Central Florida, Orlando, Florida 32816, USA
Author e-mail address: Michael.Chini@ucf.edu

Abstract: We study the polarization states of high-order harmonics emitted from a-cut ZnO crystals driven by femtosecond mid-infrared laser pulses. The polarization states of even and odd harmonics are sensitive to structural symmetries of the crystal.

OCIS codes: (320.0320) Ultrafast optics; (320.7150) Ultrafast spectroscopy

1. Introduction

High-order harmonic generation (HHG) from bulk crystals [1], for which the crystal axes can be precisely aligned with respect to the laser polarization, has provided a unique tool for studying the role of spatial symmetry in strong-field interactions [1-4]. In particular, studies of the dependence of harmonic spectra on the crystal orientation have been instrumental to explaining the underlying mechanisms of HHG in solids. For example, the dependence of harmonic spectra on the laser polarization for centrosymmetric crystals has provided information about the real-space electron trajectories in crystals [3] and the interplay between inter- and intra-band trajectories [5]. For solids lacking inversion symmetry, investigating the orientation dependences and polarization states of even and odd harmonics has also highlighted different underlying mechanisms responsible for HHG. Namely, the parallel-polarized (with respect to the driving laser polarization) emission of even harmonics in α-quartz has been attributed to strong coupling between bands [6] or asymmetric dipole matrix elements [7], while perpendicularly-polarized even harmonics have been described as a manifestation of Berry curvature [4].

Here, we explore the orientation dependence of harmonics generated from a-plane bulk ZnO by intense mid-IR laser both experimentally and theoretically. In addition to strong even harmonics generated for driving laser polarization along the crystal's optic axis, we observe weak even harmonics along other angles. These weak symmetry features can be attributed to currents along the Zn-O bond direction, which can be explained and calculated using one-dimensional two-band Semiconductor Bloch Equations (1D SBE) in combination with the Linearly-Coupled Excitation (LCE) model. This method successfully reproduces the major features of the polarization-resolved experimental HHG spectrum, suggesting that the polarization-dependent features can be mainly explained by structural symmetry alone. Such measurements suggest HHG as an all-optical method for probing structural symmetries in solids with few-femtosecond resolution, which can complement ultrafast x-ray techniques.

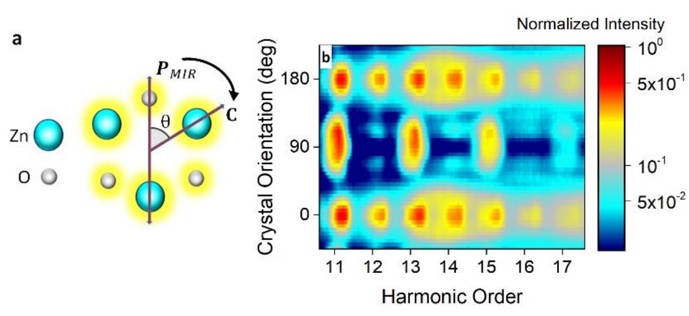

Fig. 1. a) ZnO structure, b) Orientation dependent spectrum for above-gap harmonics.

2. Experiment and results

In the experiments, idler pulses from a commercial optical parametric amplifier (Light Conversion ORPHEUS) with central wavelength of 3.6 µm, energy of 10 µJ, and pulse duration of 90 fs are focused onto the exit plane of a 0.3 mm thick a-cut zinc oxide target. First, we varied the orientation of the optic axis with respect to the laser polarization direction. Fig. 1.a shows the crystal structure, where the angle θ indicates the direction of the laser polarization with respect to the optic axis. The generated harmonics are focused into a UV-VIS-NIR spectrometer (Ocean Optics HR2000+ES) which covers the spectral range of harmonics from 4th to 17th order. As shown in Fig. 1.b, in addition to the emission of even harmonics at $\theta=0^o$ and $\theta=180^o$ (along the optic axis), relatively weaker maxima at around $\theta=65^o$ and $\theta=115^o$ are observed. This is consistent with 1D SBE which accounts for the amplitude and phase of dipole moment between valence and conduction band [7] and predicts weak even order peaks at $\theta=61^o$ and $\theta=119^o$.

Structural symmetries are known to affect the polarization states of high-order harmonics, as well as their intensities. In Figure 2, the orientation dependence of above-gap harmonics ($\Delta\varepsilon_{ZnO} = 3.3$ eV) is plotted for harmonics polarized parallel (a, b) and perpendicular (c, d) to the driving laser polarization. Theoretical calculations (b, d) based on the LCE model, and including the phase of the transition dipole matrix element, are in good agreement with the experimental results. By comparing Fig. 1 to Fig. 2(a) and (c), we find that odd harmonics mainly remain parallel to the driving beam polarization for $\theta = 0^o$, $\theta = 90^o$, and $\theta = 180^o$, whereas weak odd harmonics appear with perpendicular polarization with respect to the laser polarization at $\theta=15^o$ and $\theta=165^o$, as well as $\theta=72^o$ and $\theta=108^o$, which is consistent with the theoretical calculations shown in Fig. 2(b) and (d). On the other hand, even harmonics are emitted parallel to the laser polarization along the optic axis, and likewise for the observed peaks at $\theta=65^o$ and $\theta=115^o$ in Fig. 1. This agrees well with theoretical predictions in Fig. 2(b). Similar to odd harmonics at $\theta=15^o$ and $\theta=165^o$ we observe relatively weak maxima in the spectrum of the perpendicular polarization component shown in Fig. 2(c), which are reproduced at slightly different angles of $\theta=35^o$, $\theta=145^o$ in the calculated spectra.

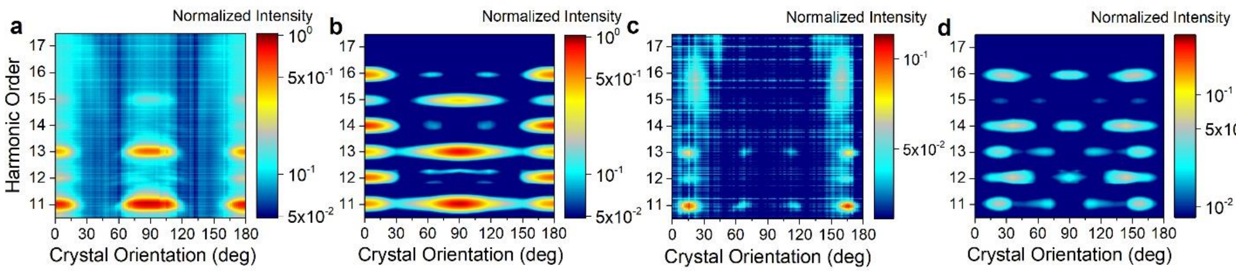

Fig.2 polarization resolved above-gap harmonic spectrum for (a) parallel (c) perpendicular emission component in experiment and (b) parallel (d) perpendicular emission component in theory.

Finally, we analyze the polarization states of the generated harmonics by rotating the polarizer angle with respect to the driving beam polarization for particular orientation angles of the crystal. For driving laser polarization along the crystal's optic axis, odd and even harmonics are polarized parallel to one another, as expected. In contrast, when the driving laser polarization is perpendicular to the optic axis, odd and even harmonics are perpendicularly polarized. The LCE model well describes the dependence of the odd and even harmonic yields and polarizations on the input laser orientation, suggesting the importance of symmetry in determining the polarization properties of HHG.

4. Summary

In this study, the polarization properties of high-order harmonics generated by intense mid-IR laser have been investigated experimentally and theoretically. We find that the polarization states of harmonics act as a sensitive probe of crystal symmetries, and that harmonic polarization states differ from the fundamental laser polarization when symmetry is broken. Due to the few-femtosecond timescale of HHG, polarization-sensitive measurements can potentially be used to probe dynamic structural changes in a crystal, such as phase transition or melting, and can therefore complement time-resolved x-ray and electron diffraction imaging for probing the dynamics in solids.

The work done at UCF was supported by the Air Force Office of Scientific Research (FA9550-16-1-0149) and by the National Science Foundation (1806135). SJ and R. L. are supported by NSF of China (21373113), the Fundamental Research Funds for the Central Universities (30920140111008 and 30916011105), and the NSF of Jiangsu Province (BK20170032). C.-D.L is supported in part by Chemical Sciences, Geosciences and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy, under Grant No. DE-FG02-86ER13491.

5. References

[1] S. Ghimire *et. al.*, "Observation of high-order harmonic generation in a bulk crystal" Nat. Phys. **7,** 138–141 (2011).
[2] S. Gholam-Mirzaei *et. al.*, "High harmonic generation in ZnO with a high-power mid-IR OPA" APL **110**, 061101 (2017).
[3] Y. S. You *et. al.*, "Anisotropic high-harmonic generation in bulk crystals" Nat. Phys. **13**, 345–349 (2017).
[4] H. Liu *et. al*.,"High-harmonic generation from an atomically thin semiconductor" Nat. Phys. **13**, 262–265 (2017).
[5] N. Tancogne-Dejean *et. al.*, "Ellipticity dependence of high-harmonic generation in solids originating from coupled intraband and interband dynamics" Nat. Commun. **8**, 745 (2017).
[6] F. Langer *et. al.*, "Symmetry-controlled temporal structure of high-harmonic carrier fields from a bulk crystal" Nat. Photon. **11**, 227–231 (2017).
[7] S. Jiang *et. al.*, "Role of the transition dipole amplitude and phase on the generation of odd and even high-order harmonics in crystals" Phys. Rev. Lett. **120**, 253201 (2018).