Polarization-state-resolved high-harmonic spectroscopy on crystalline solids

Nicolai Klemke[1,2], Nicolas Tancogne-Dejean[4,5], Giulio M. Rossi[1,2], Yudong Yang[1], Roland E. Mainz[1,2], Angel Rubio[4,5], Franz X. Kärtner[1,2,3], and Oliver D. Mücke[1,3]

[1] *Center for Free-Electron Laser Science CFEL, Deutsches Elektronen-Synchrotron DESY, Notkestraße 85, 22607 Hamburg, Germany*
[2] *Physics Department, University of Hamburg, Luruper Chaussee 149, 22761 Hamburg, Germany*
[3] *The Hamburg Center for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany*
[4] *Max Planck Institute for the Structure and Dynamics of Matter, Luruper Chaussee 149, 22761 Hamburg, Germany*
[5] *European Theoretical Spectroscopy Facility (ETSF)*
nicolai.klemke@desy.de

Abstract: We investigate the polarization states of high-harmonics generated from different semiconductors. We find significant deviations from the driving pulses' polarization and circularly polarized harmonics from elliptically polarized pulses.

The polarization of high-harmonics generated from solids can deviate from the polarization of the driving laser. This has been demonstrated with linearly polarized fields and was interpreted as the influence of crystal symmetries [1], band structure [2] as well as Berry curvature [3] on high-harmonic generation (HHG). Recently, we investigated the polarization states of high-harmonics generated in silicon with elliptically polarized driving fields [4,5]. Among other things, we found that the polarization states of individual harmonics were qualitatively different for the same driving conditions and demonstrated coherent circularly polarized harmonics with elliptically polarized excitation. The polarization-states of the emitted harmonics depend sensitively on the driving pulse's ellipticity as well as the sample rotation, showing distinct 'islands' of high ellipticity for certain driving conditions [5]. It is therefore interesting to explore if a similar behavior is observed on other crystals than Si.

Here, we present polarization-state-resolved high-harmonic spectroscopy from (0001)-cut, 50-µm-thin ZnO. High-harmonics are generated with 2.1-µm, 70-fs driving pulses with a vacuum peak intensity of ~0.7 TW/cm². We detect the harmonics with an *Ocean Optics* HR4000 spectrometer. Due to the spectrometer used, the highest harmonic order observed is the ninth harmonic (HH9). The driving ellipticity is varied by usage of a quarter- and a half-waveplate to keep the major axis constant. Fig. 1 summarizes some of our most important results.

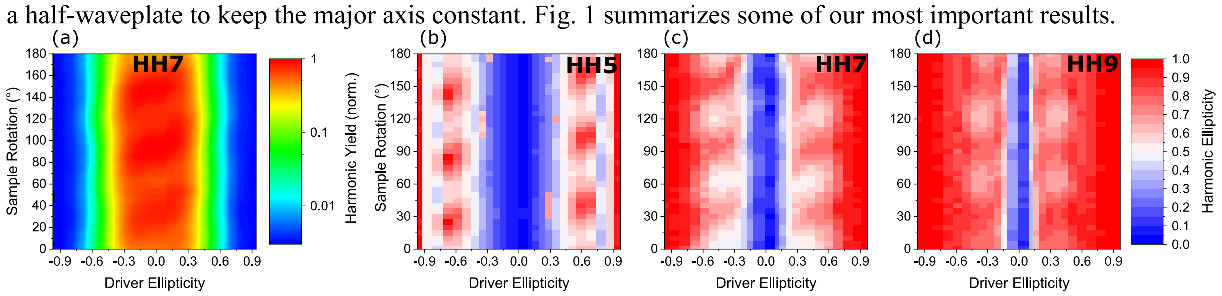

Figure 1. (a) Harmonic yield of HH7 vs. driver ellipticity and sample rotation. (b)-(d): Harmonic ellipticity maps of HH5-HH9. Ellipticity of 0 refers to linear, 1 to circular polarization.

In accordance with earlier results on ZnO [6] and in contrast to results from Si [4,5] and MgO [4,7], the harmonic yields (see, e.g., HH7 in Fig. 1(a)) do not exhibit strong non-atomic ellipticity dependences, meaning the harmonic yield of all observed harmonics drops monotonically with increasing driving ellipticity. However, the harmonic ellipticity maps in Fig. 1(b)-(d) show distinct features that are unknown from gas HHG. Most strikingly, HH9 exhibits ellipticities larger than 0.7 for most driving ellipticities with small "islands" of slightly lower harmonic ellipticities for driving ellipticities of $|\varepsilon| \approx 0.3$. Islands of this kind can also be found in HH7 and, as islands of high ellipticity, for HH5. The features reproduce the hexagonal 60°-symmetric crystal structure of (0001)-cut ZnO. The band structure of ZnO is simpler than for Si with only one conduction band involved in this photon energy range. It is interesting to note that also the features in the harmonics' ellipticity-maps seem less pronounced. This makes a simpler theoretical analysis feasible, which could analyze the influence of interband and intraband mechanisms on the harmonics' polarization-states with elliptically polarized excitation.

Apart from a straightforward approach to generate circularly polarized attosecond pulses, our work reveals fundamental insights into the lightwave-driven currents underlying HHG from solids.

References:

[1] F. Langer *et al.*, Nat. Phot. **11**, 227 (2017)
[2] K. Kaneshima *et al.*, Phys. Rev. Lett. **120**, 243903 (2018); Y. S. You *et al.*, Opt. Lett. **44**, 3 (2019)
[3] T. T. Luu, H. J. Wörner, Nat. Commun. **9**, 916 (2018)
[4] N. Tancogne-Dejean *et al.*, Nat. Commun. **8**, 745 (2017)
[5] N. Klemke *et al.,* Nat. Commun., in press
[6] S. Ghimire *et al.*, Nat. Phys. **7**, 138 (2011)
[7] Y. S. You *et al.*, Nat. Phys. **13**, 4 (2017)