Surface chirality probed by circular solid-state high-harmonic generation with polarization-tailored strong fields

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High-harmonic generation (HHG) in solids is a powerful and versatile tool for the investigation of electronic properties of dielectric media, including all-optical band reconstructions [1], exciton analysis [2] valence electron mapping [3] and work on momentum-dependent states [4]. Solid targets also offer the possibility to control HHG by functionalization via structural or chemical modifications [5] as well as polarization-tailored excitation fields which can be matched with the crystal symmetry. Using for example two-color counter-circularly polarized driving fields, such as well-known from gas-phase HHG experiments [6], allows for entirely new possibilities in symmetry-resolved and chiral-sensitive strong-field spectroscopy in the condensed phase via the generation of circular high harmonics.

Here, we demonstrate the first generation of circularly-polarized high-harmonic radiation in solids using three-fold bi-chromatic driving fields [7]. By matching the symmetry of the light polarization with the symmetries of the crystals, we study orientation-resolved, chiral electronic and magnetic properties in silicon dioxide (quartz) and on the surface of magnesium oxide (MgO), respectively. In particular, by aligning the driving field with the crystal axes, we determine the point group. Furthermore, we use the circular polarization of the emitted harmonics to probe the structural chirality of quartz(0001) as well as the ferromagnetic order on the polar MgO(111) and the non-polar MgO(100) surface originating from spontaneous chiral-symmetry-breaking. The observed surface-sensitivity stems from the strong absorption of the harmonics near the band edges and indicates coupling to exciton states.

In conclusion, we introduce a novel route for chiral extreme-UV generation and spectroscopy in solids. Our approach is transferable, in principle, to any semiconductor or insulator and, moreover, the inherently short pulse duration in the femtosecond regime and the broad spectrum of harmonics from solids opens new paths for time-resolved chiral spectroscopy in the condensed phase.

References

[1] G. Vampa *et al.*, Phys. Rev. Lett. **115**, 193603 (2015).

[2] F. Langer *et al*., Nature **533**, 225–229 (2016).

[3] H. Lakhotia et al., Nature ***583***, 55–59 (2020).

[4] Bai, Y. et al., Nat. Phys. 1–5 (2020) doi:10.1038/s41567-020-01052-8.

[5] M. Sivis *et al.*, Science 357, 303–306 (2017).

[6] A. Fleischer, O. Kfir, T. Diskin, P. Sidorenko & O. Cohen, Nat. Photonics **8**, 543–549 (2014).

[7] T. Heinrich *et al.*,, ArXiv:201005558 (2020).