CFEL SCIENCE

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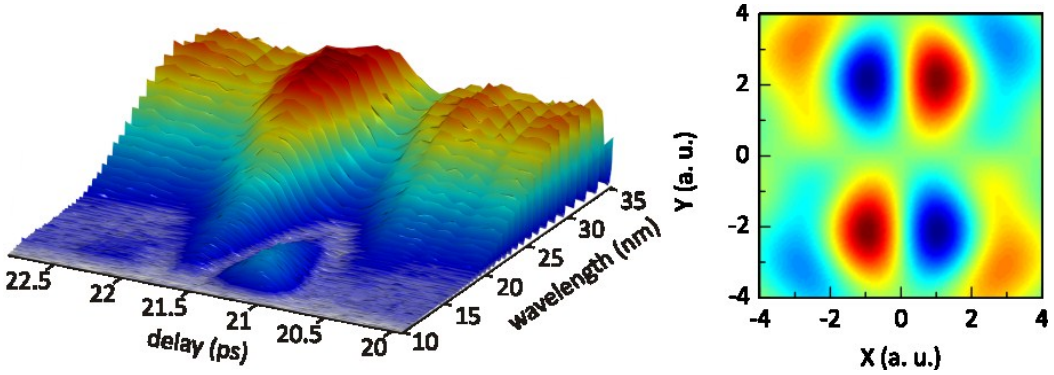
A generalized approach to molecular orbital imaging by high-order harmonic generation



A macroscopic ensemble of molecules exposed to an intense laser pulse emits coherent XUV radiation appearing as high-order harmonics of the laser frequency. The spectral amplitude and phase of the radiation produced by single molecules is encoded in the macroscopic emission. Molecular imaging by high-order harmonics [1] requires the measurement in amplitude and phase of the harmonic field emitted by a single molecule for different molecular orientations.

By modulating the angular distribution of the molecular ensemble and by exploiting a self-referencing approach, we show that both the amplitude and phase of single-molecule XUV emission can directly be retrieved from harmonic intensity spectra as a function of photon energy and angular orientation. This result is based on the exploitation of few-cycle mid-infrared laser pulses [2].

The outcomes are exploited for a tomographic reconstruction of the outermost molecular orbital of CO_2 [3]. In particular the approach to tomographic reconstruction presented here provides a route for the solution to major difficulties in contemporary HHG spectroscopy of complex species. These findings redeem the idea of HOMO spectroscopy by high-order harmonic generation and open new perspectives on the imaging of molecular orbitals.

[1] J. Itatani et al., Nature 432, 867 (2004).
[2] C. Vozzi et al., Opt. Lett. 32, 2957 (2007).
[3] C. Vozzi et al., Nat. Phys. 7, 822 (2011).

Host: Franz X. Kärtner, CFEL General Seminar