Simulation of High Harmonic Generation in Solids

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Synopsis We simulate high harmonic generation in dielectrics by ultrashort laser pulses using TDDFT. We show that inclusion of decoherence by dephasing into TDDFT is crucial for a realistic description.

To overcome fundamental limitations in high harmonic generation (HHG) from single atoms, solid targets have moved into the focus of interest. For single atoms the increase of the high-energy cut-off with increasing wavelength, $E_{\text{cut}} \propto \lambda^2$, goes hand in hand with the dramatic decrease of the harmonic intensity, $I \propto \lambda^{-5.5}$. In principle, solids promise to overcome this intensity limitation due to the large density of target atoms. Indeed, HHG from dielectrics has already been observed with well-pronounced harmonics up to very high orders. In parallel, theoretical methods are sought for to identify the source of the emitted radiation and to help optimizing the emission process.

We use a real-space real-time implementation of time-dependent density functional theory (TDDFT) employing periodic boundary conditions to model the interaction of ultrashort laser pulses with solids [1]. However, when applied to a realistic 3D solid for one fixed intensity I of the IR pulse (in this case for diamond) only very few harmonics on top of a broad background above the band-gap energy are recognizable (top panel). This is in contrast to the clean high-harmonic spectrum observed in experiment (starting with [2]).

We show that a realistic description of solid-state harmonics requires the treatment of the average over the inhomogeneous intensity distribution as well as decoherent processes. Forming a coherent average over the ensemble of harmonic spectra generated by the inhomogeneous intensity distribution in the laser spot clearly "cleans" the spectrum by amplifying the harmonics while damping other frequencies (bottom panel). Moreover, electron-phonon and electron-electron scattering not accounted for in standard TDDFT further purifies the spectrum and dampens the post-pulse ringing. We have developed an open-system TDDFT which accounts for decoherence without changing the time-depending density of the system [3]. We find an almost noiseless HHG spectrum (decoherence time $\tau = 2$ fs, center panel).

Open-system TDDFT in combination with intensity averaging promises to allow for a realistic simulation of HHG in dielectrics.

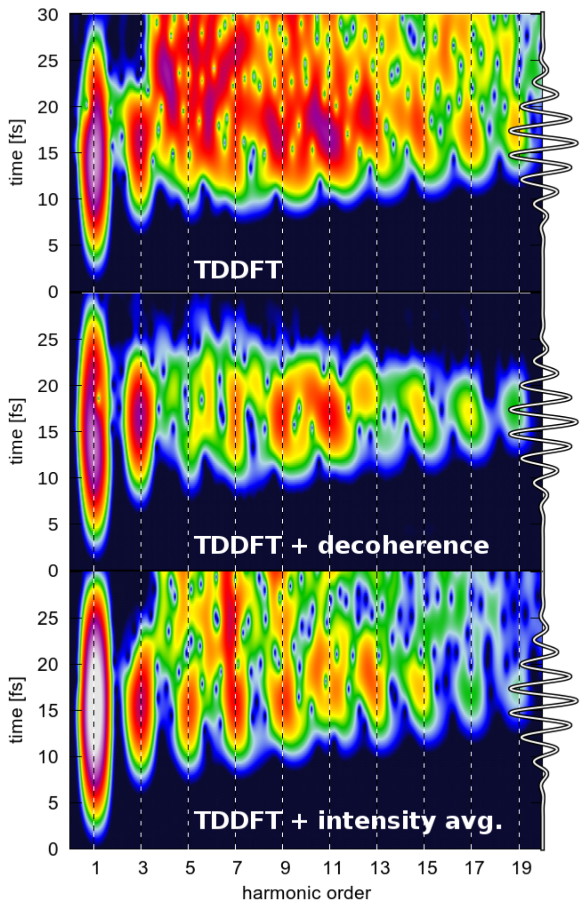

Figure 1. Wavelet transforms of radiation emitted from diamond due to irradiation by an ultrashort laser pulse. TDDFT (top panel), TDDFT with decoherence (middle panel), and intensity average of TDDFT results (bottom panel).

References

[1] T. Otobe *et al.* 2009, *J. Phys.: Condens. Matter* **21** 064224

[2] S. Ghimire *et al.* 2011 *Nature Physics* **7**, 138

[3] I. Floss *et al.* 2017, in preparation

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