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Generation of high harmonics in solids

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Synopsis We have developed an *ab-initio* multi-scale simulation to model light synthesis and propagation through transparent media accounting for the non-linear response of the solid. Using this approach we are able to investigate the build-up of high harmonics induced by strong and short laser pulses along the propagation direction inside a large band-gap dielectric, diamond. Detailed analysis of the results allows to identify minimum requirements necessary to realistically model laser-solid interactions.

Irradiation of crystalline insulators with strong and short laser pulses gives rise to non-linear response of the material and the generation of high harmonics (HHG). In recent experiments pronounced peaks at odd (and even) multiples of the laser frequency have been observed in the HHG spectra from crystals with (broken) inversion symmetry [1]. In contrast, simulated high harmonic spectra obtained from microscopic *ab-initio* models were found to be affected adversely by signals of coherent particle-hole pair excitations resulting in noisy spectra without clear harmonic peaks above the band-gap energy.

Several remedies for this problem have been proposed most of which only marginally footed on physical principles (e.g. extremely short decoherence times). As it turns out, laser-solid interaction is a true multi-scale problem which cannot be simplified below a certain level. We find that in order to reach at least qualitative agreement with experiment it is imperative to use both a sophisticated description of the electronic dynamics induced in a crystal cell on the microscopic level and the inclusion of mesoscopic effects of the extended system accounting for the pulse propagation in the dense medium and the inhomogeneous illumination of the crystal.

We combine refined single-cell simulations (time-dependent density functional theory and solution of semiconductor Bloch equations based on *ab-initio* band structures) with Maxwell's equations and include the transverse intensity distribution of the focal spot resulting in a clear and pronounced high harmonic spectrum [2]. Furthermore, we have accounted for decoherence in TDDFT on a phenomenological level [3].

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Analyzing the buildup of the final HHG spectrum within the crystal we are able to identify the ingredients necessary for realistic simulations of HHG from solids that yield agreement with experiments: 1) a fully 3-dimensional microscopic simulation of the crystal unit cell with an accurate band structure and a consistent set of coupling matrix elements, 2) the coupling of the microscopic response to the macroscopic pulse propagation which should also account for the transverse intensity distribution of the laser spot, and 3) accounting for decoherence with typical decoherence times $\tau_{\rm dec} > 10$ fs. Such simulations are able to model the transition from a noisy spectrum in the entrance layer to the clear HHG spectrum exiting the diamond layer after a propagation distance of 1 μm (Fig. 1).

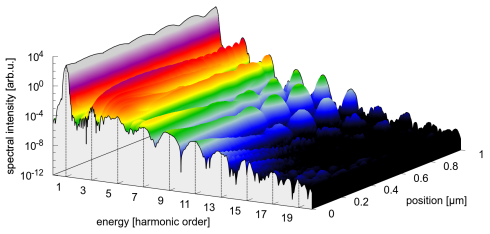

Figure 1. High harmonic spectrum (log-scale) induced in diamond by a linearly polarized few-cycle laser pulse with a carrier wavelength of 800 nm and peak intensity of 2×10^{13} W/cm^2 as a function of the position along the propagation direction inside a 1 μm thick diamond crystal.

References

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