# Project title: High-harmonic spectroscopy of vibrating chains

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### Current state of the art

Linear chains are important model systems in molecular and condensed matter physics. For instance, the seminal Su-Schrieffer-Heeger (SSH) model [1] was initially intended to describe solitons in polyacetylene but later, in a simplified version without mobile ions, became the paradigmatic, simplest toy model for topological condensed matter [2]. High-harmonic generation (HHG) in such (topological) linear chains was investigated theoretically in detail by us both with time-dependent density functional (TDDFT) theory [3, 4] and in tight-binding (TB) approximation [5]. TDDFT, with common, practicable exchange-correlation functionals, takes electron-electron interaction into account only on a mean-field level, i.e., without strong electron correlations. The SSH TB model does not take electron-electron interaction into account at all. In both kinds of modelling a huge difference in the harmonic yield between the topological and the trivial phase was found [3, 4, 5]. The HHG spectra in the metallic phase (i.e., in the midst of the topological phase transition when the band gap closes) are very different for TDDFT and TB because screening due to polarization of the chain is extremely important but only taken into account in TDDFT.

The role of nuclear motion on HHG has been thoroughly investigated on a fully quantum mechanical level in full dimensionality only for the smallest molecule $H_2^+$ [6] where double-slit like interference effects have been observed. For larger molecules and condensed matter, the ion motion is usually treated classically via Ehrenfest dynamics. As the ion motion typically occurs on a 10-femtosecond or longer time scale, and the IR laser pulses that generate HHG have durations on a similar time scale, HHG in principal facilitates the probing of vibrational motion [7]. The question is how to conclude from the HHG signal that phonons have been excited.

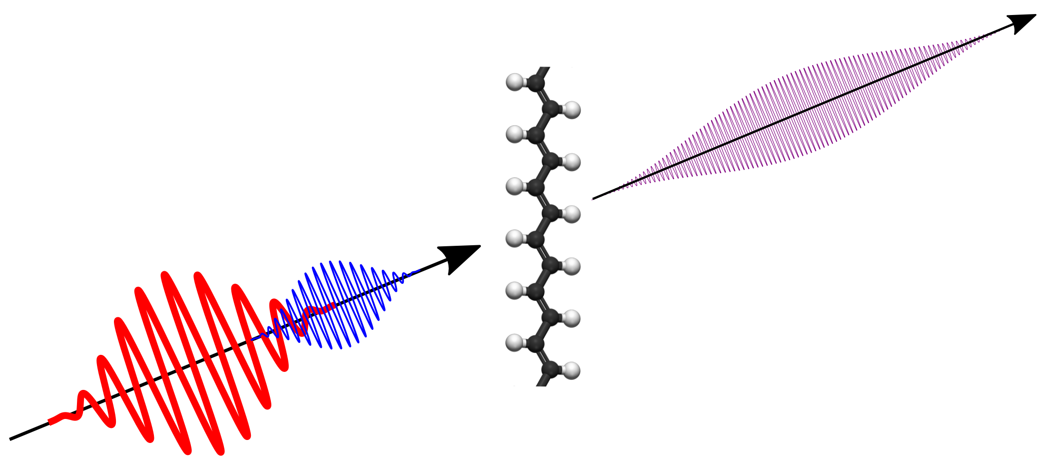

Figure 1: Sketch of the principal setup. The incoming laser pulses (pump and probe) initiate and probe electron dynamics in the chain-like organic molecule (e.g., polyacetylene). Ions will start to move on the 10-fs time scale and affect the harmonic radiation (purple).

### Research goals and working program

This project is aiming at a detailed understanding of (i) the vibrational motion that is initiated in the chains by the HHG-generating laser itself, (ii) the features that these vibrations cause in the HHG spectra, and (iii) the features in HHG spectra caused by dedicated pump pulses.

The principal setup is shown in Fig. 1. The polarization(s) of the incoming laser pulse(s) are under a certain angle with respect to the molecular axis. The most interesting dynamics is expected if the electrons are driven along the chain, possibly polarizing it and therefore counteracting the electric field of the laser (dynamical screening). The nuclei will start to move and vibrate after a while, which is expected to modify HHG. In a pump-probe setup, the dependence of the HHG spectra will be analyzed as a function of the pump-probe delay.

Concerning the theoretical treatment, we will use (a) the original SSH model [1] (coupled to the laser field) where the non-interacting electrons are treated in tight-binding and the ions are described classically, (b) TDDFT for the electrons and Ehrenfest dynamics applied to the ions, and (c) the Hubbard-Holstein model [8] where the interacting electrons are described with the Hubbard-Hamiltonian and nuclear vibrations are taken into account quantum mechanically via "local phonons".

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