

The mechanism of high harmonic generation in liquids and its application for electron scattering spectroscopy

Content

High harmonic generation (HHG) takes place in all phases of matter. In gases it has been extensively studied and is well-understood. In solids research is ongoing, but a consensus is forming for the dominant HHG mechanisms. In liquids however, no established theoretical model exists yet, and approaches developed for gases and solids are generally inapplicable. Here there are many open questions such as cutoff scaling laws, the dominant HHG mechanisms, and more. Advancement on this front may lead to novel light sources, and are especially appealing for ultrafast spectroscopy of chemistry in solutions. In particular, the great success of high harmonic spectroscopy in both gas and solid phases motivates extending this technique to the liquid phase, but this is hindered by the lack of a microscopic understanding of the underlying light-driven electron dynamics. In this talk, I will present our recent collaborative effort in tackling this problem by combining HHG experiments from thin liquid flat jets, and theory based on a novel ab-initio approach that relies on clusters we have developed. We have employed these techniques to study HHG in different liquids and laser conditions, focusing on the microscopic mechanism and the cutoff scaling. Surprisingly, and opposed to the established cutoff scaling in gases and solids, we found that the liquid HHG cutoff does not scale with the driving wavelength, and scales very weakly with the driving power. We propose an extended semi-classical model which takes into account the electron-ion scattering processes in the liquid to explain our findings. The simple model qualitatively describes the main structure of the HHG spectra, and identifies the electron mean free paths as a principal factor in the HHG process in liquids. Our work thus paves the way to the utilization of HHG spectroscopy for probing electron mean free paths in liquids in the low energy regime ($<10\text{eV}$), and has potential for probing ultrafast chemistry in solutions.

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