Ab-initio cluster approach for high harmonic generation in three-dimensional liquids: characteristic features

Ofer Neufeld[1,2,*], Zahra Nourbakhsh[1], Nicolas Tancogne-Dejean[1], Angel Rubio[1,3]

[1]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, 22761, Germany.
[2]Physics Department and Solid State Institute, Technion - Israel Institute of Technology, Haifa, 320003, Israel.
[3]Center for Computational Quantum Physics (CCQ), The Flatiron Institute, New York, NY, 10010, USA.

High harmonic generation (HHG) takes place in all phases of matter. In gases, it has been extensively studied and is very well-understood. In solids research is ongoing, but a consensus is forming for the dominant HHG mechanisms. In liquids however, no theoretical model exits yet. Approaches developed for gases and solids are generally inapplicable to liquids, because liquids lack long-range correlations, and are infinite systems. Here there are many unanswered fundamental questions such as cutoff scaling laws, the dominant HHG mechanisms, and more. Advancement on this front, which may pave the way to novel light sources and ultrafast spectroscopies, is hindered by the lack of theoretical frameworks to study complex liquids interacting with strong fields. We present an *ab-initio* cluster-based approach for the nonlinear interaction of liquids and intense arbitrary laser pulses. We employ it for HHG calculations in water, ammonia, and methane liquids, and compare the characteristic response of polar and non-polar liquids. We analyze the time-frequency and spectral structure of liquid-HHG and find that the spectrum routinely separates to two plateaus that are generated by distinct mechanisms. A semi-classical model is proposed to explain our findings. Our work paves the way to feasible calculations of liquid HHG, and illustrates the unique nonlinear nature of liquids.