High Harmonic Spectroscopy of Binary Liquids

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High Harmonic Spectroscopy has been a very powerful tool in ultrafast physics allowing access to electron dynamics and structure in various media. The underlying physics is well understood in gases and is being extensively studied in solids, but in liquids, the mechanism is still unclear since there are relatively little experimental and theoretical investigations. The main goal of this project is to understand the physical mechanism of high harmonic generation in liquid phase. Key questions are the role that intermolecular structure plays in the liquid mechanism and can one extract ultrafast solvation dynamics. To this end, we have studied HHG in different solvent classes and in varying binary mixtures.

I will describe recent studies on thin liquid phase targets that have introduced a new wrinkle to strong field physics defined by the local short-range order. I will show that mixtures composed of a methanol solvent manifest an interference effect in the harmonic emission indicative of local solvation dynamics in a binary solution and correlate with radial distribution functions derived from classical molecular dynamics simulations (MDS). In addition, a 1D TDSE model shows that a local scattering potential can mediate the interference consistent with the experiment and the MDS. This work illustrates the potential of high harmonic spectroscopy in liquids and the ability to extract information about the solvation structure on the sub-angstrom scale in addition to further exploring the mechanisms of HHG in liquid phase.