Contribution ID: **193** Type: **Contributed talk**

Attosecond-resolved probing of recolliding electron wave packets in liquids and aqueous solutions

Monday, June 23, 2025 11:30 AM (20 minutes)

We establish high-harmonic generation (HHG) in liquids as a powerful ultrafast probe for tracking spatial and temporal electron dynamics on attosecond time scales. Beyond the traditional three-step model, we uncover new nonlinear features such as multi-plateau structures and bandgap modification. These findings open pathways to attosecond-scale exploration of solvation dynamics, light–matter interactions, and electronic structure in complex environments.

High-harmonic generation (HHG) in bulk liquids has been recently explained by the "scattering-limited three-step model" [1]. In liquids such as H_2O, D_2O, and alcohols, the harmonic cut-off energy remains fixed and independent of laser wavelength, intensity, and pulse duration [1,2]—an outcome of strong electron scattering and dominant on-site recombination. However, this gas-like picture fails to account for the emergence of higher-order nonlinearities.
Here, we report the discovery of a second plateau in the HHG spectra of multiple liquids [3], marking a new regime of electron dynamics driven by recombination with neighboring molecules—particularly from the second solvation shell—enabled by hole delocalization [4,5]. This plateau displays unique signatures, such as a weak cutoff scaling and distinct ellipticity dependence, supported by advanced experiments, ab-initio simulations, and semi-classical models. Additionally, two-color interferometric measurements [6,7] provide attosecond-resolved access to the recollision process, revealing a large linear atto-chirp and an effective, field-induced reduction of the electronic band gap by several electron volts. Aqueous salt solutions exhibit spectral minima whose positions and depths are sensitive to anion type and con-centration. These features are well-described by a two-emitter interference model and reflect modulations in the relative phase and band structure induced by chemical environment and laser field. Together, these findings establish HHG in liquids as a versatile tool for probing ultrafast phenomena—capturing both temporal recollision dynamics and spatial recombination pathways. By moving beyond the simplistic gas-phase analogy, we unlock the potential of HHG to explore complex light–matter interactions, electronic structure modifications, and solvation dynamics on attosecond time scales.

References

[1] A Mondal et al., Nature Physiscs 19, 1813-1820 (2023)
[2] A Mondal et al., Optics Express 31, 34348–343461 (2023)
[3] A Mondal et al., Nature Photonics, under review
[4] I Jordan et al. Science 369, 974-979 (2020).
[5] X Gong et al. Nature 609, 507–511 (2022).
[6] N Dudovich et al. Nature Physics 2, 781–786 (2006).
[7] G Vampa et al Nature 522, 462–464 (2015).

Author: HANS JAKOB, Worner (ETH Zürich)

Presenter: HANS JAKOB, Worner (ETH Zürich)

Session Classification: Session 2 - Gas Phase Dynamics