# Theoretical Description Of High Order Harmonic Generation In Liquids

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**ABSTRACT**

High-order harmonic generation (HHG) is a non-linear strong-field process that produces coherent extreme ultraviolet (XUV) radiation and enables attosecond spectroscopy of ultrafast electronic dynamics [1]. While HHG has been extensively investigated in gases [2] and solids [3], its study in liquids has remained limited due to significant experimental challenges, despite the central role of liquid environments in chemical and biological processes. The first experimental observation of HHG in liquids in 2018 [4], followed by the first theoretical study only in 2022 [5], marked the beginning of a rapidly developing field in which many fundamental aspects remain to be elucidated.

In this work, we contribute to closing the gap between theory and experiments by developing an efficient fully quantum one-dimensional model tailored to capture HHG in disordered liquid phases. In parallel, we are using ab initio Ehrenfest molecular dynamics [6] simulations using the deMon2k software package [7] to incorporate the coupled electron–nuclear motion and the fluctuating local environments characteristic of liquids. These simulations will employ optimized Gaussian basis sets specifically adapted to describe continuum electronic states in HHG [8], which are essential for accurately modeling strong-field ionization and high-harmonic generation in condensed-phase systems.

These complementary approaches aim to provide a robust foundation for understanding strong-field interactions in liquid-phase systems. The methods currently under development are intended to clarify the key mechanisms underlying HHG in liquids and to guide upcoming theoretical and experimental developments in attosecond science applied to complex media.

## REFERENCES

1. P. Antoine, A. L'Huillier, and M. Lewenstein. *Attosecond pulse trains using high-order harmonics*, Physical Review Letters, vol. 77, no. 7, pp. 1234–1237, 1996.
2. K. Miyazaki and H. Sakai. *High-order harmonic generation in rare gases with intense subpicosecond dye laser pulses*, Journal of Physics B: Atomic, Molecular and Optical Physics, vol. 25, no. 3, 1992.
3. S. Ghimire and D. A. Reis. *High-harmonic generation from solids*, Nature Physics, vol. 15, pp. 10–16, 2019.
4. T. T. Luu, Z. Yin, A. Jain, *et al. Extreme-ultraviolet high-harmonic generation in liquids*, Nature Communications, vol. 9, p. 3723, 2018.
5. O. Neufeld, Z. Nourbakhsh, N. Tancogne-Dejean, and A. Rubio. *Ab Initio Cluster Approach for High Harmonic Generation in Liquids*, Journal of Chemical Theory and Computation, vol. 18, no. 7, pp. 4117–4126, 2022.
6. G. Friesecke and M. Koppen. *On the Ehrenfest theorem of quantum mechanics*, Journal of Mathematical Physics, vol. 50, no. 8, p. 082102, 2009.
7. A. de la Lande, A. Alvarez-Ibarra, *et al. Molecular Simulations with in-deMon2k QM/MM, a Tutorial-Review*, Molecules, vol. 24, no. 9, p. 1653, 2019.
8. E. Coccia and E. Luppi. *Optimal-continuum and multicentered Gaussian basis sets for high-harmonic generation spectroscopy*, Theoretical Chemistry Accounts, vol. 135, p. 43, 2016.