# PhD project :
# Theoretical description of high-order harmonic generation in liquids

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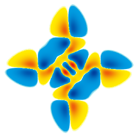

## Summary

High order harmonic generation is a highly non-linear process triggered when an intense infrared laser interacts with atoms or molecules. Since the 80s, this spectroscopic process has allowed to follow electronic dynamics in real time at their natural time (attosecond) and space (angström) scales. Recent experimental advances allowed to observe high order harmonic generation in the liquid phase, but the mechanism is yet to fully uncover. This thesis project is about the theoretical modelling of high order harmonic generation in the liquid phase. The methods that will be used are ab initio simulations for quantitative results, approached analytical and semi-analytical calculations and toy model numerical calculations for qualitative and interpretative results and modelling.

## Supervision

The PhD will be supervised by Marie Labeye, Maîtresse de Conférences (Tenured Assistant Professor) and co-supervised by Anne Boutin, Directrice de Recherche (Research Director), in the Theoretical Chemistry group at PASTEUR laboratory, Ecole Normale Supérieure, Paris.

## Candidate profile

The candidate should have a Master's degree diploma in physics or chemistry, basic knowledge in quantum mechanics and spectroscopy, and a sufficient understanding of English for scientific reading. Moreover some skills or experience in programing, numerical simulations, or optics would be greatly appreciated.

## Context

High-order harmonic generation (HHG) is a highly non-linear process triggered by the interaction of a high intensity infrared laser pulse with atoms or molecules. It consists of the emission of a very short coherent light pulse, composed of harmonics of the incoming light, and that were used to measure the fastest dynamics every observed [1, 2].

In addition to its interesting properties as a light source, HHG also contains both structural and dynamical information on the emitting system itself. It is thus used as a spectroscopy, called High-order Harmonic Spectroscopy (HHS), that allows to detect and characterize in real time ultrafast atomic and molecular processes, and to image electronic properties. Recent examples include the detection of ultrafast dynamics in gas and solids [3, 4], and imaging of orbitals via tomography [5]. In order to extract information from the HHG spectrum, a solid theoretical understanding of the physical processes is required. These experimental advances were supported by theoretical developments, both numerical and analytical, to which Marie Labeye has contributed [6, 7].

Very recently, HHG was observed for the first time in the liquid phase [8, 9], a theoretical models were proposed [9]. This opens exciting opportunities for theoreticians to develop models that would allow to transpose the successful experimental and theoretical toolbox from the gas and solid phase to the liquid phase.

## Références


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[2] S. Beaulieu et al. Attosecond-resolved photoionization of chiral molecules. *Science*, 358(6368) :1288 (2017).

[3] S. Baker et al. Probing Proton Dynamics in Molecules on an Attosecond Time Scale. *Science*, 312(5772) :424 (2006).

[4] T. T. Luu et al. Extreme ultraviolet high-harmonic spectroscopy of solids. *Nature*, 521(7553) :498 (2015).

[5] P. Peng et al. Attosecond imaging of molecules using high harmonic spectroscopy. *Nature Reviews Physics*, 1(2) :144 (2019).

[6] M. Labeye et al. Optimal Basis Set for Electron Dynamics in Strong Laser Fields : The case of Molecular Ion H2+. *J. Chem. Theory Comput.* (2018).

[7] —. Dynamical distortions of structural signatures in molecular high-order harmonic spectroscopy. *Phys. Rev. A*, 99(1) :013412 (2019).

[8] T. T. Luu et al. Extreme–ultraviolet high–harmonic generation in liquids. *Nature Communications*, 9(1) :3723 (2018).

[9] A. Mondal et al. High-harmonic spectroscopy of low-energy electron-scattering dynamics in liquids. *Nat. Phys.*, 19(12) :1813 (2023). Number : 12 Publisher : Nature Publishing Group.