



**Mardi 10 Novembre 2026 | 11:00 | Auditorium de l'IPCMS**

**Thomas Schnappinger**

*Stockholm University*

***From Ultrafast Processes to Strong Coupling: An Ab Initio Molecular Perspective on Light-Matter Interactions***

Light can control chemistry in many ways. Short laser pulses can drive molecules far from equilibrium in a matter of femtoseconds. Optical cavities can reshape the electromagnetic environment in which molecules react. Experiments in both cases record spectra, and ab initio simulations are needed to translate these spectra into the motion of electrons and nuclei. In the first part of my talk, I will demonstrate how simulations can be used to decode time-resolved spectra of photoinduced processes. A pump pulse excites or ionizes the molecule, which then responds. A probe pulse records this response. Multiconfigurational methods, such as RASSCF/RASPT2 with spin-orbit coupling included, describe the valence- and core-excited states of neutral and ionized molecules throughout this process. Since core-level spectroscopy is element-specific, simulated core-level spectra reveal the location of charge in a molecule at any given moment. I will illustrate this approach using charge transfer in  $\text{CF}_3\text{I}^+$  after strong-field ionization. Attosecond extreme-ultraviolet transient absorption spectroscopy in the group of H. J. Wörner, combined with our simulations, revealed a 1.5-fs delay in population transfer [1]. In the second part, I will turn to polaritonic chemistry. Although strong vibrational coupling in optical cavities has been reported to modify the reactivity and properties of molecules without any chemical change, the underlying mechanism is still debated. To address this issue using ab initio theory, we developed a cavity Born–Oppenheimer Hartree–Fock approach that treats cavity photon modes as nuclear coordinates [2]. Using this approach, we computed vibro-polariton spectra [3] and polarizabilities [4] under strong vibrational strong coupling. This method is now available to the scientific community in the form of the open-source package psi4-cbo [5]. To begin, I will discuss the meaning of light-matter coupling strength for a real cavity in the context of ab initio methods and explain why dipole self-energy matters. Using water clusters as an example, I will demonstrate how ab initio simulations can interpret vibro-polariton infrared and Raman spectra and relate them to transmission spectra measured in experiments. Together, these two topics demonstrate how ab initio simulations can provide molecular insights ranging from attosecond charge transfer to molecules in a cavity.

[1] Matselyukh et al., Nat. Commun. 16, 7211 (2025).

[2] Schnappinger et al., JPCL 14, 8024 (2023).

[3] Schnappinger & Kowalewski, JCTC 19, 9278 (2023).

[4] Schnappinger & Kowalewski, JCTC 21, 5171 (2025).

[5] Schnappinger & Kowalewski, arXiv:2609.17711 (2026).