Theory and simulation of ultrafast processes in molecules

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The interaction of light and matter is responsible for a variety of photophysical and photochemical phenomena occurring in nature, like photosynthesis, in the human body, like vision, and in technological devices, like photovoltaics. Theoretical modeling of these phenomena requires to be able to describe the complex interplay of electronic and nuclear motion beyond the Born-Oppenheimer approximation [1], ie including nonadiabatic effects, over ultrafast time scales ranging from femtoseconds to picoseconds.

The exact factorization of the electron-nuclear wavefunction is a formalism introduced in 2010 by Gross and coworkers to analyze and to simulate nonadiabatic processes [2,3]. Its original electron-nuclear formulation has been used to derive various flavors of trajectory-based algorithms [4,5] to simulate ultrafast relaxation processes initiated by photoexcitation, like photoisomerization [6] or photodissociation [7]. However, extensions of the original formalism to treat electron-only systems (exact electron factorization) [8,9] and photon-electron-nuclear systems (exact PEN factorization) [10, 11] have been proposed to study processes such as the dynamics of electrons in a strong laser field or photodynamics in the strong coupling regime.

In this talk, I will present an introduction to the theory of nonadiabatic ultrafast dynamics with the exact factorization and I will give an overview of its recent applications.

[1] F. Agostini, B. F. E. Curchod, WIREs Comput. Mol. Sci. (2019).
[2] A. Abedi, N. T. Maitra, E. K. U. Gross, Phys. Rev. Lett. (2010).
[3] F. Agostini, E. K. U. Gross, Eur. Phys. J. B (2021).
[4] S. K. Min, F. Agostini, E. K. U. Gross, Phys. Rev. Lett. (2015).
[5] C. Pieroni, F. Agostini, J. Chem. Theory Comput. (2021).
[6] C. Pieroni, E. Sangiogo Gil, L.-M. Ibele, M. Persico, G. Granucci, F. Agostini, J. Chem. Theory Comput. (2024).
[7] A. Pollien, E. Villaseco Arribas, D. Lauvergnat, F. Agostini, Mol. Phys. (2024).
[8]. A. Schild, E. K. U. Gross, Phys. Rev. Lett. (2017).
[9] S. Giarrusso, F. Agostini, ChemPhysChem (2024).
[10] L. Lacombe, N. M. Hoffmann, N. T. Maitra, Phys. Rev. Lett. (2019).
[11] E. Sangiogo Gil, D. Lauvergnat, F. Agostini, J. Chem. Phys. (2024)