Joined experimental/theoretical study of the control of the ring opening in spiropyrane molecules : role of the quantum interferences

Fabien Gatti,
Institut des Sciences Moléculraires d'Orsay, Université Paris-Sud, France
B. Gonon, B. Lasorne,
Institut Charles Gehardt de Montpellier, France,
G. Karras, E. Hertz, F. Billard, B. Lavorel, O. Faucher,
University of Dijon, France

Most industrial chemistry is performed using temperature and pressure control which results in a waste of energy and the production of unwanted by-products. A major breakthrough consists in developing *a novel chemistry* based on a systematic use of quantum superposition of molecular quantum states ("pure state"), i.e. *quantum coherence*, induced by the excitation with *laser pulses* [1]. The central tool of control, in order to carry out photochemical reactions selectively and efficiently, is the laser pulses whose recent progress features high power, precise tunability, arbitrary shaping down to the ultimate subfemtosecond and attosecond timescales.
The correct theory to describe the corresponding dynamics is Molecular Quantum Dynamics [2]. In contrast to standard quantum chemistry calculations, where the nuclei are treated classically, molecular quantum dynamics can cover quantum mechanical effects in their motion. Although the calculation of large systems still presents a challenge - despite the considerable power of modern computers - new strategies have been developed to extend the studies to systems of increasing size.
In particular, we use here the Multi-Configuration Time-Dependent Hartree method (MCTDH) [3,4,5]. We focus mainly on a multidimensional quantum mechanical treatment applied to femtosecond ring opening process in spiropyrane molecules underlying the possibility to play with the wave properties of matter to change reactivity.

[1] F. Gatti, "Molecular dynamics simulated by photons", *Nature News & Views*, 557, 641 **(2018)**.

[2] *Quantum Physics, Applications to Chemistry*, F. Gatti, B. Lasorne, H.-D. Meyer and A. Nauts, Lectures Notes in Chemistry, Springer (2017).

[3] H.-D. Meyer, U. Manthe, and L.S. Cederbaum, *The multi-configurational time-dependent Hartree approach.* Chem. Phys. Lett. **165** (1990), 73.

[4] M. H. Beck, A. Jäckle, G. A. Worth and H.-D. Meyer, *The multiconfiguration time-dependent Hartree method: A highly efficient algorithm for propagating wavepackets*, Physics Reports **324** (2000), 1.

[5] H.-D. Meyer, F. Gatti, and G. A. Worth, editors, *Multidimensional Quantum Dynamics: MCTDH Theory and Applications.*, Wiley-VCH,(2009),Weinheim.