
Quantum description of spiropyran ring-opening dynamics

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Résumé

The purpose of this work is to understand and control the light induced ring opening of spiropyran molecules. This mechanism is characterized by a femtosecond time scale explained by a non-radiative decay at a conical intersection.[1] Thus we investigate the insight of this dynamic process by non-adiabatic quantum dynamics.

We construct coupled electronic potential energy surfaces taking into account dynamical electronic correlation at a post-CASSCF ab-initio level (XMCQDPT2 [2]). This is a particularly tedious task because of the strong anharmonicity of the surfaces, on the one hand, and of the need to describe correctly the topography and non-adiabatic couplings around the conical intersection on the other hand. To this end, we set up a general approach restricted to a few degrees of freedom containing the essential physical properties of the system.

We use the Multi Configuration Time Dependent Hartree (MCTDH) approach to perform non-adiabatic quantum dynamics simulation. With that, we simulate the experiments currently carried out in the group of Dijon (FRANCE), studying the ring-opening process by femtosecond spectroscopy coupled with LASER pulse control. These experiments already demonstrated the ultrafast time scale of the process and proved the possibility to change the yields, specifically using quantum interferences created by LASER coherent light.

Simulations show good agreement with this experimental absorption spectrum and dynamics time scale. Moreover, preliminary results tend to reproduce the effect of the control LASER pulse on the reactivity.

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2014, *140*(4) , 044301

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