Nonadiabatic Dynamics in Molecules – From Ultra-Fast Detection of Concical Intersections to Control of Avoided Crossings with Quantum Light

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Non-adiabatic processes are responsible for non-radiative decay in a large number of photochemical and photobiological processes. These fast sub-100-femtosecond decay channels are dominated by conical intersections (CIs). At a CI, the electronic and nuclear degrees of freedom frequencies are comparable and strongly mix due to the breakdown of the Born-Oppenheimer approximation. A major challenge for their direct detection is the rapidly varying gap between the electronic surfaces in their vicinity. Modern XUV/X-ray light sources provide spectral broad and temporal short pulses, which potentially allow for monitoring CIs directly. We present theoretical studies on ultra fast Raman methods [2], which make use of ultrashort X-Ray laser pulses and compare the results for different molecules.

In the second part of the presentation we explore how coherent control, which is usually done with laser fields, can be done with quantum light. Here we use a quantum description of the light field which enables us to engineer the quantum state of light and allows to manipulate the light-matter interaction [1]. We will demonstrate the different principles of control with quantum light on the avoided crossing in lithium fluoride. Using a quantum description of light together with the non-adiabatic couplings and vibronic degrees of freedoms opens up new ways to look at quantum control. We will show the deviations from control with purely classical light field and how back action of the light field becomes important in a few photon regime.

References

[1] András Csehi, Gábor J. Halász, Ágnes Vibók, and Markus Kowalewski. Quantum Control with Quantum Light of Molecular Nonadiabaticity. *Phys. Rev. A*, 100:053421, 2019.

[2] Markus Kowalewski, Kochise Bennett, Konstantin E Dorfman, and Shaul Mukamel. Catching conical intersections in the act: Monitoring transient electronic coherences by attosecond stimulated x-ray raman signals. *Phys. Rev. Lett.*, 115(19):193003, 2015.