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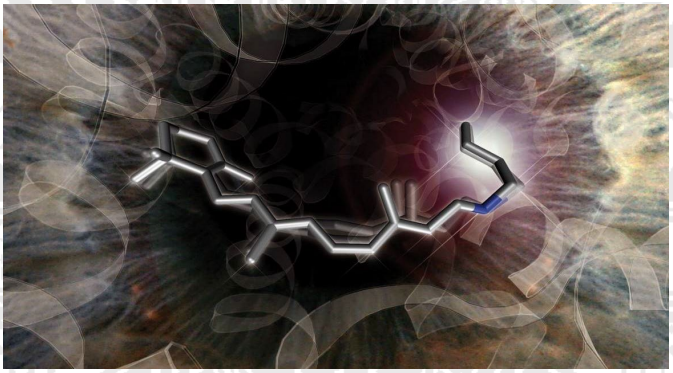

REAL-TIME OBSERVATION OF CONICAL INTERSECTION IN BIOMOLECULES

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Conical intersections (CIs) are regions of the potential energy landscape of a molecule where the electronic and nuclear degrees of freedom become strongly mixed and the Born–Oppenheimer approximation breaks down. CIs are ubiquitous features in the photophysics and photochemistry of molecules and can be considered as “doorways” through which the photoexcited wavepacket is efficiently funneled to a lower-energy electronic state. CIs play a dual role in the interaction of biomolecules with light: either to promote efficient conversion from a reactant to a product state in a photochemical reaction or to enable efficient dissipation of excess electronic energy, preventing a potentially harmful photochemical reaction [1]. An example of the first case are visual opsin proteins, in which the photoexcited retinal chromophore exploits a CI to promote ultrafast photoisomerization to a ground-state photoproduct which triggers visual transduction. An example of the second case are nucleobases, the building blocks of DNA, for which CIs are used to promote rapid dissipation of excited state energy, preventing photoreactions which could damage the genetic code.
Given the extreme speed of the processes leading to CIs, ultrafast optical spectroscopy is the elective tool for their observation. However, the direct visualization of a wavepacket passing through a CI is challenging, because the energy gap between the interacting levels changes very rapidly over a short time, calling for the combination of high temporal and spectral resolution. In this talk I will present examples of real-time visualization of CIs in biomolecules (opsin proteins [2] and nucleobases [3, 4]) using a specially developed ultrafast transient absorption spectroscopy system combining sub-20-fs time resolution with broad frequency tunability, from the UV to the infrared [5]. I will also discuss the potential of X-ray FELs to open new spectroscopic windows for the detailed study of the CI dynamics, via either element-specific probing or ultrafast structural dynamics.

[1] G. Cerullo and M. Garavelli, Proc. Natl. Acad. Sci. USA 117, 26553 (2020).
[2] D. Polli et al., Nature 467, 440 (2010).
[3] R. Borrego-Varillas et al., J. Am. Chem. Soc. 140, 16087 (2018).
[4] R. Borrego-Varillas et al., Nature Commun. 12, 7285 (2021).
[5] C. Manzoni and G. Cerullo, J. Opt. 18, 103501 (2016).