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Investigations of molecular photoenergy conversion using ultrashort x-ray pulses

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The conversion of light energy into other energy forms in molecules is the result of a concerted and ultrafast motion of electrons and nuclei, often under breakdown of the Born-Oppenheimer approximation. This talk is about ultrafast experiments aimed at resolving light induced ultrafast molecular dynamics with x-ray probe pulses using free electron lasers.
The molecules in the center of this talk are nucleobases, which encode genetic information in life. Although possessing really high UV absorption cross-sections, damaging events by UV absorption are relatively rare. The ultrafast transfer of electronic energy into harmless vibrational energy plays an important role as internal photoprotection mechanism. We present experiments probing the very first steps of this process by femtosecond resonant x-ray absorption spectroscopy at the oxygen K-edge. We deduce a less than 100 fs $\pi\pi \rightarrow n\pi$ transition, which plays a crucial role in the photoprotection of this nucleobase [1].
Thiolated nucleobases show an efficient and ultrafast relaxation into long-lived triplet states, contrasting with the ultrafast relaxation to the ground states observed in canonical nucleobases. This gives rise to interesting applications as photoinduced-cross linkers as well as problems related to its current use of thionucleobases as medication. We investigate the dynamics of 2-thiouracil via x-ray probing at the sulfur L-edge using the new URSA-PQ instrument for gas-phase spectroscopy at FLASH [2]. We find a direct connection between the charge moving within the molecule and the binding energy shifts observed in the photoelectron spectrum. This manifests itself into coherently modulated signals due to oscillating electronic population.

[1] Probing ultrafast $\pi\pi/n\pi$ internal conversion in organic chromophores via K-edge resonant absorption, T. J. A. Wolf, R. H. Myhre, J. P. Cryan, S. Coriani, R. J. Squibb, A. Battistoni, N. Berrah, C. Bostedt, P. Bucksbaum, G. Coslovich, R. Feifel, K. J. Gaffney, J. Grilj, T. J. Martinez, S. Miyabe, S. P. Moeller, M. Mucke, A. Natan, R. Obaid, T. Osipov, O. Plekan, S. Wang, H. Koch and M. Gühr
Nature Communications 8, 29 (2017)

[2] Following UV-induced electronic dynamics of thiouracil by ultrafast x-ray photoelectron spectroscopy, D. Mayer, F. Lever, D. Picconi, J. Metje, S. Alisauskas, F. Calegari, S. Düsterer, C. Ehlert, R. Feifel, M. Niebuhr, B. Manschwetus, M. Kuhlmann, T. Mazza, M.S. Robinson, R.J. Squibb, A. Trabattoni, M. Wallner, P. Saalfrank, T. J. A. Wolf, M. Gühr
https://arxiv.org/abs/2102.13431 (2021)

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