European XFEL Theory Seminar

Thursday, 17th June 2021, 16:00 *Zoom meeting*

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Photodissociation of ironpentacarbonyl (FeCO5) - from initial bursts of CO release to branching pathways in solution

Theoretical excited state molecular dynamics simulations of the initial non-adiabatic transitions after photoexcitation of ironpentacarbonylreveal an intricate ultra-fast dynamics. Coupled electron-nuclear dynamics involving metal-to-ligand charge-transfer (MLCT) excitationsand dissociative metal-centered excited states results in a gradual transition from coherent bond oscillatory motionto reoccurring bursts of carbonmonoxide release [1].These initial events set the stage for sequential CO release in gas phase [2] and complex pathways in ethanol solution [3]as experimentally observed in X-ray photoelectron spectroscopy (XPS) and iron L-edge resonant inelastic X-ray scattering (RIXS).Theoretical simulations of gas phase XPS and solution RIXS spectra based on multi-configurational quantum chemistryallow to kinetic modeling of the processes on different time-scales. The impact and challenges for explicit dynamical modellingof time-resolved X-ray spectra are discussed.

[1] A. Banerjee, M. R. Coates, M. Kowalewski, H. Wikmark, R. M. Jay, P. Wernet, M. Odelius,Photoinduced bond oscillations in ironpentacarbonyl give delayed, synchronous bursts ofcarbonmonoxide release, Unpubl. (20XX)

[2] P. Wernet, T. Leitner, I. Josesfsson, T. Mazza, P. S. Miedema, H. Schröder, M. Beye,K. Kunnus, S. Schreck, P. Radcliffe, S. Düsterer, M. Meyer, M. Odelius, A. Föhlisch,Communication: Direct evidence for sequential dissociation of gas-phase Fe(CO)5 via asinglet pathway upon excitation at 266 nm, J. Chem. Phys. 146 (2017) 211103.

[3] P. Wernet, K. Kunnus, I. Josefsson, I. Rajkovic, W. Quevedo, M. Beye, S. Schreck, S. Grübel,M. Scholz, D. Nordlund, W. Zhang, R. W. Hartsock, W. F. Schlotter, J. J. Turner,B. Kennedy, F. Hennies, F. M. F. de Groot, K. J. Gaffney, S. Techert, M. Odelius, A. Föhlisch,Orbital-specific mapping of the ligand exchange dynamics of Fe(CO) 5 in solution,Nature 520 (2015) 78–81

Host: Nils Brouwer

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