Structural dynamics of nano-/bio-molecules driven by a femtosecond XUV/XFEL pulse

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Highly excited and/or ionized molecules generated by the irradiation of femtosecond XUV/XFEL pulse undergoes exotics chemical reaction dynamics. In this talk, I will discuss my recent theoretical works on the XUV/XFEL induced structural dynamics of nano-/bio-molecules as follows:

1. Ultrafast nonadiabatic cascade and subsequent photofragmentation of XUV excited caffeine molecule [1]

We investigated the XUV driven femtosecond dynamics of caffeine as a model of prebiotic molecule. We observed a decay of excited cationic states with a time constant of 40 fs *via* femtosecond XUV-pump–IR-probe experiments. Guided by *ab-initio* many-body theory, this timescale is interpreted in terms of a non-adiabatic cascade *via* 10^2 highly correlated states (Fig. 3). These results show that both nonadiabatic coupling and electron correlation are the keys for ultrafast reaction dynamics in the highly-correlated electronic excited states.

2. XFEL induced Coulomb explosion of fullerene C_{60} and its application to imaging of the mid-IR induced coherent vibration [2,3]

We found that the time series of the angular anisotropy $\beta(t)$ of fast C^+ and C^{2+} fragments of C_{60}^{60+} produced by such an XFEL pulse reflects the instantaneous structure of C_{60} vibrationally excited by IR pulses (Fig. 2). The phases and amplitudes of excited vibrational modes and the coupling between excited modes can be successfully extracted from the expansion of $\beta(t)$ in terms of vibrational modes. This *proof-of-principle* simulation clearly demonstrates that various information of the structures and reaction dynamics of large clusters or biomolecules can be retrieved by decomposing the experimentally determined $\beta(t)$ into vibrational modes.

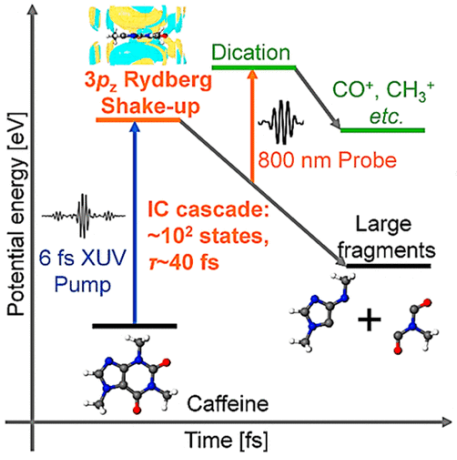

Fig. 1. Nonadiabatic cascade of caffeine [1]

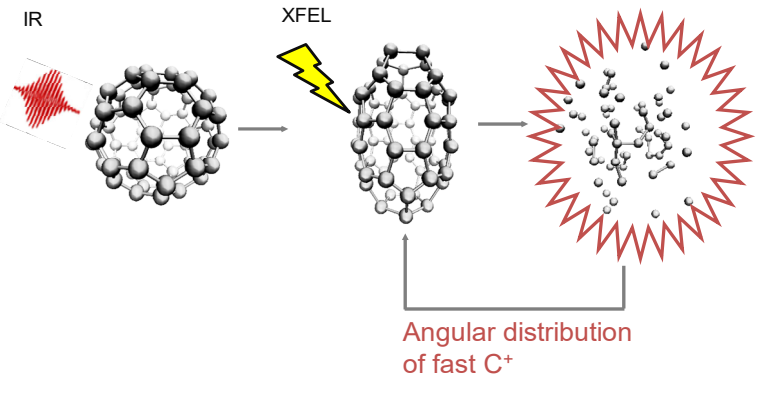

Fig. 2. XFEL Colomb explosion imaging of the mid-IR induced coherent vibration of C_{60} [3]

References (*Corresponding author):
[1] A. Marciniak, **K. Yamazaki*** *et al.*, *J. Phys. Chem. Lett.* **9**, 6927-6933 (2018).
[2] **K. Yamazaki** *et al.*, *J. Chem. Phys.* **141**, 121105 (2014).
[3] **K. Yamazaki*** *et al.*, *J. Chem. Phys.* doi: 10.1063/1.5115072 (2019), **Editor's pick**.