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Attosecond Coherent Electron Dynamics Triggered by XFEL Pulses

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Two key concepts characterize the ultrafast many-electron dynamics triggered in atoms and molecules upon interaction with ultrashort X-ray laser pulses produced by an X-ray free electron laser (FEL) source: quantum coherence [1] and quantum entanglement: the former underpins the few-femtosecond charge dynamics in molecules and the ensuing photochemical transformation; the latter limits the coherence that can be observed within each subsystem when interrogated individually by probe measurements.

I will discuss these key concepts and present results from two exemplary, combined experimental-theoretical pump-probe studies of quantum electronic coherences in molecules. The first one concerns molecular glycine [2], where few-femtosecond X-ray pulses from the FLASH X-ray FEL were used to trigger coherent electron dynamics in the glycine cation and probe it by resonant x-ray absorption and sequential double photoionization. The results provide a direct support for the existence long-lived electronic coherence up to 25 femtoseconds [2]. The second one addresses electron motion resulting from photoionization of a prototypical aromatic system (para-aminophenol) [3]. In this work [3], a pair of attosecond X-ray pulses from the LCLS-II X-ray FEL was employed to both trigger coherent dynamics in the para-aminophenol cation and track it with atomic-site specificity via attosecond X-ray absorption.

References:
[1] M. Ruberti, "Onset of ionic coherence and ultrafast charge dynamics in attosecond molecular ionisation", Physical Chemistry Chemical Physics, **21**, 17584 (2019).
[2] D. Schwickert et al., "Electronic quantum coherence in glycine molecules probed with ultrashort x-ray pulses in real time", Science Advances, **8** (22), eabn6848 (2022).
[3] T. Driver et al., "Attosecond Coherent Electron Motion in a Photoionized Aromatic Molecule", arXiv:2411.01700 **[physics.chem-ph]** (2025).

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