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Applications of Attosecond Soft-X-ray pulses to Photoemission Chronoscopy and Transient Absorption

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Attosecond soft-X-ray pulses can nowadays be produced either through high-harmonic generation (HHG) or free electron lasers (FELs). Whereas HHG sources achieve the shortest durations (43 as [1]), FELs achieve the highest peak intensities [2]. I will discuss recent experiments that exploit the complementarity of these attosecond sources. Combining attosecond soft-X-ray pulses from LCLS with circularly polarized infrared pulses, we have measured attosecond photoionization delays of N1s photoemission of a series of aromatic azabenzene molecules (pyridine, pyrazine, s-triazine) [3]. We have observed a systematic increase of the photoionization delays with increasing number of electronegative nitrogen atoms and with increasing symmetry of the molecular scaffold. Taking advantage of the excellent timing stability of HHG-based attosecond pulses, we have observed the decoherence and revival of charge migration in neutral silane molecules and the transfer of electronic coherence through conical intersections [4]. Exploiting the broad bandwidth of HHG-based sources, we have observed a charge-directed proton-transfer reaction in ionized urea solutions [5]. These experiments highlight the complementarity of HHG- and FEL-based sources and suggest promising perspectives for attosecond science.

References:

- [1] T. Gaumnitz et al., Opt. Exp. 2017, <https://doi.org/10.1364/OE.25.027506>
- [2] J. Duris et al., Nat. Photon. 2020, <https://doi.org/10.1038/s41566-019-0549-5>
- [3] J.-B. Ji, Z. Guo et al., arxiv <https://doi.org/10.48550/arXiv.2402.17685>
- [4] D. Matselyukh et al., Nat. Phys. 2022, <https://doi.org/10.1038/s41567-022-01690-0>
- [5] Z. Yin et al., Nature 2023, <https://doi.org/10.1038/s41586-023-06182-6>

Footnotes

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