

Experimental Demonstration of Two-Color Attosecond Pump–Probe Spectroscopy with an X-ray Free-Electron Laser

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Pump–probe experiments with attosecond resolution are the key to understanding electronic dynamics in quantum systems. Isolated attosecond soft X-ray pulses produced with a Free-Electron Laser (FEL) have pulse energies up to hundreds of microjoules [1]. The intense pulse energies of attosecond soft X-ray FEL pulses are sufficient for nonlinear X-ray spectroscopy and enable attosecond pump-probe experiments. In this talk, I will present the generation and control of sub-femtosecond pulse pairs from a two-color X-ray FEL [2]. By measuring the delay between the two pulses using an angular streaking diagnostic, we characterize the group velocity of the X-ray FEL and demonstrate the control of the pulse delay down to 270 as. We demonstrate the applicability of this technique to a pump–probe measurement in core-ionized para-aminophenol. These results reveal the ability to perform pump–probe experiments with sub-femtosecond resolution and atomic site specificity.

The highly flexible design of the Athos soft X-ray beamline at SwissFEL enables the generation of isolated attosecond pulses [3] and holds the potential for two-color X-ray attosecond pump-probe techniques. We are currently working on designing an angular streaking instrument at the Maloja endstation to diagnose isolated attosecond X-ray pulses and to further enable attosecond soft X-ray pump-probe measurements at SwissFEL.

REFERENCES

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- [2] Guo, Z., et al. Nature Photonics (2024): 1-7.
- [3] Prat, E., et al. APL Photonics 8.11 (2023).

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