X-ray free-electron lasers (XFELs) have heralded the regime of high-intensity coherent X-ray radiation for time-resolved measurements of atomic and molecular systems during chemical interactions involving inner-shell electronic states and single-shot studies on non-reproducible systems, including X-ray diffraction and imaging experiments.

I will show how photoelectron streaking spectroscopy has been established as a route to bridge the gap between well-controlled lab-based X-ray sources and high-brightness XFELs for the investigation of dynamics on the natural time scale of the motion of electrons. The novel concept of angular-resolved streaking relies on exciting photoelectrons in a gaseous target with the XFEL pulse in the presence of a near-circularly polarized infrared field and angle-resolved detection of the dressed photoelectrons with a multi-time-of-flight spectrometer. The experimental results provide direct information about the full time–energy distribution of the X-ray pulses with attosecond resolution on a single-shot basis, including X-ray pulse duration, polarization, intensity substructure, chirp and potentially the phase. Thus, the specific capabilities of LCLS-II are expanded to X-ray coherence measurements with attosecond temporal resolution, exemplified on the idea of probing charge migration in-situ on complex bio-molecules.