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Beyond Pump–Probe: Exploiting Temporal Correlations for Time-Resolved X-Ray Photoelectron Spectroscopy FELIX BRAUSSE, MARIO BORGWARDT, JOHANNES MAHL, MATTHEW FRAUND, Lawrence Berkeley National Laboratory, FRIEDRICH ROTH, TU Bergakademie Freiberg, WOLFGANG EBERHARDT, Center for Free-Electron Laser Science / DESY Hamburg, OLIVER GESSNER, Lawrence Berkeley National Laboratory — Time-resolved x-ray photoelectron spectroscopy (tr-XPS) is a well-established tool for studying the temporal evolution of excited-state dynamics of atoms, molecules, and solids. The technique has been enabled by the latest generation of bright, short-pulse x-ray light sources. Here, we present recent progress in developing a tr-XPS technique that does not rely on traditional pump–probe settings, but instead exploits the temporal correlations between photoelectrons generated by different pulses of the x-ray pulse train. The point-in-time properties of a molecular ensemble in chemical equilibrium statistically fluctuate around a mean (expectation) value. These fluctuations are encoded in the time-dependent photoelectron signal, and its temporal autocorrelation can reveal the underlying, time-invariant quantities, like diffusion constants and reaction rates. We demonstrate that, in a traditional pump–probe experiment, auto- and cross-correlations of the time-stamped photoelectron signal stream provide access to much of the same dynamics information as the pump–probe spectra. The results represent an important milestone toward the long-term goal of developing XPS techniques capable of determining the microscopic, short-term processes that underlie dynamic equilibria.

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