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Time-resolved photoemission spectroscopy at free-electron lasers

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Free-electron lasers (FELs) are currently the only ultrashort-pulsed photon sources that can provide sufficient photon flux for time-resolved pump-probe solid-state photoemission measurements in the soft and hard x-ray regime. In principle, FELs enable powerful combinations of photoemission techniques, augmented by femtosecond time resolution, in a single experiment. We particularly envision a pump-probe photoemission experiment in which the electronic structure dynamics in energy-momentum space, the chemical dynamics at atomic sites, and the local structural dynamics around atomic sites are simultaneously tracked at the fundamental timescales of electronic and atomic motion by combined time-resolved ARPES, XPS, and XPD. Intriguingly, when hard x-ray FEL radiation is used, the nonequilibrium electron and lattice dynamics can be probed even in the bulk of materials or at buried interfaces. This is especially useful in complex materials or device-like structures where the complete ultrafast photoemission movie provides direct dynamical information on, e.g., the couplings between electronic and structural degrees of freedom or the interfacial carrier dynamics.
Here, we give a brief overview of the current status of time-resolved photoemission at FELs [1-5]. Specifically, we present results obtained from complex materials such as 1T-TaS2, YbInCu4, VO2, and SrTiO3 at the soft x-ray FEL FLASH and the hard x-ray FEL SACLA, respectively. The results demonstrate the practical viability and possible wider impact of FEL-based time-resolved photoemission, but they also illustrate the need for high-repetition-rate FELs such as FLASH and the European XFEL.

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