Photoemission Orbital Tomography: Imaging Molecular Wave Functions in Reciprocal and Real Space

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The photoemission orbital tomography (POT) technique, a variant of angle-resolved photoemission spectroscopy, has been very useful in the characterization of the electronic properties of molecular films. It is a combined experimental and theoretical approach that is based on the interpretation of the photoelectron angular distribution in terms of a one-electron initial state. This includes the unambiguous assignment of emissions to specific molecular orbitals, their reconstruction to real space orbitals in two and three dimensions, the deconvolution of complex spectra into individual orbital contributions beyond the limits of energy resolution, the extraction of detailed geometric information such as molecular orientations, twists and bends, the precise description of the charge balance and transfer at interface, the detection of momentum-selective hybridization with the substrate, and the tracing of orbital images on ultrafast time scales, to name only a few examples. In its simplest form, POT relies on the plane-wave approximation for the final state. While this works surprisingly well in many cases, this approximation does have its limitations, most notably for small molecules and with respect to the photon-energy dependence of the photoemission intensity. Regarding the latter, a straightforward extension of the plane wave final state leads to a much-improved description while preserving the simple and intuitive connection between the photoelectron distribution and the initial state.