From Density Functional Theory to Orbital Cinema: Imaging Molecular Electronic States at Surfaces

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Understanding how molecular electronic states emerge, hybridize, and evolve at solid interfaces is central to many problems in nanoscience, from organic electronics to photocatalysis. In this talk, I will present an overview of my research on the electronic, optical, and structural properties of π-conjugated organic molecules adsorbed on metal surfaces and atomically thin insulating layers, with a particular emphasis on how ab-initio theory can be directly connected to modern photoemission experiments.

In the first part, I will give a brief introduction to the theoretical framework underlying my work, including density functional theory, time-dependent DFT, and many-body perturbation theory (GW and Bethe-Salpeter equation), and discuss how these methods are used to describe charge transfer, level alignment, and excitonic effects at hybrid organic-inorganic interfaces. I will then introduce photoemission orbital tomography (POT), a methodology that allows molecular orbitals to be reconstructed from angle-resolved photoemission data, and outline my role in developing and applying this approach in close collaboration with experimental colleagues.

In the second part, I will highlight selected recent results that illustrate how POT provides new physical insight into interface phenomena. These include the microscopic origin of circular dichroism in photoemission from achiral molecules on surfaces, and the extension of POT to the time domain, where femtosecond pump-probe experiments allow the imaging of transient excited states and excitons. In this context, I will discuss results from the ERC Synergy project Orbital Cinema, which aims to visualize electronic and excitonic dynamics at interfaces on femtosecond time scales.