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Stacking the Deck: Leveraging Surface Interactions to Tune Interfacial Electronic Structure¹ BRET MAUGHAN, CALLEY EADS, University of Arizona, PERCY ZAHL, Brookhaven National Laboratory, PETER SUTTER, University of Nebraska-Lincoln, OLIVER MONTI, University of Arizona — We present results from a series of experiments aimed at understanding and controlling molecular interactions in phthalocyanine (Pc) thin-films on Cu(110) to tailor the interfacial electronic structure. Using low-temperature scanning tunneling microscopy (LT-STM), we identify interactions that drive surface-molecule coupling, molecular self-assembly and thin-film order. We provide evidence that interactions with native Cu adatoms play a pivotal role in self-assembly of Pc systems, along with anisotropic nanoribbon growth dynamics, supported by an agent-based kinetic Monte Carlo (AB-KMC) simulation. We show further that self-assembled nanoribbon length can be controlled using surface diffusion barriers and that ordered 2D thin-film growth is promoted by diminishing surface-molecule interactions that otherwise dominate native Cu(110) interfaces. Altogether, this detailed structural understanding allows us to interpret interfacial electronic structure and dynamics, uncovered through ultraviolet (UPS) and two-photon photoemission (2PPE) spectroscopy experiments, in molecular configuration-specific detail. In all, our understanding of interfacial processes guides strategic modifications to both surface and molecule to harness interfacial interactions and thereby modify the collective electronic structure of the interface.

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