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Electron Injection to Control Self-Assembly and Disassembly of Phenylacetylene on Gold[1] ARTHUR P. BADDORF, QING LI, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, CHENGBO HAN, J. BERNHOLC, Department of Physics, North Carolina State University, HUMBERTO TERRONES, BOBBY SUMPTER, MIGUEL FUENTES-CABRERA, JIEYU YI, ZHENG GAI, PETER MAKSYMOVYCH, MINGHU PAN, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory — The power of two-dimensional organic molecular systems for applications including electronics, functionalization and nanolithography is enabled by our ability to produce structures through self-assembly on a surface. Unfortunately, relying on thermal fluctuations to drive the surface attachment reactions has limited self-assembled molecules (SAMs) to little beyond alkanethiols on gold. We demonstrate a seminal example of non-thermal control over molecular self-assembly, where hot-electron injection rather than thermal fluctuations transform a disordered layer of weakly bonded hydrocarbon molecules into an ordered, dense monolayer. The process is reversible, in that injection of holes reverts to a disordered state. Since electron and hole injection is accomplished with a STM, unprecedented local control over ordered and disordered domains is achieved. STM imaging and correlated density functional calculations reveal that ordered domains consist of molecules vertically aligned and more strongly attached to the gold substrate through the acetylene tail, while disordered domains contain weakly bound molecules lying flat.

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