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Electron-induced Three Dimensional Self-assembly and Disassembly of Molecules on a Gold Surface QING LI, Oak Ridge National Laboratory, CHENGBO HAN, North Carolina State University, MIGUEL FUENTES-CABRERA, HUMBERTO TERRONES, BOBBY SUMPTER, Oak Ridge National Laboratory, JERRY BERNHOLC, North Carolina State University, JIEYU YI, ZHENG GAI, ART BADDORF, PETRO MAKSYMOVYCH, MINGHU PAN, Oak Ridge National Laboratory — The immensely successful methodology of molecular self-assembly on surfaces has produced thousands of new applications and paved ways to new research areas, such as molecular electronics and the dip-pen nanolithography. Here we demonstrate a seminal example of non-thermal control over molecular self-assembly, where hot-electrons transform a largely disordered layer of hydrocarbon molecules, into a highly ordered, densely packed and three-dimensional monolayer on a gold surface. Subsequently, hot-electron/hot-hole injection can heal the defects within the self-assembled layer, and even entirely and reversibly disassemble it. From a theoretical analysis we have identified that electron-induced processes allow the formation of a very strongly-bonded molecule, and yet it is inaccessible by thermally-activated reactions due to a large number of competing processes. This work thus demonstrates the feasibility of accessing and controlling non-thermal reaction pathways that may lead to unique and controllable order-disorder transitions in supported molecular layers.

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