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Three-Dimensional Imaging of Complex Molecular Electronic States via Atomic Manipulation Reconstruction ERIC CHATTERJEE, DOMINIK RASTAWICKI, ALEX CONTRYMAN, YAN SUN, DYLAN REUTER, HARI MANOHARAN, Stanford Univ — We describe a method based on STM atomic manipulation for experimentally capturing the complete three-dimensional electronic structure of complex molecules. Using techniques we have recently developed for assembling molecular graphene and related materials, we vary specific site potentials, intersite hopping amplitudes, and Fermi energy in 2D nanostructures which when wrapped into a 3D container represent a new probe molecule. Here we present the design methods and analyses of a number of such molecules, focusing on those containing rotational symmetry. We show how various types of fullerenes, of interest due to their electronic and vibrational properties, can be unwrapped on a 2D surface, reprogrammed, and rewrapped to 3D. Examples of the unwrapping methods include cutting selected bonds in order to sever adjacent faces or sites. Analysis of the local density of states for 2D correspondents to fullerenes yields the presence of peaks at the highest occupied molecular orbital and lowest unoccupied molecular orbital, with an energy gap between these levels. The replication of these properties of fullerenes in 2D space serves as evidence of the significant potential of STM assembly and spectroscopy in studying the applicability of exotic 3D molecules in electronics.

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