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Giant spatially-resolved self-assembled donor-acceptor molecular heterojunctions¹ JEFFREY R. GUEST, Center for Nanoscale Materials, Argonne National Laboratory, Argonne, IL, USA, JOSEPH A. SMERDON, Jeremiah Horrocks Institute of Mathematics, Physics and Astronomy, University of Central Lancashire, Preston, UK, NOEL C. GIEBINK, Department of Electrical Engineering, The Pennsylvania State University, University Park, PA, USA, NATHAN P. GUISINGER, PIERRE DARANCET, Center for Nanoscale Materials, Argonne National Laboratory, Argonne, IL, USA — Despite theoretical models predicting that rectification ratios (RR) > 1000 should be achievable in molecular rectifiers, demonstrations of this have been rare. It has also been extremely challenging to unravel the structure-function relationships on the nanometer length scales that determine their behavior. Using scanning tunneling microscopy (STM) and spectroscopy (STS), we show that RRs >1000 at biases <500 mV are realized in the two-molecule limit for self-assembled donor-acceptor bilayers of pentacene on C_{60} on Cu. We show that the system behaves as a molecular analog to a Schottky diode due to strong electronic coupling of C_{60} to the metallic substrate, and electronic transport is dominated by sequential tunneling from semiconducting pentacene to metallic C_{60} . Furthermore, we demonstrate the extreme sensitivity of the low-bias I(V) characteristics to the molecularly-resolved structure of the heterojunction (HJ), which leads to negative differential resistance and $\sim 100 \times$ variation in the rectification ratio within 2 nm of the edge of the molecular HJ.

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