Supplementary Information for

Two-Dimensional van der Waals Supramolecular Frameworks from Co-hosted Molecular Assembly and C_{60} Dimerization

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Initial growth of C_{60} 2D crystal

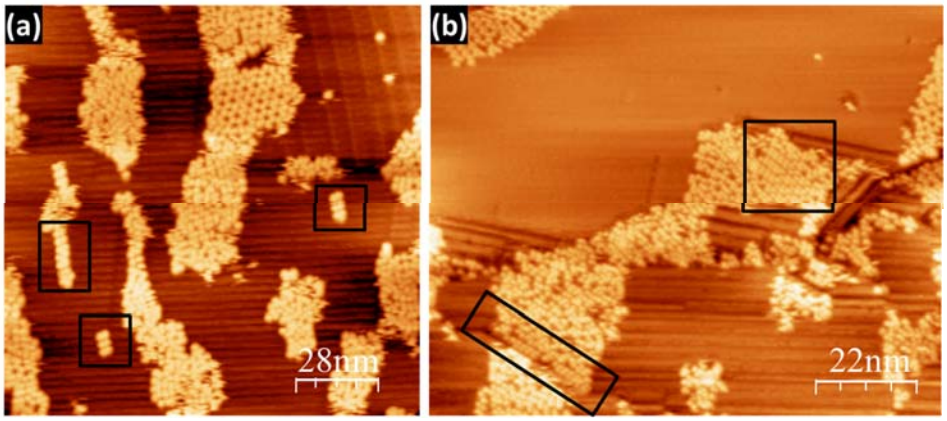

Figure S1. The initial growth of C_{60} 2D crystal at RT. (a) V_{bias}=1.2 V, I_t=300 pA and image size: 85 nm; (b) V_{bias}=-1.5 V, I_t=50 pA and image size: 90 nm × 110 nm; the areas marked with black rectangles show the appearance of C_{60} pairs.

The stability of C_{60} 2D crystal

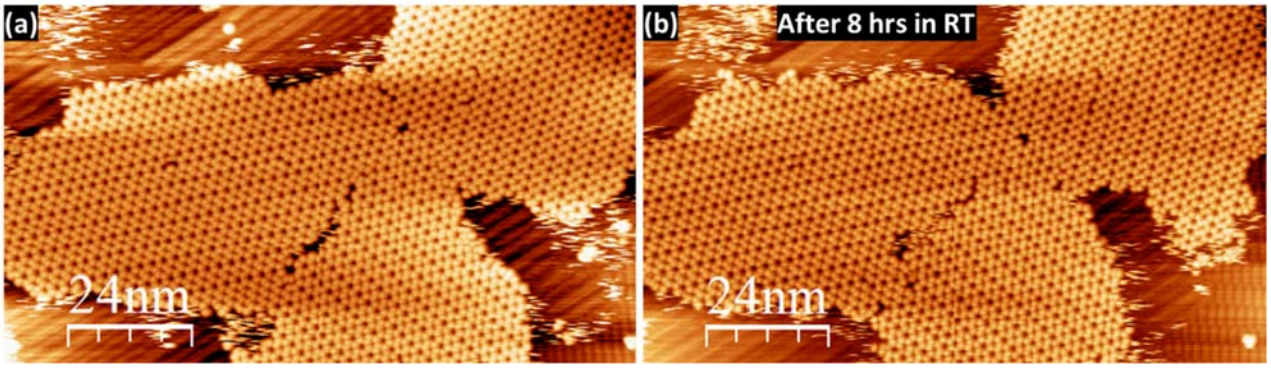

Figure S2. The stability of C_{60} 2D crystal at RT. (a) an original patch of C_{60} 2D crystal; (b) after staying at RT about 8 hours; both images are taken with V_{bias}=1.0 V, I_t=100 pA.

Further depositing C_{60} molecules onto the porous C_{60} framework

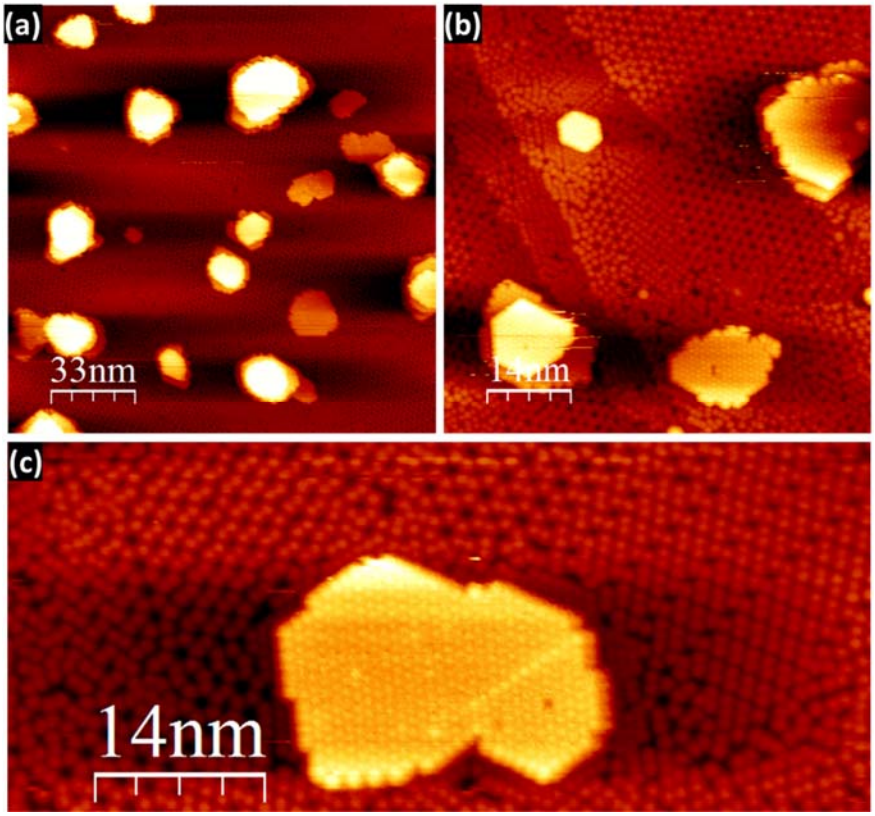

Figure S3. Further depositing C_{60} molecules onto the porous C_{60} framework. (a) V_{bias}=1.8 V, I_t=300 pA and image size: 165 nm; (b) V_{bias}=1.5 V, I_t=300 pA and image size: 70 nm; (c) High resolution image shows both close-packed C_{60} molecules on multilayers island and porous C_{60} framework in background, V_{bias}=1.8 V, I_t=300 pA.

STM images taken at 77 K with various biases

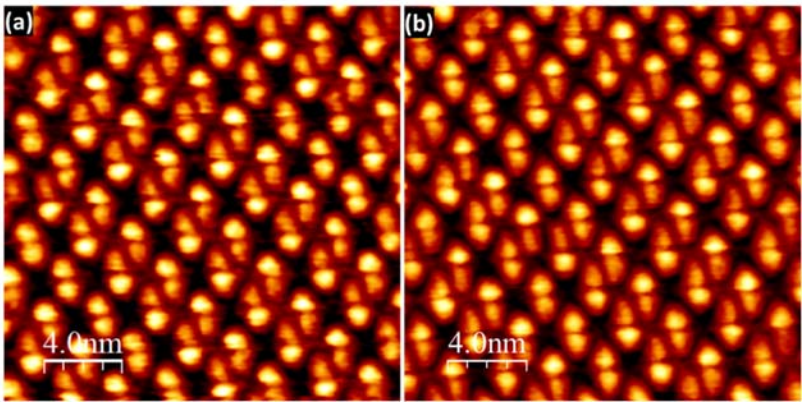

Figure S4. STM images taken at 77 K with various biases. (a) V_{bias}=-0.3 V, I_t=200 pA and image size: 20×20 nm^2; (b) V_{bias}=+0.3 V, I_t=200 pA; both images shows a feature of one bright and one dark for the C_{60} dimers.

Optimized structure of two C_{60} molecules, calculated isosurfaces and STM simulations

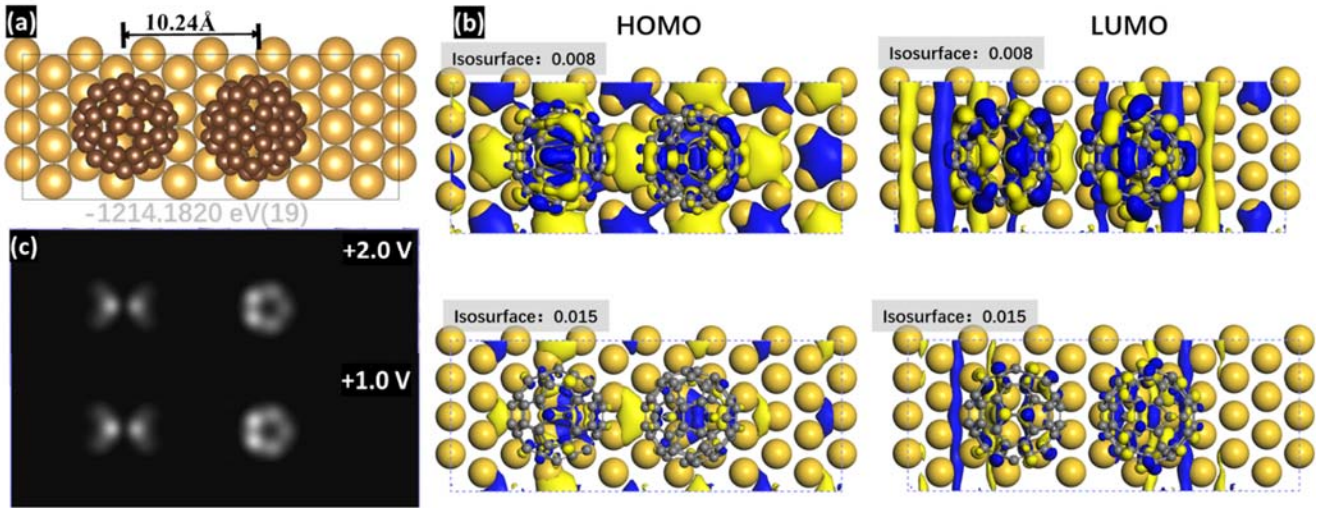

Figure S5. (a) A ball-stick model shows an optimized structure of two C_{60} molecules with a distance of 10.24 Å. (b) The calculated isosurfaces of 0.008 and 0.015 for the HOMO and LUMO orbitals of two C_{60}, which show no charge-transfer in between. (c)The simulated STM images for such C_{60} dimer with the biases of +2.0 V (upper) and +1.0 V (lower).