SUPPORTING INFORMATION

Detailed experimental procedures are described.

Large area scans (**Figure S1**) show the overall morphology of the surface following decoration by a C_{60} coverage controlled to create C_{60} chains one atom wide along step edges. From such large area images, specific features can be localized under the STM tip, and higher-resolution images can be made, as shown in enhanced color in **Figure S2**. **Figure S2** shows both decorated adatom islands and vacancies. Sequential scans of an entire closed ring, such as **Figure S2c**, are used to create movies (**S3**) of the structural fluctuations of the entire ring structure.

EXPERIMENTAL METHOD

Ag(111) thin film preparation procedure has been described previously.[15, 32, 33] In a UHV chamber (base pressure $< 3 \times 10^{-11}$ torr), Ag(111) thin films are Ar-sputtered and annealed. After many cycles, surfaces become atomically clean, as confirmed by Auger, LEED and STM. Under proper annealing conditions, monolayer Ag islands are generated on the clean Ag(111) films. The size of islands ranges from several nm to more than 100 nm. The distributions of island size and density are sensitively related to the annealing conditions.

C_{60} is evaporated from powder in a PBN crucible with the clean Ag(111) film held at room temperature. Before deposition, we carefully degas the evaporator and the C_{60} containing crucible, so that during deposition, the pressure in the chamber never exceeds

1.0×10^{-10} torr. The deposition rate is ~ 0.01 ML/min. By controlling deposition time, the C_{60} coverage varies from only few C_{60} molecules on the whole surface to C_{60} molecules to fully covering steps and island edges. After deposition, the samples are transferred onto the STM stage in the same chamber.

The typical tunneling conditions are 0.07 to 0.11 nA at −1.55 to −1.75 V. To investigate the decorated island shape fluctuations, we employ relatively fast STM scans over individual islands to obtain ensembles of topography images at consecutive time. The fastest scan speed is 13.1 s per 256 × 256 pixel image and 26.2 s per 512 × 512 pixel image, respectively. Each ensemble contains 50 to several hundred sequential images. Via image processing methods, the position of each C_{60} molecule can be extracted, and the island shapes are digitized by the positions of all C_{60} molecules in the chains. The typical size of the decorated islands we investigated is from 20 nm to 50 nm in diameter. In our measurements, we find that the decorated islands in this size range do not have observable net motion across the surface of the film. For smaller decorated islands typically with size less than 10 nm, we do see island decay, in which the surrounding C_{60} rings shrink by coalescence, e.g. in which the total number of C_{60} molecules does not change.

SUPPORTING FIGURES AND MOVIE

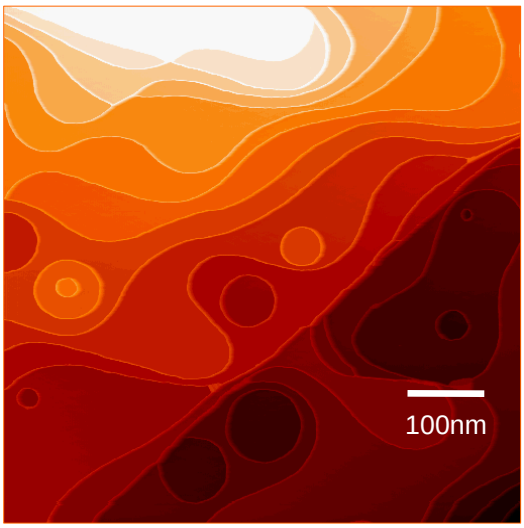

Figure S1 STM topography image of Ag thin film morphology decorated by single-molecule wide C_{60} chains. The tunneling conditions are $U_{sample} = -1.71$ V and $I_t = 86$ pA.

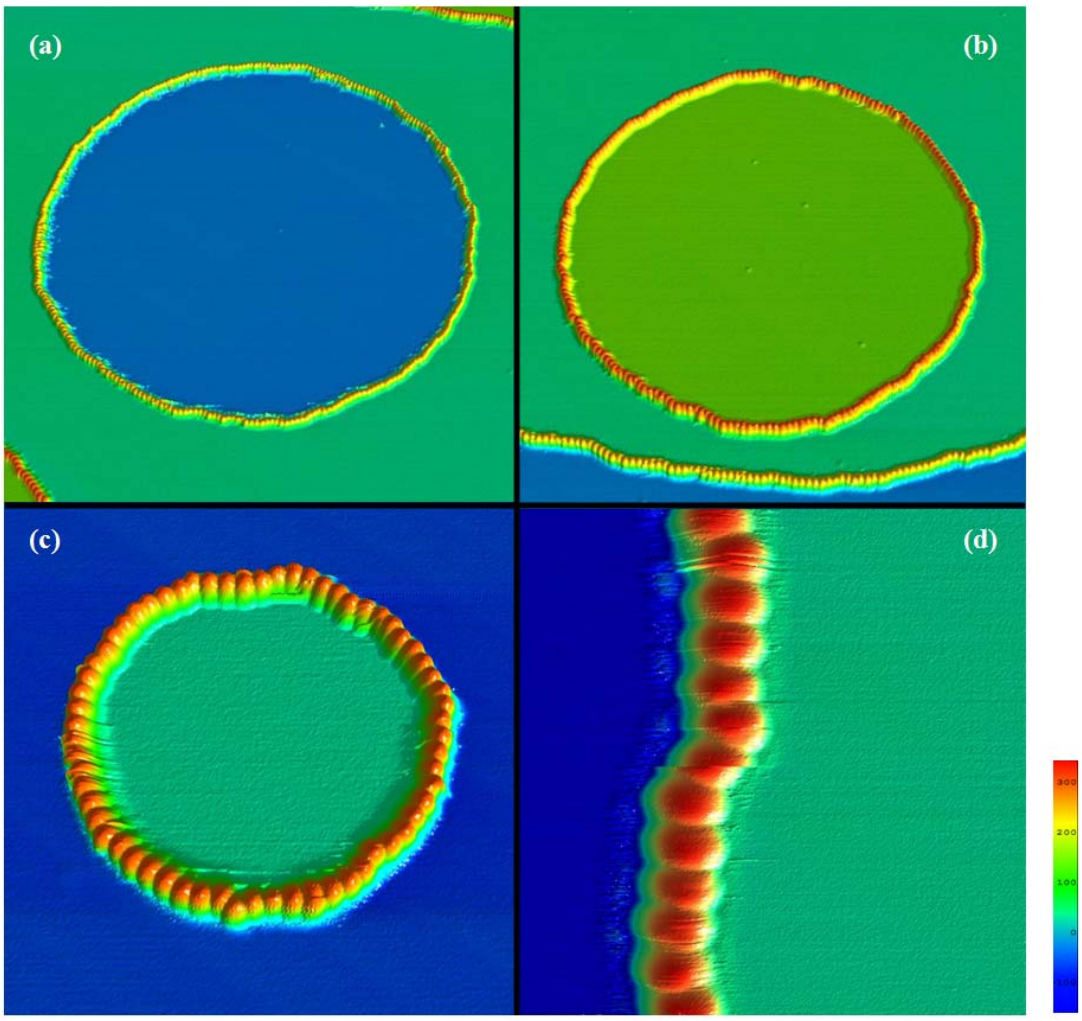

Figure S2 STM images (3D view) of different types and size scales of decorated islands. The color bar indicates height, which differs slightly in offset and scale for each image. **(a) (upper left)** large vacancy island decorated by a C_{60} ring (island diameter ~ 96 nm), **(b) (upper right)** large adatom island decorated by a C_{60} ring (island diameter ~ 72 nm), **(c) (lower left)** small adatom island decorated by a C_{60} ring (with 64 C_{60} molecules, diameter ~ 21 nm), **(d) (lower right)** close up view of an edge segment. The horizontal streaks and the "split" molecule near the center of the image indicate hopping of C_{60} molecules between sequential STM line scans.

Movie S3 A movie of a STM image ensemble including 66 sequential images with time interval 52.4 s. Images are as shown in **Fig. 2b**, in which the blue dots represent the STM topography image of C_{60} molecules, and the orange dots are the digitized C_{60} positions.