Supporting Information

In situ observations of fullerene fusion and ejection in carbon nanotubes

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S 1 Experimental procedure

S 1.1 SWNT synthesis

The starting SWNTs were synthesized via a laser evaporation route, and prior to the filling process they were opened by annealing in air at 653 K for 60 min.[1] This oxidation process also removes any amorphous carbon present in the sample, as discussed in [2]. The catalyst particles, in our as-synthesized SWNTs sample, are observed to be encapsulated with graphitic matter. No acid treatment was used to remove catalyst particles; this helped to minimize any damage or surface functionalization of the SWNTs. The peapods were then prepared by the hot vapor filling method.[3] High purity fullerenes (MER Corp) were mixed with the opened SWNT and then heated in a sealed quartz tube under high vacuum (ca. 10^{-7} hPa) at 723 K for 7 days. After the filling step, the peapods were annealed in dynamic high vacuum (ca. 10^{-6} hPa) at 1598 K for 90 min. This annealing process converts the peapods into DWNT. At the same time most excess fullerenes remaining in the sample, which have not incorporated within SWNT, sublime out of the sample.

S 1.2 HREM imaging and simulations

The time-sequence in-situ HREM imaging was performed using a third order aberration corrected TITAN[3] transmission electron microscope at 80kV. The HREM simulations were performed using JEMS software.[4] The chirality of the tubes was determined by analyzing the fast Fourier transformed TEM images and measuring the corrected SWNT diameter in the manner described by Sato et al.[5] The TEM imaging parameters used for simulated HREM images were obtained from the Cs-correction measurements during experimental HREM imaging. (Electron beam current: $1x10^{-2}$ - $1x10^{-1}$ pA/nm^2, coefficient of spherical aberration (Cs): 0.001 mm).

S 1.3 HREM post image processing method

The post image processing was done using fast fourier transformation (FFT) filtering of each individual frame followed by inverse FFT to remove unwanted noise and slightly enhance the contrast of each image frame. The band pass mask of Gatan® Digital Micrograph™ software was used for this process. The region of interest was cropped from each image processed individual frame and compiled into movie using iMovie HD software.

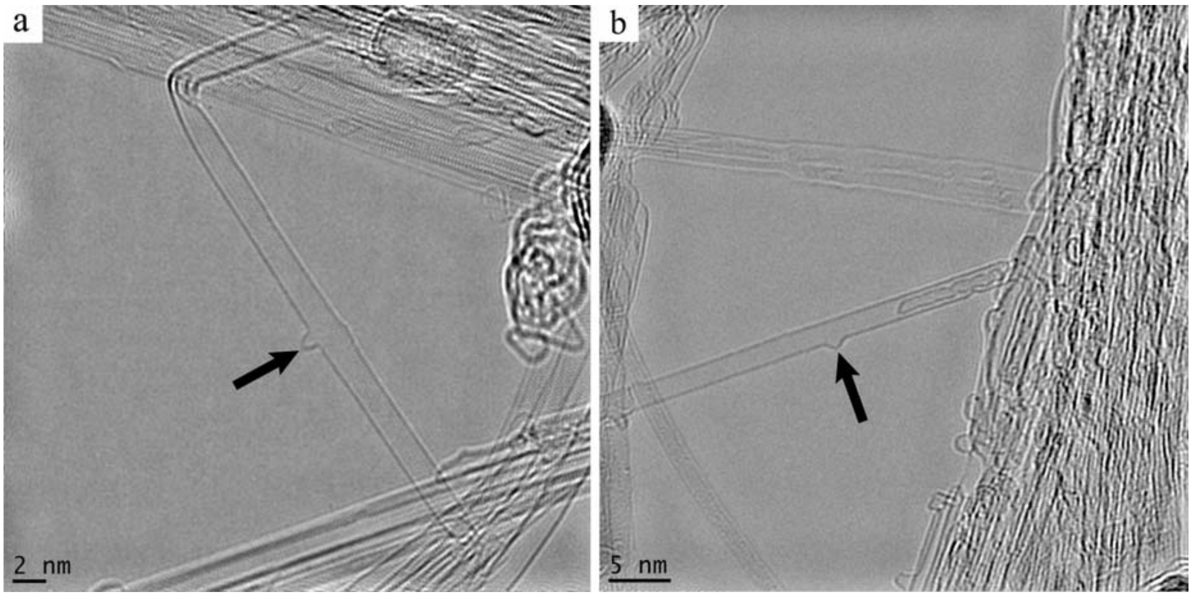

Figure S1. Overview bright field TEM images of humped-SWNTs observed in our as-annealed SWNT samples. The arrows show the nanohump geometry of covalently fused C_{60} fullerene with outer sidewall of SWNT.

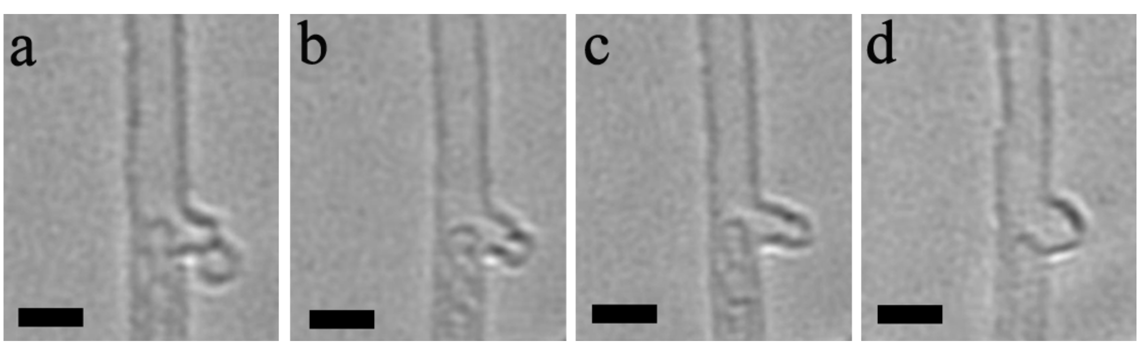

Figure S2. In situ HREM observations of a C_{60} fullerene molecule interacting and covalently fusing with a hSWNT. The scale bar is 2 nm.

S 2 HREM time-sequence movie "Movie_S1.mpg"

In-situ HREM image series movie, showing the interaction dynamics leading to the formation of humped-SWNT. The duration of each frame in the movie is reduced to 1/30 of a second. The real-time interval between individual images used in this movie, during the TEM image recording, is 1 second.

S 3 HREM time-sequence movie "Movie_S2.mpg"

In this movie we observe the covalent coalescence of nanohump with an additionally adsorbed C_{60} fullerene. The duration of each frame in the movie is reduced to 1/30 of a second. The real-time interval between individual images used in this movie, during the TEM image recording, is 1 second.

S 4 HREM time-sequence movie "Movie_S3.mpg"

In this movie we show the *in-situ* HREM observations of electron beam irradiation driven morphing-entry of the fullerene inside the host SWNT. The duration of each frame in the movie is reduced to 1/30 of a second. The real-time interval between individual images used in this movie, during the TEM image recording, is 1 second.

References:

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