Supporting Information

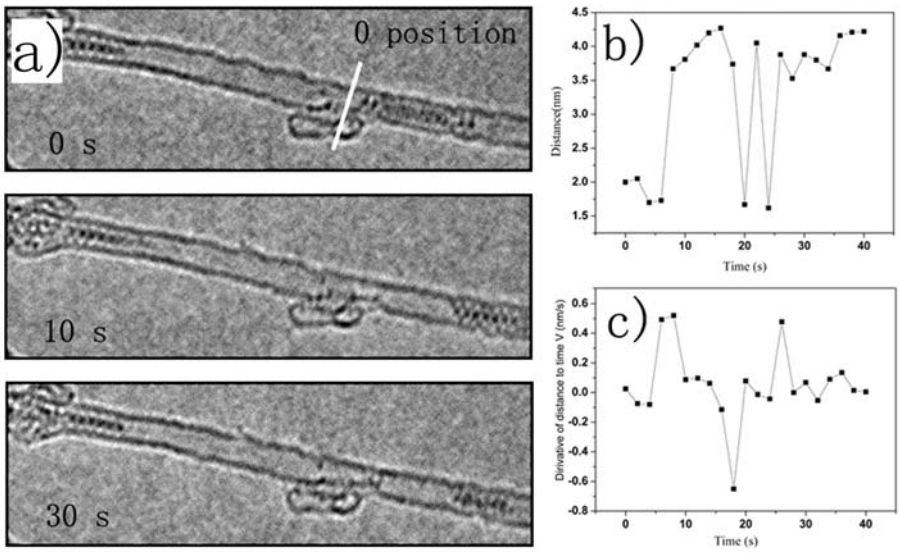

Fig. S1

The iodine atoms inside SWNTs are not immobile but occasionally move inside the tube. The movement of iodine atoms is ascribed to the electron irradiation-induced displacements Figure S1a shows sequential HRTEM images of a single iodine chain inside a SWNT with the diameter of ~1 nm. (See Movie 1 for movie file of iodine atoms moving inside the SWNT). The distance from the center of iodine atomic chain to the initial position (shown in figure S1a) versus the observation time are shown in figure S1b. Fig. S1c shows the relative displacement per observation sequence. The movement of iodine atoms is ascribed to the electron irradiation-induced displacement during the observation. The movement of iodine atoms could be blocked at a defect or maybe some contamination of SWNT. This phenomenon may have the implications for the utilization of the defects or contaminations as hurdles to control the movements of the atom chains.

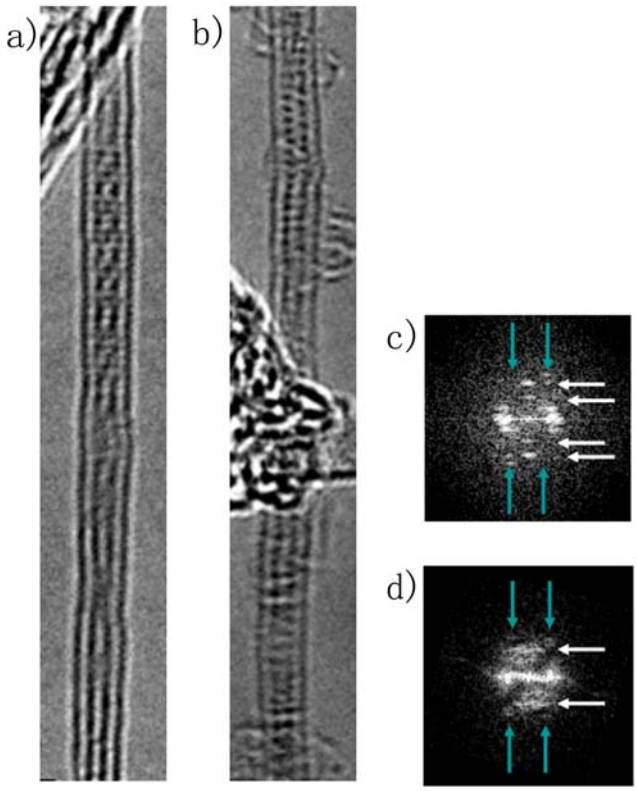

Fig. S2 shows HRTEM images of double helical structures of iodine atomic wires taken by an aberration corrected TEM. As one can see, the helical pitch is not constant even in a single SWNT and can vary between 1.4 nm and 5 nm in Fig. S2a. From the FFT analyses of the HR-TEM images (Fig. S2c and S2d), we can estimate the inter-atomic distance of iodine atom as 0.29-0.30 nm (indicated by blue arrows) which differs from the bulk iodine crystal (0.27 nm in orthorhombic structure). We were unable to correlate the nanotube chiral angle (indicated white arrows) with the iodine helical pitch in the FFT images.

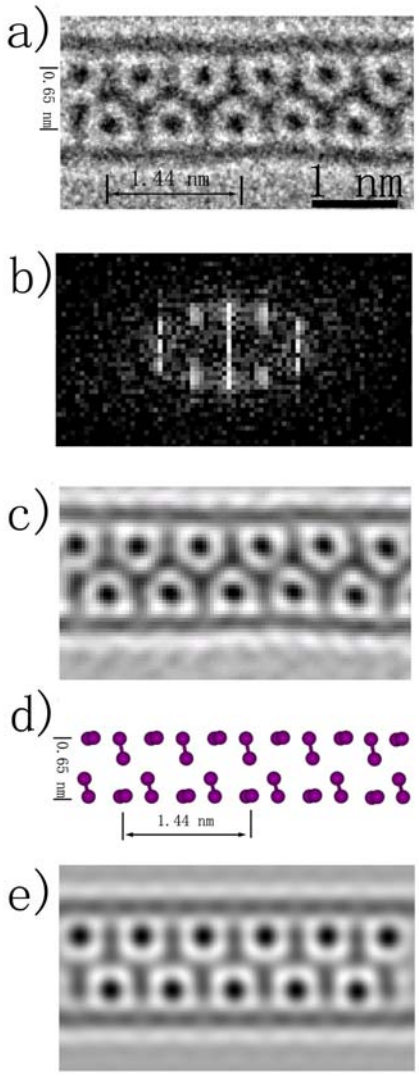

Figure S3. shows the iodine crystal confined in the SWNTs with slight different diameters. (a) HRTEM images of iodine crystal confined in a SWNT of 1.45 ± 0.05 nm in diameter (b) The FFT pattern of the images. (c) Reconstructed inverse FFT images. (c) The schematic atomic model for the iodine crystal. The iodine-iodine distance in the dimer was set to 0.29 nm (d) The simulated images based on the crystal model encapsulated in a SWNT

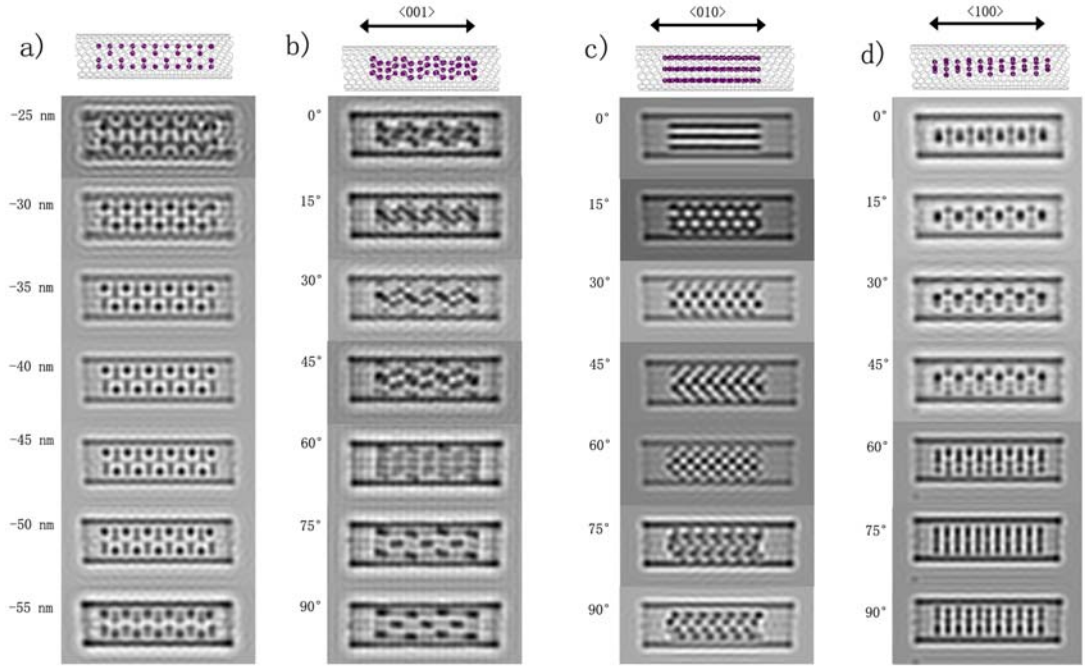

Figure S4

Systematic HR-TEM image simulations were performed to corroborate the observed HR-TEM images (Figure 3). Over 300 HR-TEM image simulations (all not shown) for any possible structures with different defocus and orientational conditions were made to carefully compare with the observed image. Apart from the fig S4(a) showing our proposed crystal structure (a new phase), no other image simulation based on the bulk crystal structures S4(b)-(d) was able to reproduce the experimental HR-TEM image. The bulk structure with the <100> along the tube axis gives a similar lattice constant as 1.44nm (S4(d)), the structure does not reproduce well the ladder shaped contrast in the experiment. The image simulation of the structure (a) with -40 nm defocus obviously shows the best accordance with the experiment.