Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Ultra-stretchable Conductors Based on Buckled Super-aligned Carbon Nanotube Films

Yang Yu,[b] Shu Luo,[b] Li Sun,[a] Yang Wu,[a] Kaili Jiang,[a] Qunqing Li,[a] Jiaping Wang,*[a] and Shoushan Fan[b]

[a] *Department of Physics and Tsinghua-Foxconn Nanotechnology Research Center, Tsinghua University, Beijing 100084, China*
Collaborative Innovation Center of Quantum Matter, Beijing 100084, China
**E-mail: jpwang@tsinghua.edu.cn*

[b] *Department of Physics and Tsinghua-Foxconn Nanotechnology Research Center*
School of Materials Science and Engineering
Tsinghua University, Beijing 100084, China

I. Supplemental Figures

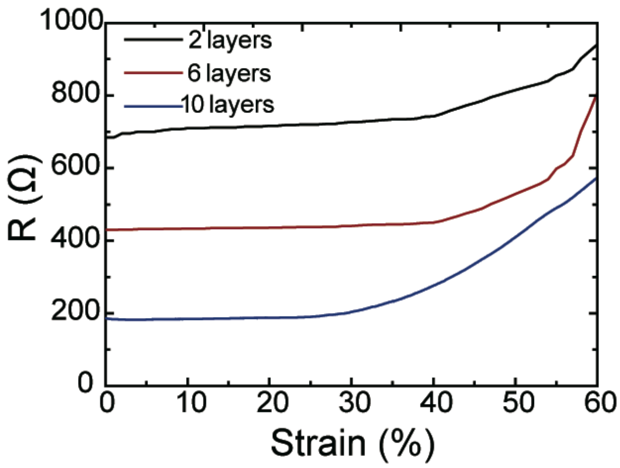

Figure S1. Resistances of the 40% pre-strained SACNT/PDMS conductors with 2, 6, and 10 layers of SACNT films as a function of the applied tensile strain.

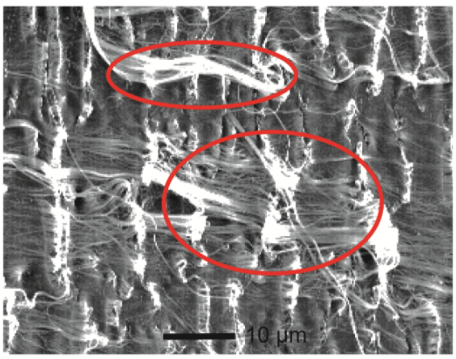

Figure S2. SEM image of the 40% pre-strained 10 layers SACNT/PDMS film after a loading-unloading cycle at 40% strain, with detached CNT bundles marked by the red circles.

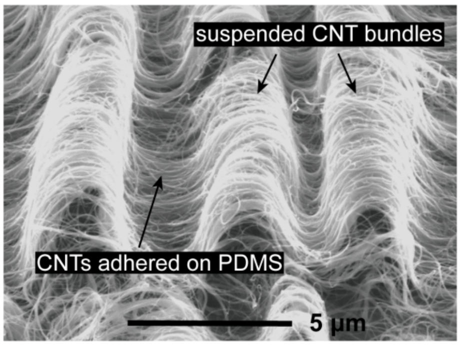

Figure S3. SEM image of a pre-strained parallel SACNT/PDMS film, showing the wavy CNT structures formed on the PDMS substrate.

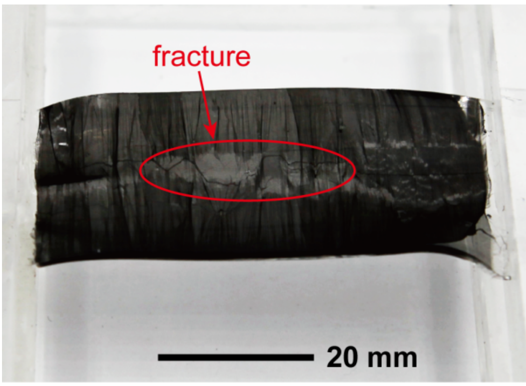

Figure S4. Photograph of a SACNT/PDMS film made with 6 layers of cross-stacked SACNT films and 60% pre-strain.

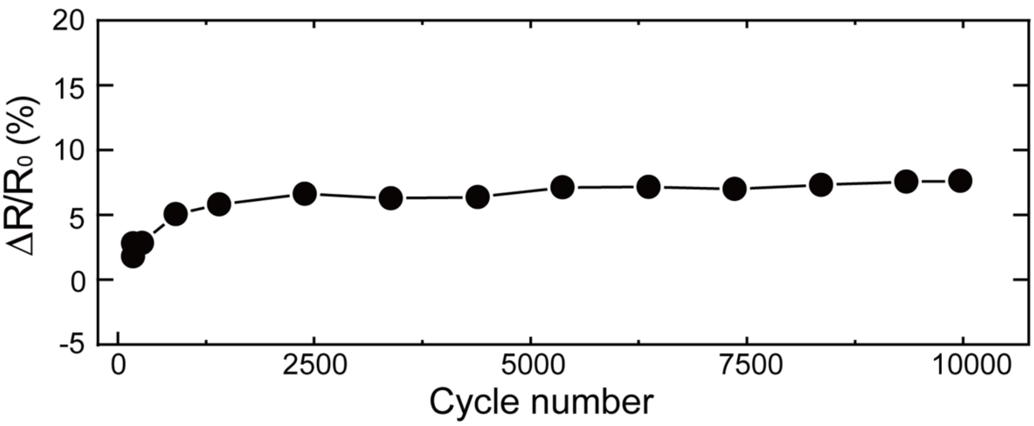

Figure S5. Normalized resistance changes of a cross-stacked SACNT/PDMS film with 40% pre-strain during 10,000 cycles at 40% strain.

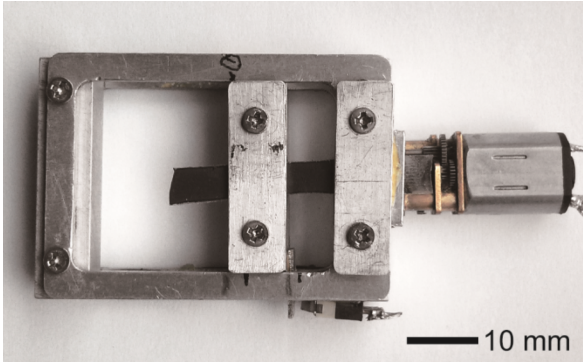

Figure S6. A lab-designed tensile module to stretch the SACNT/PDMS films within the SEM chamber for *in situ* morphology observation.

II. Experimental Details

Preparation of the SACNT film and the PDMS substrate:

SACNT arrays with a diameter of 20–30 nm and a height of 300 μm were synthesized on silicon wafers by chemical vapor deposition with acetylene as the precursor and iron as the catalyst. Details of the synthesis method can be found in previous papers.[18, 19] Continuous SACNT films were directly drawn from the SACNT arrays by an end-to-end joining mechanism. The alignment of CNTs in the film was parallel to the drawing direction. The PDMS substrate with thickness of 0.6 mm was prepared with Sylgard 184 (Dow Corning) by mixing the base and the curing agent at a weight ratio of 10:1. The mixture was cured at 70 °C for 1 h. After cooling down to room temperature, the PDMS substrate was cut into sizes of 70 mm × 20 mm.

Fabrication of the buckled SACNT/PDMS film conductor:

Firstly, the PDMS substrate was uniaxially stretched with various strains of 10%, 20%, 30%, 40%, 100%, 150%, and 200%. Controlled layers of uni-directional or cross-stacked SACNT films (denoted as "||" or "⊥" CNT films) were coated on the surface of the pre-strained PDMS substrate. Ethanol was then dropped onto the PDMS substrate. SACNT films shrank and tightly contacted with the pre-strained PDMS substrate upon the evaporation of ethanol.

Afterwards, the PDMS substrate was released to its original length and the SACNT film formed buckled structures on the PDMS substrate.

In situ SEM observation:

The morphologies of the SACNT/PDMS films were examined by scanning electron microscopy (SEM, Sirion 200, FEI). A lab-designed tensile module (Figure S6) was integrated within the SEM chamber to stretch the SACNT/PDMS samples with 20%, 40%, 60%, and 80% strains for in-situ morphology observation. The gauge length was 5 mm and the strain rate was 10% s^{-1}.

Characterization:

The resistance of the SACNT/PDMS conductor under applied strain up to 200% was characterized. The dimension of the sample was 70 mm × 10 mm × 0.6 mm. Two Cu foils were attached at both ends of the sample by conductive silver paste for resistance measurement. Tensile strain was applied using an Instron 5848 microtester with a gauge length of 20 mm and the resistance of the SACNT/PDMS sample was monitored by a Keithley 2400 Source Meter. The durability of the SACNT/PDMS conductor was characterized by performing cyclic tensile tests with 150% strain for 10,000 cycles and the resistance of the sample was measured at each stretching cycle. The morphologies of the SACNT/PDMS film were further examined by SEM before and after the cyclic tensile testing.