Supplementary Information

Interfacial Thermal Resistance and Thermal Rectification in Carbon Nanotubes Film-Copper System

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1. **Transmissivity Measurement of the CNT film**

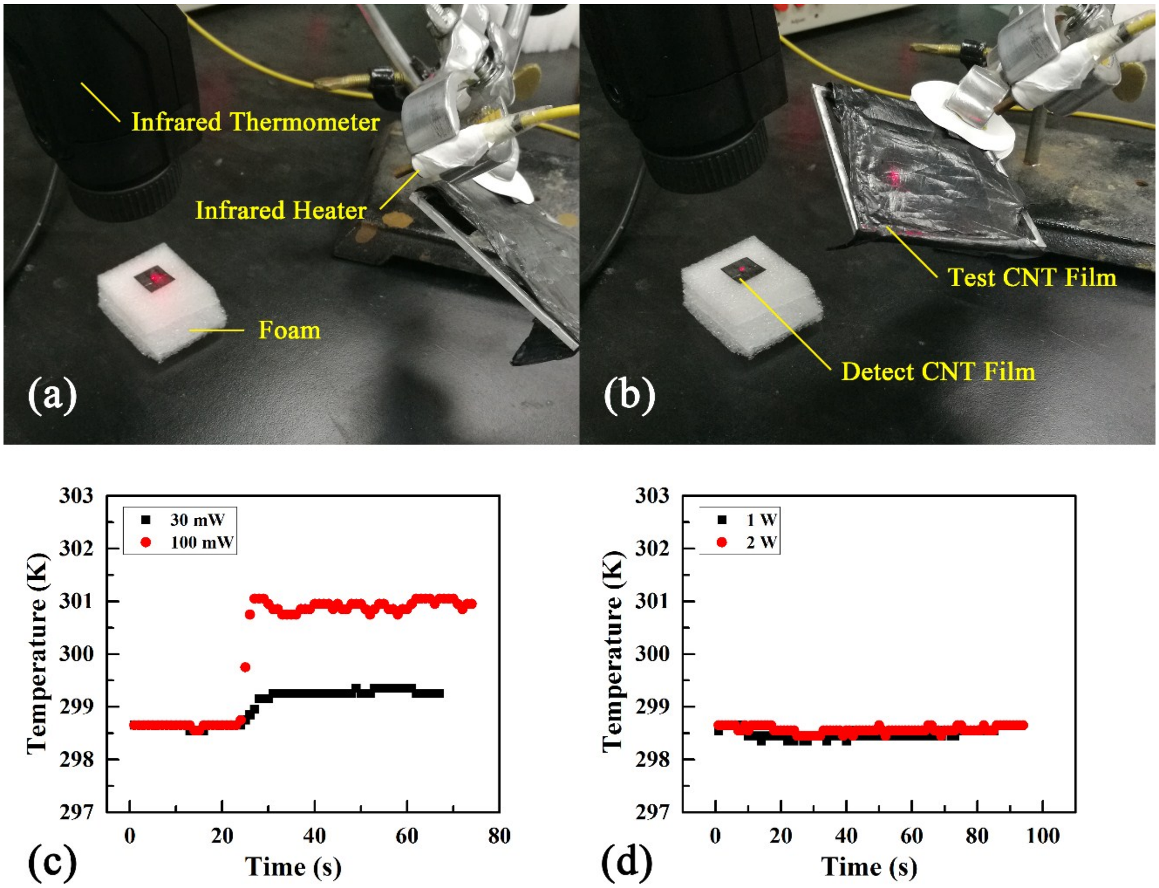

Fig. S1 Digital Photographs of the system to measure the transmissivity of the CNTs (a and b) and the results (c and d).

We experimentally confirmed the transmissivity of the CNT film being zero. First, we utilized the infrared laser to heat the CNT film directly (Fig. R1 (a)). The reason we employ the CNT film as detector rather than copper film is that the CNT film possesses a much higher emissivity than copper, so the CNT film is more sensitive to infrared heating. Foam is put under the CNT film as an insulation. An infrared thermometer is employed to monitor the temperature of the detect CNT films. The results are shown in Fig. R1 (c). The temperature rose even though the power of infrared laser was set as 30 mW, which is the minimum power of the device. The temperature of the detector rose remarkably when the power was set as 100 mW. Then we put a 1000-layer super-aligned CNT film between the infrared heater and the detector (Fig. R1 (b)). Within the precision of the temperature monitor, no temperature increase was observed neither the power of the infrared laser was 1 W nor 2 W (Fig. R1 (d)). So we believe it is reasonable to conclude that the CNT films are opaque and the transmissivity is zero.

2. Conductivity Measurements of CNT films

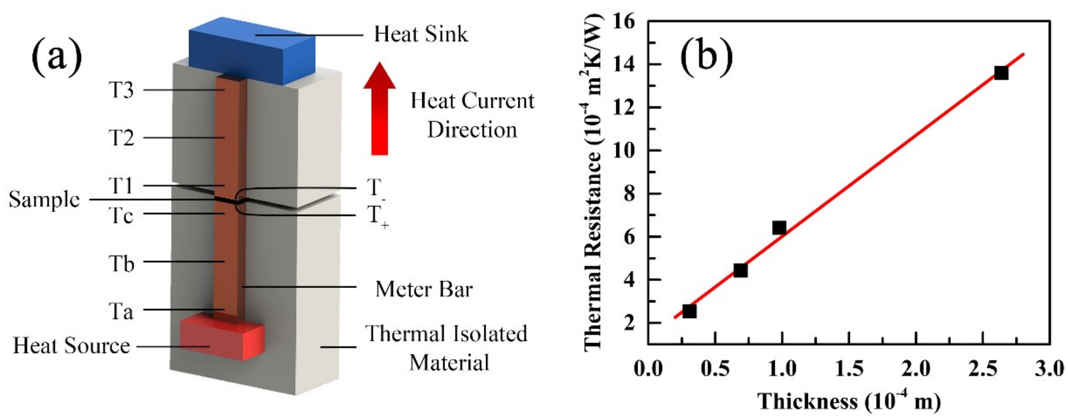

Figure S2. (a) Schematic of the system for thermal conductivity measurement of CNT films. (b) Thermal resistance versus thickness of CNT films obtained under a pressure of 1.1 MPa with optimal fit (red).

Shown in Figure. S2(a) is the schematic of the apparatus for CNT films conductivity measurement, which is consistent with ASTM (American Society of Testing Materials) D5470 standard. The samples were set between two copper bars with a constant pressure. No thermal grease was glued on the surfaces of the CNT films because the softness of the samples naturally leads to well contact. Heat flows from bottom to top and the temperatures are monitored at six different locations (Ta-Tc, T1-T3). The heat flux can be obtained as (Fourier's Law)

$$J = Sk_m \frac{Ta - Tc}{xa - xc} \tag{S1}$$

where S and k_m are the cross section area and thermal conductivity of the meter bar respectively, xi (i represents a-c, 1-3, or + -) is the location of a specific point. Then we can obtain $T+$ by extrapolation

$$T_+ = Tc - \frac{xc - x_+}{xa - xc}(Ta - Tc) \tag{S2}$$

and $T-$ could be obtained in a similar way. The total thermal resistance obtained from system contains two parts, one of which is the intrinsic resistance of CTN films (R_{CNT}), and the other one is interfacial thermal resistance of CNT films with two contacted surfaces (R'_{int}). With the help of Eq. S1 and Eq. S2, we can obtain

$$R_{total} = R'_{int} + \frac{l}{k_{CNT}} = \frac{T_+ - T_-}{J/S} \tag{S3}$$

where l and k_{CNT} are the thickness and conductivity of the CNT films, respectively.

Figure S2(b) shows the thermal resistance as a function of thickness of CNTs with the linear fit, yielding thermal conductivity of 0.213 W/m·K and the summation of ITRs in two direction of 1.32×10^{-4} m^2K/W.

3. Uncertainty Analysis

The error bars shown in Fig. 5 consist of random error, the uncertainties induced from the resolution of the device, and the uncertainty induce from the thermal leakage of the system.

We can obtain the uncertainty induced from the resolution of the device with the help of Eq. (2), which shows that

$$R_{int} = \frac{\Delta T_{int}}{j} = \left(\frac{\Delta T_{OA}}{\Delta T_{Cu}} - 0.04\right)\frac{l_{Cu}}{k_{Cu}} - \frac{l_{CNT}}{k_{CNT-r}} \tag{2}$$

The second term on the right hand side of the Eq. (2) is a constant. The temperature resolutions of the thermal imager and thermocouple are 0.08 K and 0.1 K, respectively. The uncertainty of the thickness of the CNT film caused by the resolution of the micrometer is 0.005 mm. If there is a function $N=f(A, B, C\ldots)$, the standard uncertainty transfer equation can be expressed as

$$\Delta_N = \sqrt{(\frac{\partial f}{\partial A})^2\Delta_A^2 + (\frac{\partial f}{\partial B})^2\Delta_B^2 + (\frac{\partial f}{\partial C})^2\Delta_C^2 + \ldots} \tag{S4}$$

We can substitute Eq. (2) into Eq. (S4) and obtain

$$\Delta_{device} = \frac{l_{Cu}}{k_{Cu}}\sqrt{(\frac{1}{T_{AB}})^2\Delta_{T_O}^2 + (\frac{T_{OB}}{T_{AB}^2})^2\Delta_{T_A}^2 + (\frac{T_{OA}}{T_{AB}^2})^2\Delta_{T_B}^2 + (\frac{k_{Cu}}{l_{Cu}k_{CNT}})^2\Delta_{l_{CNT}}^2} \tag{S5}$$

With the similar method, the uncertainty caused by thermal leakage could be expressed as

$$\Delta_{leakage} = \frac{l_{Cu}\Delta T_{OA}}{k_{Cu}\Delta T_{Cu}}\Delta_{T_{Cu}} \tag{S6}$$

where Δ_{TCu} could be estimated as

$$\Delta_{T_{Cu}} = [1 - (\frac{\Delta T_{CD}}{\Delta T_{AB}})^{\frac{1}{3}}] \tag{S7}$$

Then, the total uncertainty could be expressed as

$$\Delta_{total} = \sqrt{\Delta_{random}^2 + \Delta_{device}^2 + \Delta_{leakage}^2} \tag{S8}$$