A Demo Opto-electronic Power Source Based on Single-Walled Carbon Nanotube Sheets

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ABSTRACT It is known that single-walled carbon nanotubes (SWNTs) strongly absorb light, especially in the near-infrared (NIR) region, and convert it into heat. In fact, SWNTs also have considerable ability to convert heat into electricity. In this work, we show that SWNT sheets made from as-grown SWNT arrays display a large positive thermoelectric coefficient (p-type). We designed a simple SWNT device to convert illuminating NIR light directly into a notable voltage output, which was verified by experimental tests. Furthermore, by a simple functionalization step, the p- to n-type transition was conveniently achieved for the SWNT sheets. By integrating p- and n-type elements in series, we constructed a novel NIR opto-electronic power source, which outputs a large voltage that sums over the output of every single element. Additionally, the output of the demo device has shown a good linear relationship with NIR light power density, favorable for IR sensors.

KEYWORDS: single-walled carbon nanotube · opto-electronic · thermoelectric · power source · near-infrared light

Single-walled carbon nanotubes (SWNTs) were previously reported to strongly absorb light, especially in the near-infrared (NIR) region, and convert it into heat.[1−5] The absorption coefficient is extremely high (10^4 to 10^5 cm^{-1} extending from the ultraviolet to the far-infrared region), at least 1 order of magnitude greater than that of mercury−cadmium−telluride, the most popular photoconductor for 2D arrays of IR photodetectors.[2] A forest of vertically aligned SWNTs behaves very similarly to a blackbody.[3] The enormous heat emitted from SWNTs when absorbing NIR light can even be utilized to kill cancer cells.[4,5] On the other hand, we accidently found that SWNT sheets by vacuum-filtrating as-grown SWNT arrays have a large positive thermoelectric coefficient (~70 μV/K), indicating a strong capability to convert heat into electricity. Therefore, we consider that, by illuminating one end of such a SWNT sheet strip using NIR light, there will be a large temperature difference between the two ends and thus will be a considerable voltage output. This is the initial step and the basis of the work presented in this article.

Further, we find that the as-prepared SWNT sheets simply functionalized by polyethyleneimine (PEI) display a large negative thermoelectric coefficient (~−87 μV/K), that is, an inversion from p-type to n-type. When the above p-type NIR light-induced thermoelectric elements were imitated, n-type elements could also be constructed. By integrating p- and n-type SWNT elements in series, we propose a novel NIR opto-electronic power source, which outputs a large voltage that sums over the output of every single element. We have fabricated such a demo device using 50 pairs of p- and n-type elements, and the test results have satisfactorily met our expectations. Additionally, the output of the demo device has shown a good linear relationship with the NIR light power density. This work also provides a new mechanism to design infrared opto-electronic devices, which may have application potentials in various areas such as telecommunication, remote sensing, thermal photovoltaics, and solar cells.[6]

The raw SWNT materials are chemical vapor deposition (CVD)-grown SWNT arrays. Their diameters and lengths are around 2 nm and 0.36 mm, respectively. Detailed processes of the fabrication of as-prepared SWNT sheet (p-type) and PEI-coated SWNT sheet (n-type) are given in the Methods section. Scanning electron microscopy images of the surfaces of the two types of SWNT sheets are shown in Figure 1. From both images, we can see that such sheets are continuous and exhibit an excellent uniformity. PEI is uniformly coated on the SWNT bundles and distributed on the sheet (see Figure 1b). The thickness of the PEI-coated sheet is around 25 μm, a little thicker than that of the as-prepared sheet (~20 μm), implying that PEI penetrated the sheet well.

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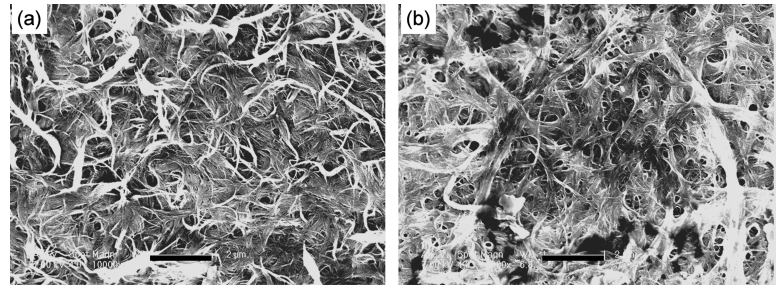

Figure 1. Scanning electron microscopy images of the surfaces of as-prepared (a) and PEI-coated (b) SWNT sheets. Scale bars: 2 μm.

The experimental setup and the results of NIR light-induced thermoelectric test of as-prepared SWNT sheets are shown in Figure 2. A sheet strip with a size of 16 × 2 mm^2 (length × width) was suspended over two supporting blocks (Figure 2a). Electrical contacts to the sample sheet were made with silver paste. The NIR light with a wavelength of 980 nm was provided by a direct diode laser system. The diameter of the illuminated area was around 6 mm. The output voltage was read by a Keithley 2182 nanovoltmeter, and the temperature data were collected by an Optris LS infrared thermometer. The diameter of the temperature recording point is *ca.* 0.8 mm, and the edge of the point is very close to the electrode (<0.2 mm).

The light power density dependence of the output voltage ($V_{AB} = V_A - V_B$) is plotted in Figure 2b. This simple device successfully converts NIR light directly into a voltage output. The output shows a good linear relationship with NIR power density, indicating that SWNT sheets are a good candidate for IR sensors, although the magnitude of the output is relatively small (sensitivity ~12.6 μV/(mW/cm^2)). In the following, we will see that this value can be amplified by integrating the opto-electronic elements. When the light was illuminating, the temperature of both ends was recorded (Figure 2c). The temperature of the illuminated end increases linearly with the light power density, while the unilluminated end remains at room temperature (19.0 °C). According to the calculation formula of thermal conductivity, we estimate that, under 90 mW/cm^2 illumination, the temperature difference between the two ends may exceed 100 °C. Therefore, this seemingly

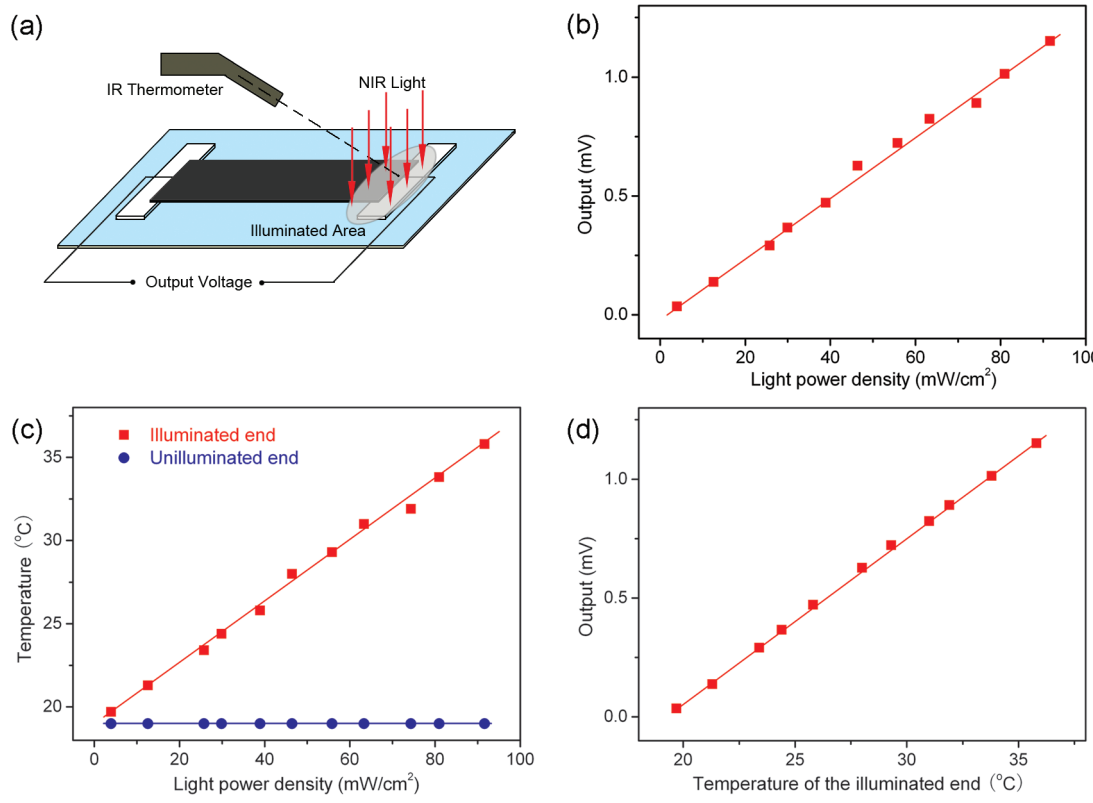

Figure 2. Experimental setup (a) and the results (b−d) of NIR light-induced thermoelectric test of as-prepared SWNT sheet. (b) Light power density dependence of the output voltage (V_{AB}). (c) Light power density dependence of the temperature of the two ends when one end was illuminated by NIR light. (d) Plot of the output voltage *versus* the temperature of the illuminated end.

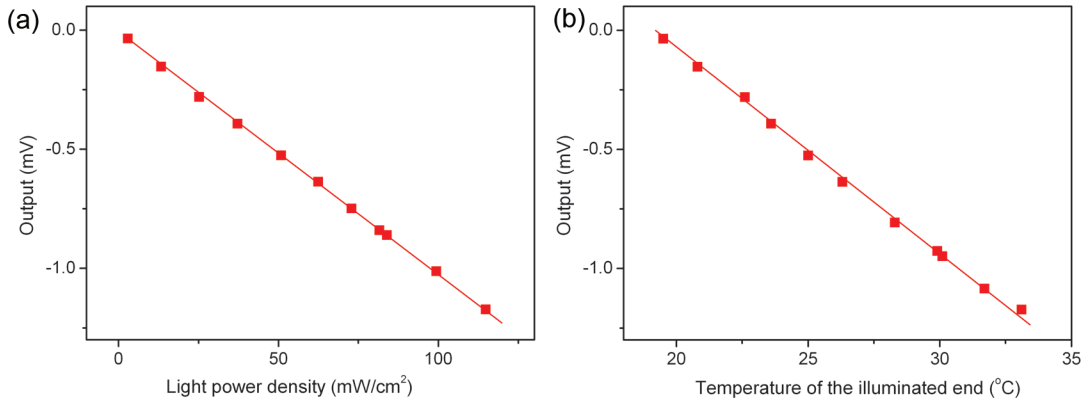

Figure 3. (a) Light power density dependence of the output voltage (V_{AB}) of a PEI-coated SWNT sheet strip. (b) Plot of the output voltage *versus* the temperature of the illuminated end of the PEI−SWNT strip.

peculiar result is in fact natural. Additionally, the supporting frame of the unilluminated end acts as sort of a heat sink, which conducts and accommodates the limited heat. The thermoelectric coefficient is determined by $S = \Delta V/\Delta T$, where ΔT is the temperature difference between the two ends. The output voltage shows a good linear relationship with the temperature of the illuminated end (Figure 2d), which means the NIR light-induced S can be deemed as a constant in this temperature range. From the slope, we calculate it to be 70.2 μV/K. This value is in excellent agreement with the thermoelectric coefficient obtained by directly heating one end of the sheet strip without the use of NIR light (~70 μV/K). The thermoelectric coefficient of our sheet is generally larger than that reported previously,[7−11] which is mainly ascribed to the loose contacts between the individual SWNTs and SWNT bundles[9] (see Figure 1a). Additionally, we intentionally left the Fe catalyst nanoparticles in the sheet during the fabrication, although the amount was small. Therefore, the remnant Fe particles might also contribute to the large thermoelectric coefficient known as Kondo effect in the one-dimensional system.[8] The results presented in Figure 2 validate the feasibility of our proposition at the beginning that, by illuminating one end of a strip of SWNT sheet, there would be a large temperature difference between the two ends and thus would be a considerable voltage output.

In recent years, the origin of the photoresponse of carbon nanotube (CNT) films or CNT−polymer composites in both visible and NIR regions has been under much controversy in terms of whether the photoresponse is due to photon-induced charge carriers (excitonic)[12−17] or due to heating of the CNT networks (bolometric).[2,16−19] For instance, Itkis *et al.* detected a notable photoconductivity for a large-area SWNT film suspended in vacuum under very low NIR light power densities, which was attributed to heating of the SWNT network.[2] Pradhan *et al.* have also observed a large NIR photoconductivity in CNT−polymer composites and attributed it to a combination of thermal and excitonic effects together with the thermal fluctuation-induced tunneling model.[16,17] Lien *et al.* reported that the photoresponse maxima occurred when the (green) light illuminated the SWNT film−electrode contacts and ascribed it to photoexcitation at the tube−metal junction.[12] St-Antoine *et al.* carefully studied the laser position dependence of the photoresponse of a suspended SWNT film and found that the photoresponse maximum was located along the trenches and, therefore, away from the film−electrode contacts.[18] They demonstrated that the origin of the photoresponse in suspended films was a photothermoelectric effect and linked to non-uniform doping.[18] From the NIR light-induced thermoelectric coefficient in this work, we infer that the NIR photoresponse in our sheet mainly originates from heating of the SWNT network. Despite these investigations, studies on practical applications of the photoresponse in SWNT-related materials have not been reported yet.

The crucial step toward the integrated, large-output opto-electronic device is the fabrication of n-type SWNT sheets with large negative thermoelectric coefficients. Previous studies revealed that SWNTs can be functionalized by amine-rich polymers such as PEI to switch their operation from p-type to n-type irreversibly.[20] Up to now, the application of this functionalization is limited to field-effect transistors.[20−22] Here, we report that PEI-coated SWNT sheets display a large negative thermoelectric coefficient. The experimental setup for the NIR light-induced thermoelectric test of PEI-coated SWNT sheets is identical to that for the as-prepared sheet (Figure 2a). The light power density dependence of the output voltage (V_{AB}) is plotted in Figure 3a, implying an n-type characteristic. Meanwhile, the temperature of both ends was also measured. Like the results of as-prepared sheet, the temperature of the illuminated end increases linearly with the light power density, while the unilluminated end remains at room temperature. The dependence of the output on the temperature of the illuminated end also shows a good linear relationship (Figure 3b). We calculate the NIR light-induced thermoelectric coefficient to be −87.4 μV/K, an astonishing value which is even larger than that of

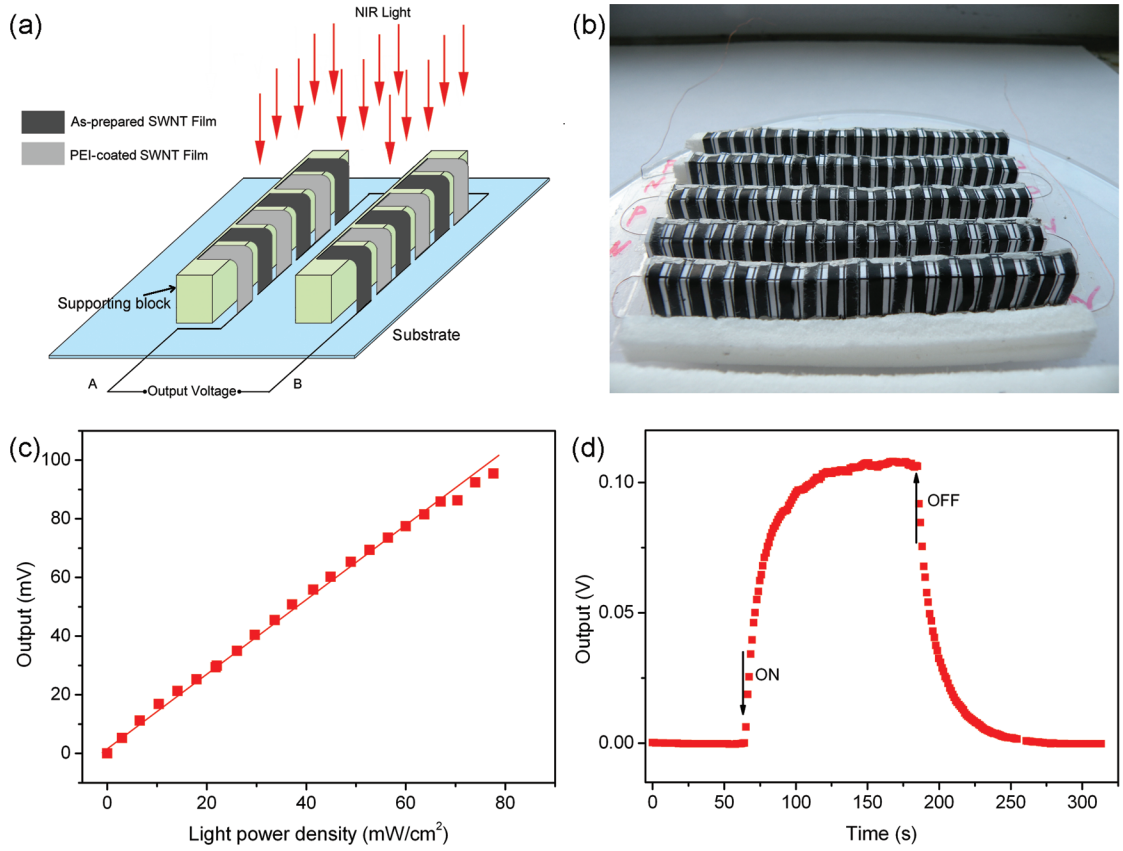

Figure 4. (a) Schematic illustration of an integrated device. In practical experiment, the number and the arrangement of the sheets can be adjusted. (b) Optical photograph of a demo device composed of 50 pairs of p- and n-type elements. (c) Light power density dependence of the output voltage (V_{BA}) of the demo device. (d) Dynamic response of the demo device to NIR illumination (power density ~84.6 mW/cm^2).

the as-prepared sheet in magnitude. This result indicates that PEI-coated SWNT sheets have application potentials in thermoelectric areas, especially when flexibility is required. We note that, under the same NIR light power density, the output of the PEI-coated sheet is a little smaller than that of the as-prepared sheet (sensitivity ~10.2 μV/(mW/cm^2)). This can be ascribed to the relatively larger heat capacity due to the incorporation of PEI in the sheet.

By integrating p- and n-type SWNT elements, we designed a novel opto-electronic power source (Figure 4a) and fabricated a demo one (Figure 4b). The p-type and n-type elements were arranged alternatively and linked with each other in series. The demo device was composed of 50 p-type elements and 50 n-type elements. For an individual p-type (or n-type) element, one end of the sheet strip (proportion in length ~1/4) was exposed to the NIR light, and the rest was curved down to avoid the illumination. The excellent flexibility of SWNT sheets ensured this process. More information about the fabrication of the demo device can be found in the Methods section. To further prevent the upright part of the strips from possible illumination, in practical experiment, we put long printing paper strips over the trenches. The light power density dependence of the output voltage (Note: V_{BA}) is plotted in Figure 4c, showing a good linear relationship. Satisfactorily, the sensitivity of the integrated device approaches 1.30 mV/(mW/cm^2). This value is basically 100 times larger than a single p-type (or n-type) element, which has met our expectation that the overall output sums over that of every single element. The dynamic response of the demo device to NIR illumination (84.6 mW/cm^2) is shown in Figure 4d. The demo power source successfully outputs a macroscopic voltage exceeding 0.1 V. It is reasonable to deduce that, by integrating more elements, a larger output can be obtained. The electric power is also evaluated using $P = V \times I$, where the I value was determined by Ohm's law. The P value is calculated to be on the order of microwatts. The low power is mainly ascribed to the large resistance of the SWNT sheets (~100 Ω for one sheet strip). A direct way to improve P is to optimize the sheet structure, for example, by reducing the strip lengths, to reduce the resistance. Another and also the critical way is to improve the thermoelectric performance, for example, *via* eliminating the metallic tubes and/or incorporating other thermoelectric materials.

It is worth mentioning that, under a given power density NIR light, the output voltage is only dependent on the number of the elements. Employing modern industrial technique, we believe that the elements can be

integrated more compactly, that is, a large voltage output from a small illuminated area. It has been reported that the NIR photoresponse of SWNT films can be greatly enhanced by vacuumizing the experimental environment.[2,13] This behavior is interpreted as depressing the thermal coupling between the SWNT films and the ambient air.[2] Here, we also expect an enhancement in the NIR response in our device by reducing the ambient air pressure. Furthermore, reducing the contact area between the sheets and the substrates is also an effective way to improve the response. In short, we believe there is still a large space for the improvement and optimization of the performance of the proposed power source.

The opto-electronic response in the SWNT sheets is intrinsically a thermoelectric effect. Therefore, it is meaningful to discuss their performances as thermoelectric materials. The most valuable parameter that describes a thermoelectric material is figure of merit $ZT = S^2\sigma T/k$, where S, σ, and k are thermoelectric coefficient, electrical conductivity, and thermal conductivity, respectively. For as-prepared SWNT sheets, the electrical conductivity is measured as *ca.* 35 S/cm and the thermal conductivity *ca.* 5 W/m · K. We calculate the figure of merit to be 1.1×10^{-3} at 300 K. This value exceeds the expected figures of merit for pure CNTs ($\sim 10^{-4}$)[23] and compares favorably to those in the previous studies with conjugated polymers ($\sim 1 \times 10^{-3}$),[24,25] yet it is lower than those of typical inorganic semiconductor materials such as silicon ($ZT \sim 0.01$ at 300 K).[26] For PEI-coated sheets, the figure of merit is expected to be even larger due to the enhanced thermoelectric coefficient and depressed thermal conductivity.

In general, the figure of merit is very low for many practical applications. However, there is still some space for the improvement of the thermoelectric performance of the SWNT sheets, for example, *via* eliminating the metallic tubes, and incorporating other thermoelectric materials, as mentioned above. We are expecting some progress in this field. Further, the unique properties of the SWNT sheets rest with NIR conversion and are suitable for large-scale fabrication at low cost. Additionally, the excellent flexibility of the sheets offers great convenience during the fabrication of the device especially when specific structures are desired.

In fact, by studying the working principle of the device, we can see that any material that meets the following requirements can be employed for opto-electronic power sources proposed in this work: (i) strongly absorbs light and converts it into heat; (ii) has a large thermoelectric coefficient; and (iii) has a relatively small specific heat capacity. For SWNTs, the specific heat capacity is around 600 mJ/g · K,[27,28] which ensures a large temperature difference between the illuminated end and the unilluminated one of the SWNT sheet strip.

Recently, St-Antoine *et al.* have reported that visible light (wavelengths at 544 and 685 nm) can also lead to a significant thermal effect.[18] Ajayan *et al.* have observed that fluffy SWNTs can even burn when exposed to a camera flash (white light without the ultraviolet constituents), which means the local temperature has reached 600−700 °C.[29] Therefore, the opto-electronic power source proposed here may also be able to work under visible light; that is, this work provides a new approach to build solar cells.

To summarize, employing the strong capability of absorbing light and converting heat of SWNTs, and the large thermoelectric coefficients of both as-prepared (p-type) and PEI-coated (n-type) SWNT sheets, in this article, we proposed a novel opto-electronic power source. This power source is designed by integrating a large number of these two types of SWNT sheets in series and outputs a large voltage that sums over the output of every single element. We have fabricated such a demo device, and the test results using NIR light have satisfactorily met our expectations. Additionally, the output of the demo device has shown a good linear relationship with the NIR light power density, which is favorable for IR sensors. This type of opto-electronic power source can be conveniently constructed in a large scale. This work provides a new mechanism to design opto-electronic devices.

METHODS

Fabrication of As-Prepared and PEI-Coated SWNT Sheets. Raw SWNT arrays were first detached from the substrates and dispersed in an ethanol solution using an ultrasonic crasher (1000 W, 10 min). Then the suspension was filtered through a microporous membrane with the aid of vacuum, which led to a random packing of SWNTs in the form of a dense sheet. After the remnant ethanol was evaporated, the sheet was peeled off the membrane.

The average molecular weight of PEI used here is around 10 000. The as-prepared SWNT sheet was fully immersed in an ethanol solution of PEI (0.1 g/mL) for over 12 h and then washed with ethanol. After the remnant ethanol was evaporated, the fabrication of PEI-coated SWNT sheet was finished.

Fabrication of the Demo Integrated Device. The large-area as-prepared and PEI-coated SWNT sheets were first cut into a large number of small quadrate strips (length × width = 13 × 2 mm^2). Then 10 pairs of SWNT sheet strips were placed on a printing paper (75 × 17 mm^2) in a queue, where the p-type and n-type strips were arranged alternatively. The separations between neighboring p-type and n-type strips were around 1.6−1.8 mm. Both ends of every single sheet strip were stuck to the printing paper with the aid of double-faced adhesive tape. The strips were linked with each other in series using silver paste. Then one side of the printing paper was laid on a sponge rubber cuboid (length × width × height ~ 75 × 4 × 9 mm^3, preplaced on a polystyrene plate with glue), while the rest was curved down to avoid the illumination. We chose sponge rubber as the substrate because of its excellent adiabaticity. Another 40 pairs of elements were fabricated. Altogether, the demo device composed of 50 pairs of p- and n-type SWNT sheet strips, all of which were linked in series with the aid of silver paste.

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