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Directly measuring of thermal pulse transfer in one-dimensional highly aligned carbon nanotubes

SUBJECT AREAS:

CHARACTERIZATION
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CONDENSED-MATTER PHYSICS

CARBON NANOTUBES AND
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Using a simple and precise instrument system, we directly measured the thermo-physical properties of one-dimensional highly aligned carbon nanotubes (CNTs). A kind of CNT-based macroscopic materials named super aligned carbon nanotube (SACNT) buckypapers was measured in our experiment. We defined a new one-dimensional parameter, the “thermal transfer speed” to characterize the thermal damping mechanisms in the SACNT buckypapers. Our results indicated that the SACNT buckypapers with different densities have obviously different thermal transfer speeds. Furthermore, we found that the thermal transfer speed of high-density SACNT buckypapers may have an obvious damping factor along the CNTs aligned direction. The anisotropic thermal diffusivities of SACNT buckypapers could be calculated by the thermal transfer speeds. The thermal diffusivities obviously increase as the buckypaper-density increases. For parallel SACNT buckypapers, the thermal diffusivity could be as high as $562.2 \pm 55.4 \text{ mm}^2/\text{s}$. The thermal conductivities of these SACNT buckypapers were also calculated by the equation $k = C_p \alpha \rho$.

The macroscopic materials based on carbon nanotubes (CNTs)^{1–6}, such as CNT buckypapers and CNT yarns *et al.*, have been widely studied and are attracting more and more attentions. These CNT-based macroscopic materials are self-supported and inherit the excellent properties of individual CNTs^{2,7,8}. So they could be easily manipulated and will be widely used in the future. The super-aligned carbon nanotube (SACNT) buckypaper^{2,9}, where all individual CNTs are highly aligned, is an outstanding member of the CNT-based macroscopic materials. It has excellent properties of tensile strength, thermal and electric conduction. Hence the SACNT buckypapers have been extensively studied for enormous applications such as transistors, super-capacitors and electrodes on which some novel physical properties were applied^{2,10–12}. The thermo-physical properties, including thermal conductivity and thermal diffusivity, are important factors of the CNT buckypapers and would play critical roles in the performance and stability of those buckypaper-based devices. On the other hand, it is well known that the individual CNTs have very excellent thermal properties^{13–15}, but when they constitute macroscopic materials, the thermal properties would be reduced because of enormous interspaces and interfaces in the macroscopic materials. So far there is few reported experimental result to show exactly how the tube-tube contacts and interspaces between individual CNTs in these CNT-based macroscopic materials affect their thermo-physical properties. In view of this, further studies of the thermo-physical properties will help us sufficiently understand the thermal transfer mechanisms of macroscopic materials based on CNTs. Altogether, it is very important to investigate the thermo-physical properties of the SACNT buckypapers.

The thermo-physical properties of different types of materials have been widely studied by several different measuring methods, such as the laser flash method^{16–18}, the self-heating 3ω method^{16,19,20}, the optical heating and electrical thermal sensing technique^{21–23}, etc. These methods have been developed for many years and are still frequently used to determine the thermo-physical properties of different materials at present. Nevertheless, all of them have some limitations. For example, the laser flash method is based on the pulsed-laser heating. The thermo-physical properties of measured materials could be calculated by the heat diffusion time. Therefore, this method needs superior quality vacuum environment to minimize the heat dissipation to surroundings²⁴. In spite of this, the influence of the surroundings is impossible to be eliminated. It is also very hard to precisely estimate the experimental error arising from the surroundings in the laser flash method. For another example, the self-heating 3ω method measures the 3ω signal in the self-heating materials and uses the resistance difference to determine the thermo-physical properties^{16,21}. But the 3ω technique has to suffer from poor signal to noise ratio and also the various noises in the power source and the environment²¹. In view of this, we set up a simple and



precise instrument system to investigate the thermo-physical properties of SACNT buckypapers in the present work. Our measuring techniques didn't need vacuum environment and were operated in the atmosphere, but the convection and the radiation wouldn't affect our measured results. In comparison, it is a great advantage over other measuring methods.

Results

Sample information. We had fabricated a series of SACNT buckypapers with different densities in our previous work². These SACNT buckypapers had shown very outstanding transport and mechanical properties. The individual CNTs in the SACNT buckypapers are all multiwalled and their diameters are about 10–20 nm. The SACNT buckypapers were produced in two steps, *i.e.*, the preparation of super-aligned CNT films^{2,25} from super-aligned CNT arrays and the pressing process (more details are described in the Methods section and Ref. 2). The super-aligned CNT arrays that

constructed the buckypapers were fabricated by the chemical vapor deposition³. The individual multiwalled CNTs (MWCNTs) in the buckypapers were configured in very well parallel to each other, and they would band together closely when high pressures were exerted (Figure 1d & 1e). In this paper, we used six different SACNT buckypaper samples with density in the range of 0.34–0.93 g/cm³ and thickness in the range of 102–23 μm. The scanning electron microscope (SEM) images of the SACNT buckypapers with different densities 0.45 g/cm³ and 0.81 g/cm³ were shown in the Figure 1d & 1e respectively. The SEM images indicate that individual CNTs in the buckypapers with higher density were configured more closely. The SEM images also show that, when higher pressures were exerted, the SACNT buckypapers surface becomes much smoother and crooks of individual CNTs could be reduced (more images and details were provided in our previous paper Ref. 2). So there are fewer crooks of individual CNTs in higher density SACNT buckypapers.

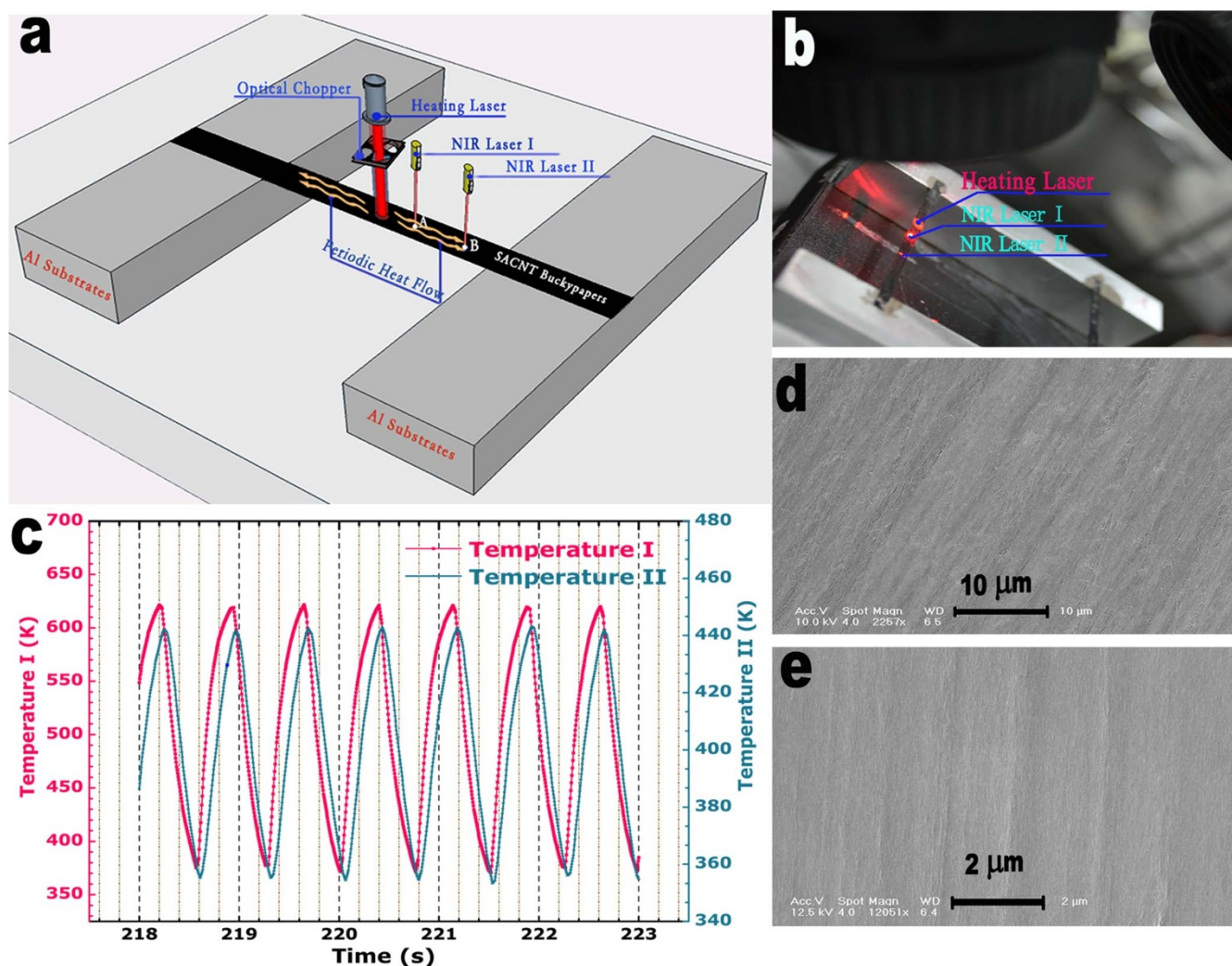


Figure 1 | The measurements of thermo-physical properties of SACNT buckypapers with different densities. (a) Schematic illustration of our measuring method and instrument system. The buckypaper ribbon was pasted on a pair of aluminum substrates by a high-purity silver paste. At the middle point of the suspended ribbon, a heating laser and an optical chopper heated the suspended SACNT buckypaper ribbons by the sinusoidal condition. The NIR Laser I and NIR Laser II were set to measure the temperature distributions at points A & B respectively. (b) The electronic photo of our measurement. (c) The temperature distributions of separated points A & B on the suspended buckypaper ribbon. This is the real-time data of our one measurement. The figure clearly shows that the two temperature waves have obvious time phase differences and different amplitudes. (d) Scanning electron microscope (SEM) images of the SACNT buckypapers with the density 0.45 g/cm³. (e) SEM images of the SACNT buckypapers with the density 0.81 g/cm³. These SEM images show that the higher density buckypaper have a smoother surface, and they also have fewer crooks and interspace between individual CNTs.



The measuring method and instrument system. The measuring method in this paper is based on the one-dimensional periodic heat transfer^{26–29}. That is, according to the one-dimensional Fourier Equation

$$\frac{\partial T(x,t)}{\partial t} = \alpha \frac{\partial^2 T(x,t)}{\partial x^2} \quad (1)$$

When the boundary condition is a sinusoidal heating flux $Asin(\omega t)$, where A is the amplitude and ω is the angular frequency. Then the temperature-solution of the Fourier Equation is

$$T(x,t) = Ae^{-\sqrt{\frac{\omega}{2\alpha}}x} \cos\left(\omega t - \sqrt{\frac{\omega}{2\alpha}}x\right) \quad (2)$$

where α is the thermal diffusivity and x is the distance from a typical point to the heating point^{26,28}. This equation indicates that the temperature distributions at typical points on samples are periodic variations with the same period but with decreasing amplitudes as the distance increases²⁸. If we measured the temperature distributions of two separated points that were Δx apart, we could get two temperature waves with different time phase and different amplitudes. Then the time phase difference Δt between the two temperature waves could be easily calculated by finding peaks. We defined the “thermal transfer speed” $v = \Delta x/\Delta t$, then the thermal diffusivity $\alpha = v^2/2\omega$ according to the temperature-equation given above^{26,28}. The thermal conductivity also could be determined by the equation $k = C_p \alpha \rho$, where k is the thermal conductivity, C_p is the heat capacity and ρ is the density of the measured material. In view of the measuring method described above, in order to determine the thermo-physical properties, key works of our measuring method are the accurate measurements of distance between two typical points and time phase difference between two temperature waves. So the amplitudes of temperature waves are not important factors. Furthermore, when the one-dimensional periodic heat transfer reaches to a dynamically stable state, the time phase of the temperature waves won't be influenced by the atmosphere, even though the amplitudes of the temperature waves could be influenced by radiation and convection. Our measuring method only depends on the time phase difference and the distance, so the atmosphere will scarcely affect our measured results. It is a great advantage over other measuring techniques. As widely known, it is very hard to accurately measure the amplitude of temperature and eliminate the influence of surroundings simultaneously.

The schematic illustration of our measuring method is shown in Figure 1a. Six kinds of SACNT buckypapers with different densities were fabricated. Then every sample was cut into 2 mm width \times 5 cm length ribbons along two different directions. In this way, we could get two groups of samples, *i.e.*, one group of samples were cut along the CNT aligned direction and another group of samples were cut perpendicular to the CNT aligned direction. Because of the very large ratio of length to width and thickness, we could reasonably treat these samples as one-dimensional materials. Then every sample was pasted on a pair of aluminum substrates by a high-purity silver paste to get perfect thermal contact conditions (Figure 1b). The aluminum substrates acted as heat sinks so that two ends of sample were always at room temperature. A near-infrared laser (Figure 1a, Heating Laser) was used to heat the suspended SACNT buckypaper ribbon at the middle point, and we set an optical chopper in the front of the heating laser to get a sinusoidal heating flow. In this way, periodic and damping temperature waves were spreading along the SACNT buckypaper ribbon. We used two same infrared thermometers (NIR Laser I & NIR Laser II in Figure 1a) to collect the temperature data of two separated points on the suspended SACNT buckypaper ribbons. The infrared thermometer is a kind of non-contact thermometer, and its spatial and temperature resolution are 1 mm and 0.1 K respectively. We could precisely set the two thermometers synchronous when they were measuring temperatures of the same

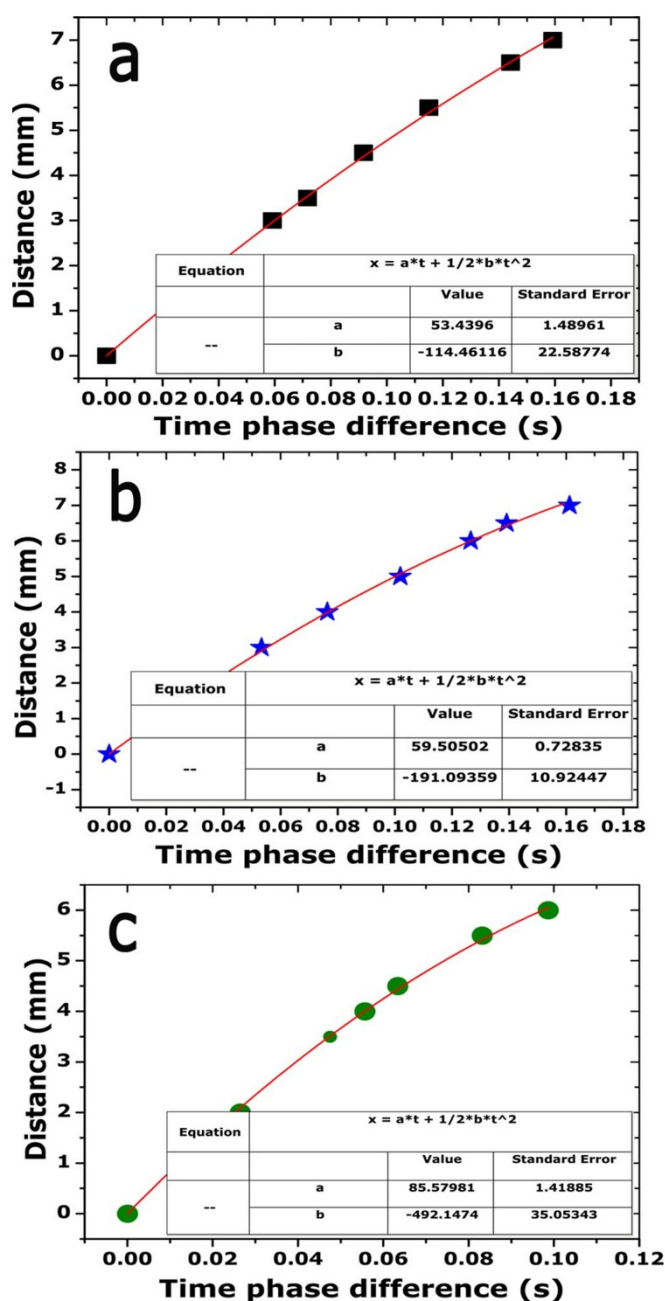


Figure 2 | The measured distance as a function of time phase difference. The figure (a), (b) and (c) show the results of the SACNT buckypapers with three densities 0.75 g/cm³, 0.81 g/cm³ and 0.93 g/cm³ respectively. The inset table is the fitting data that fitted by the uniformly variable motion equation $x = at + 1/2bt^2$. The constant “a” represent the initial parallel thermal transfer speed. These inset tables clearly show that the initial parallel thermal transfer speed increase as the density increases. The constant “b” could be treated as the acceleration of heat transfer, which could be regarded as the thermal damping factor in the SACNT buckypapers.

point. The temperature data was collect for every 5 microseconds by the thermometers. The amplitudes and time phase of temperature waves collected by the two thermometers were displayed on computer. In this way, we obtained two temperature waves with different amplitudes and different time phase (Figure 1c). Then the time phase difference (Δt) could be accurately measured by finding peaks in the Origin, and the distance (Δx) between two points (points A & B in Figure 1a) was controlled by a high-precision translation-and-lift



stage (resolution 0.01 mm). Finally, the thermal transfer speed $v = \Delta x/\Delta t$ and the thermal diffusivity $\alpha = v^2/2\omega$ could be calculated, where ω is the angular frequency of the optical chopper. For verification of our method, we also measured the thermo-physical properties of a kind of polycrystalline graphite with known thermal diffusivity, which is a kind of isotropic carbon allotrope. The graphite was cut into the same size with the SACNT buckypapers, as well as the thickness is about 100 μm . In the present work, the room-temperature thermal diffusivity of this graphite is about $31.9 \pm 3.4 \text{ mm}^2/\text{s}$, which has very good agreement with the former result achieved by the laser flash method³⁰. This agreement confirmed the validity of our measuring method.

The thermal transfer speed along the CNTs aligned direction. Based on the method described above, we could investigate the thermal damping mechanisms in the SACNT buckypapers. We changed the distances between the two measured points on the suspended SACNT buckypaper ribbons and we got a series of time phase differences with different distances (more details in method section). In this way, we could study the variation of thermal transfer speed as measured distances change. For low density parallel samples and all perpendicular samples in our experiment, they didn't show obviously regular variation of v as measured distances change. We suggest that this phenomenon is mainly due to numerous interspaces and a lot of crooks of individual CNTs in low density and perpendicular samples. When the densities of parallel samples reach to about 0.75 g/cm^3 , the parallel thermal transfer speeds of SACNT buckypapers show a novel characteristic. If we put our measured data in the rectangular coordinate system, in which the x-axis refers to the time phase difference and y-axis refers to the measured distance, we found that the data shows a similarly parabola relation. The distance-time relationships of SACNT buckypapers with three different densities were shown in Figure 2.

In order to obtain the information of thermal transfer damping mechanisms in SACNT buckypapers, we assumed that the thermal transfer is a similar uniformly variable motion. Then we tried to fit them by the uniformly variable motion equation $x = at + 1/2bt^2$, the fitting data and the standard error were shown in the inset tables in Figure 2. The constant "a" represented the initial parallel thermal transfer speed. The constant "b" could be treated as the acceleration of thermal transfer, which could be regarded as the thermal damping factor. The parallel thermal transfer speeds of three samples are $53.4 \pm 1.5 \text{ mm/s}$, $59.5 \pm 0.7 \text{ mm/s}$ and $85.6 \pm 1.4 \text{ mm/s}$ and corresponding accelerations are $-114.5 \pm 22.6 \text{ mm/s}^2$, $-191.1 \pm 10.9 \text{ mm/s}^2$ and $-492.1 \pm 35.1 \text{ mm/s}^2$ respectively. Our results indicate that the parallel thermal transfer speed increases as the density increases. We suggest that this is mainly due to the fact that there are much fewer interspaces and CNT crooks in higher density buckypapers. It is known that the interspaces between individual CNTs and crooks of CNTs could strongly influence the heat transfer. So heat will transfer more effectively in the higher density buckypapers. On the other hand, our measured results demonstrated that the absolute magnitude of acceleration also increase as the density increases. This is a novel finding and we couldn't establish the exact mechanism of the finding up to now. Nevertheless we guess that the mechanism of the finding may relate to the tube-tube contacts between individual CNTs. More specifically, in unit length of SACNT buckypapers with different densities, there are much more tube-tube contacts between individual CNTs in higher density buckypapers. Several former works had demonstrated that tube-tube contacts between individual CNTs have significant thermal resistance because of phonon-phonon interacting^{31,32}.

Thermal diffusivities and thermal conductivities at room temperature. Using the one-dimensional periodic heating method and the equation $\alpha = v^2/2\omega$, we could obtain the thermal diffusivities of all SACNT buckypapers at room temperature. Two series of thermal

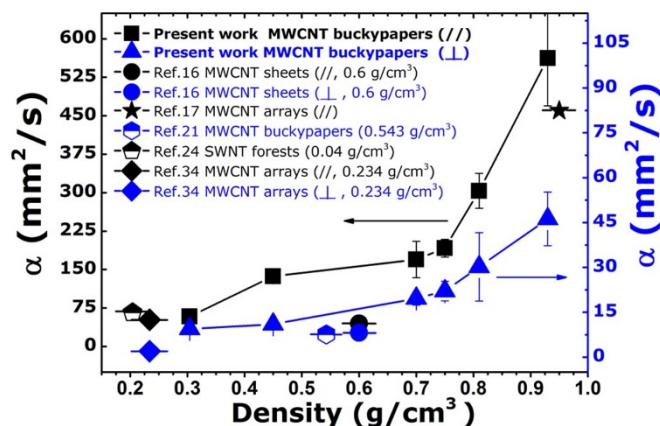


Figure 3 | The anisotropic thermal diffusivities (α) of SACNT buckypapers with different densities at room temperature. The figure clearly shows that both parallel and perpendicular thermal diffusivities increase as the density increases. The error bars include the error arising from time measurements by infrared thermometers and the error arising from repeating measurements. Several typical referenced results of different types of CNT-based macroscopic materials were listed in the figure. It is worth noting that the result of the Ref. 24 is the thermal diffusivity of single-walled CNTs (SWNT) forests. The Ref. 16 and Ref. 34 also included anisotropic values. The parallel and perpendicular values are marked by “//” and “ \perp ” respectively. The specific densities of several referenced materials were also labeled in the figure.

diffusivities were achieved because of the super-aligned arrangements of individual CNTs in the SACNT buckypapers, *i.e.*, the parallel thermal diffusivities and perpendicular thermal diffusivities (Figure 3). For simplicity, the parallel and perpendicular thermal diffusivities are marked by “//” and “ \perp ” respectively in Figure 3. As is shown in Figure 3, the two series of thermal diffusivities all increase as the density of SACNT buckypapers increases. We suggested that this phenomenon is mainly due to fewer interspaces and much closer CNT-CNT contacts in higher density SACNT buckypapers. For the lowest density SACNT buckypapers (0.30 g/cm^3), the parallel and perpendicular thermal diffusivity are $58.6 \pm 3.2 \text{ mm}^2/\text{s}$ and $9.5 \pm 0.6 \text{ mm}^2/\text{s}$ respectively. For the highest density SACNT buckypapers (0.93 g/cm^3), the parallel thermal diffusivity is as high as $562.2 \pm 55.4 \text{ mm}^2/\text{s}$. This result demonstrates that SACNT buckypapers have a very high thermal diffusivity at room temperature. For several common materials, such as copper and aluminium, their room-temperature thermal diffusivities are about $110 \text{ mm}^2/\text{s}$ and $90 \text{ mm}^2/\text{s}$ respectively³⁰. This superiority indicates that the high-density SACNT buckypapers have very good heat-transfer properties. This result also confirms that the SACNT buckypapers partly inherit the excellent properties of individual CNTs. Additionally the SACNT buckypapers have obviously anisotropic thermal diffusivities. The parallel thermal diffusivities are bigger than the corresponding perpendicular thermal diffusivities by almost one order of magnitude (Figure 3). This is mainly due to the fact that the heat transfers much more efficiently along the CNT aligned direction than that across the CNT aligned direction^{32,33}.

Our measured results and several former representative results of thermal diffusivities are shown in Figure 3. These referenced results are all experimental values that were obtained by different measuring techniques^{16,17,21,24,34}. These referenced CNT-based macroscopic materials included sheets, arrays, forests and buckypapers. Several referenced results also contained parallel and perpendicular values^{16,34}, and they were marked by different symbols and colors in Figure 3. The data in Figure 3 demonstrates that our measured results are higher than most of the former results. We believe that this

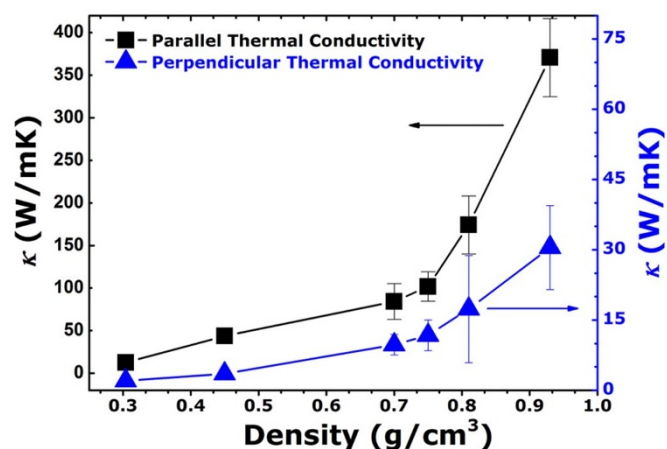


Figure 4 | The anisotropic thermal conductivities of SACNT buckypapers that were calculated by the equation $k = C_p \rho \alpha$. Both parallel and perpendicular thermal conductivities increase as the density increases. For the same density samples, the parallel thermal conductivity is much bigger than the corresponding perpendicular one.

is mainly due to the fact that individual MWCNTs in our buckypapers are arranged in very high quality alignment, and our SACNT buckypapers may have fewer impurities.

Finally, according to the equation $k = C_p \rho \alpha$, we could calculate room-temperature thermal conductivities of these SACNT buckypapers. The specific heat capacity of MWCNTs at room temperature is about $C_p = 720 \text{ J} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$, which is the same with that of graphite^{16,19}. The parallel thermal conductivities and perpendicular thermal conductivities calculated by the equation $k = C_p \rho \alpha$ are shown in Figure 4. Considering the error range, our calculated parallel thermal conductivity of the 0.93 g/cm^3 SACNT buckypapers have good agreement with our former results measured by other methods². Figure 4 shows that high-density SACNT buckypapers have very high thermal conductivity in parallel direction. Our highest measured thermal conductivity is superior to that of several common heat-transfer materials, such as aluminum and copper. Moreover, the SACNT buckypapers have much lower density. So we believe that the SACNT buckypapers will be widely used as heat-transfer materials in the future. Figure 4 also shows positive correlation between the thermal conductivities and densities. This positive correlation is owing to the much closer CNT arrangement in higher density SACNT buckypapers. The much closer CNT arrangement means fewer interspaces and bigger contact areas between individual CNTs. So heat will transfer more effectively in higher density SACNT buckypapers. The parallel thermal conductivities are much bigger than the corresponding perpendicular thermal conductivities, which is mainly due to the fact that the heat transfers much more efficiently along the CNT aligned direction than that across the CNT aligned direction^{32,33}.

Discussion

Our measuring method reported in this paper is based on the one-dimensional periodic heat transfer equation, and the thermo-physical properties were calculated by the thermal pulse transfer speed. The key works of our measurement were accurate measurements of distances and time phase differences rather than the exact amplitudes of temperature. According to the periodic temperature equation, the time phase of the temperature waves won't be influenced by the atmosphere when the one-dimensional periodic heat transfer reaches to the dynamically stable state, even though the amplitudes of the temperature waves are influenced by radiation and convection. Therefore the surroundings (including the atmosphere and the substrates) will have tiny influence on our measured results. It is a

great advantage of our method over other measuring techniques. On this basis, we tried to investigate the thermal damping mechanisms in the SACNT buckypapers. It is found that high-density ($>0.75 \text{ g/cm}^3$) SACNT buckypapers have very good heat-transfer properties along the CNT-aligned directions. Moreover, we found that the parallel thermal damping factors increase as the density of SACNT buckypapers increases. Up to now we couldn't clearly comprehend this finding and we just guessed that this is mainly due to more tube-tube contacts in unit length of higher density SACNT buckypapers. In order to verify our suggestion presented here, more investigations will be developed to study this finding in the future.

Furthermore, we obtained the thermal diffusivities and thermal conductivities of SACNT buckypapers with different densities. Their thermal diffusivities and thermal conductivities are anisotropic because of the super-aligned arrangement of individual CNTs. Both the thermal diffusivity and the thermal conductivity are strongly dependent on the density, we suggested that this is mainly owing to the much more closely CNT arrangement in higher density SACNT buckypapers. Compared with several representative reported experimental results of thermal diffusivities, our measured results are higher than most of the previous results. We suggested that this is mainly due to the very high quality alignment of individual CNTs and fewer impurities in our buckypapers. The highest calculated thermal conductivities of our samples is about 370 W/mK , this is a relatively high thermal conductivity of CNT-based macroscopic materials. Compared with common heat-transfer materials, the SACNT buckypapers reported here possess superior thermal conductivity as well as much lower density. So we believed that the SACNT buckypapers will be extensively used as heat-transfer materials in the future. In this work, we also measured the thermal properties of a kind of polycrystalline graphite for verification and comparison. Its thermal transfer speed and thermal diffusivity are about 22.3 mm/s and $31.6 \text{ mm}^2/\text{s}$ respectively. The values are obviously lower than the corresponding results of SACNT buckypapers, which confirms that our fabricated SACNT buckypapers have very good thermal properties.

In summary, we designed a simple and precisely measuring system to study the thermo-physical properties of SACNT buckypapers with different densities. Our measuring techniques could be easily manipulated and has superiority to other techniques. Specifically, our measured results were tiny influenced by the surroundings. In order to describe the thermal transfer damping mechanisms in SACNT buckypapers, we defined the parameter "thermal transfer speed" and investigated its variation as the measured distances change. We found that the higher density SACNT buckypapers have bigger parallel thermal damping factor. We just guessed that it is mainly due to the thermal resistances at the tube-tube contacts. Our results also demonstrated that SACNT buckypapers have anisotropic thermal diffusivities because of the super-aligned arrangement of individual CNTs. The thermal diffusivities of SACNT buckypapers could be as high as $562.2 \pm 55.4 \text{ mm}^2/\text{s}$ with the corresponding density 0.93 g/cm^3 . This is a relatively higher value than that of several common materials. The anisotropic thermal conductivities of SACNT buckypapers were also calculated in this paper by the equation $k = C_p \rho \alpha$. Our results indicated that our SACNT buckypapers have very outstanding heat-transfer properties.

Methods

Sample preparation. Firstly, we drew super-aligned MWNTs sheets from the super-aligned MWNT arrays fabricated by chemical vapor deposition method³. Secondly, we put hundreds of the super-aligned MWNT sheets together in one direction precisely and then we drenched them into alcohol so as to shrink the CNTs in the sheets. Thirdly, these super-aligned MWNT sheets were pressed with different pressures to get different density SACNT buckypapers. Then the SACNT buckypapers were cut into $2 \text{ mm} \times 5 \text{ cm}$ ribbons by high power laser in two directions. One is parallel to the aligned direction of individual MWNTs in SACNT buckypapers and the other is perpendicular to the aligned direction. In this way, we got two groups of samples, the parallel samples and the perpendicular samples. Then



every SACNT buckypaper ribbon was pasted on a pair of aluminum substrates by high-purity silver paste. Then we got a series of samples with suspended SACNT buckypaper ribbons pasted on substrates. Finally, all the aluminum substrates were fixed on the optical platform. The sample size was measured by a micrometer, and the density was computed by the mass and volume of samples. The microstructures of samples were probed by scanning electron microscope (Sirin 200, resolution 1.0 nm).

Measuring process and instrument information. We used a heating laser (central wavelength 960 nm) to heat the sample at the middle point. The heating power range is 0–10 W and the size of heating points could be controlled by lens. The frequency of the optical chopper was controlled by a KEITHLEY voltage source. The chopper was set at the same frequency for all measurements. The Optris LS infrared thermometer's spatial and temperature resolutions are 1 mm and 0.1 K. They were fixed on a high-precision translation-and-lift stage to ensure the precise position control, where they could be moved in three-dimensions and the spatial resolution is 0.01 mm. We fixed the NIR Laser I to measure the temperature of a typical point (point A in Figure 1a) that was very close to the heating point, the position of NIR Laser II could be precisely controlled and changed by the translation-and-lift stage. Before started the measurement, we carefully and precisely controlled the two thermometers measuring at the same point, and then we set them synchronous. We could change and control the measuring distance between the two thermometers by the translation-and-lift stage with resolution 0.01 mm. After measurement, the time phase difference could be achieved by finding peaks in the Origin.

- Liu, L., Ma, W. & Zhang, Z. Macroscopic carbon nanotube assemblies: Preparation, properties, and potential applications. *Small* **7**, 1504–1520 (2011).
- Zhang, L., Zhang, G., Liu, C. & Fan, S. High-density carbon nanotube buckypapers with superior transport and mechanical properties. *Nano Lett.* **12**, 4848–4852 (2012).
- Jiang, K., Li, Q. & Fan, S. Nanotechnology: Spinning continuous carbon nanotube yarns. *Nature* **419**, 801 (2002).
- Dalton, A. B. *et al.* Super-tough carbon-nanotube fibres. *Nature* **423**, 703 (2003).
- Nanot, S. *et al.* Broadband, polarization-sensitive photodetector based on optically-thick films of macroscopically long, dense, and aligned carbon nanotubes. *Sci. Rep.* **3**, 1335 (2013).
- Zhou, W., Bai, X., Wang, E. & Xie, S. Synthesis, structure, and properties of single-walled carbon nanotubes. *Adv. Mater.* **21**, 4565–4583 (2009).
- Zhang, H. *et al.* Electrical and thermal properties of carbon nanotube bulk materials: Experimental studies for the 328–958 K temperature range. *Phys. Rev. B* **75**, 205407 (2007).
- Jakubinek, M. B. *et al.* Thermal and electrical conductivity of array-spun multi-walled carbon nanotube yarns. *Carbon* **50**, 244–248 (2012).
- Wang, D., Song, P. C., Liu, C. H., Wu, W. & Fan, S. S. Highly oriented carbon nanotube papers made of aligned carbon nanotubes. *Nanotechnology* **19**, 075609 (2008).
- Baughman, R. H. *et al.* Carbon nanotube actuators. *Science* **284**, 1340–1344 (1999).
- Wei, Y. *et al.* Efficient fabrication of carbon nanotube micro tip arrays by tailoring cross-stacked carbon nanotube sheets. *Nano Lett.* **12**, 2071–2076 (2012).
- Yang, C., Lin, Y. & Nan, C. W. Modified carbon nanotube composites with high dielectric constant, low dielectric loss and large energy density. *Carbon* **47**, 1096–1101 (2009).
- Kim, P., Shi, L., Majumdar, A. & McEuen, P. L. Thermal transport measurements of individual multiwalled nanotubes. *Phys. Rev. Lett.* **87**, 215502 (2001).
- Li, Q. W., Liu, C. H., Wang, X. S. & Fan, S. S. Measuring the thermal conductivity of individual carbon nanotubes by the Raman shift method. *Nanotechnology* **20**, 145702 (2009).
- Pop, E., Mann, D., Wang, Q., Goodson, K. & Dai, H. Thermal conductance of an individual single-wall carbon nanotube above room temperature. *Nano Lett.* **6**, 96–100 (2005).
- Aliev, A. E. *et al.* Thermal transport in MWCNT sheets and yarns. *Carbon* **45**, 2880–2888 (2007).
- Xie, H., Cai, A. & Wang, X. Thermal diffusivity and conductivity of multiwalled carbon nanotube arrays. *Phys. Lett. A* **369**, 120–123 (2007).
- Mäklén, J. *et al.* Thermal diffusivity of aligned multi-walled carbon nanotubes measured by the flash method. *Phys. Status Solidi B* **248**, 2508–2511 (2011).
- Yi, W., Lu, L., Dian-lin, Z., Pan, Z. W. & Xie, S. S. Linear specific heat of carbon nanotubes. *Phys. Rev. B* **59**, R9015–R9018 (1999).
- Hou, J. *et al.* Thermal characterization of single-wall carbon nanotube bundles using the self-heating 3 omega technique. *J. Appl. Phys.* **100**, 124314 (2006).
- Yue, Y., Huang, X. & Wang, X. Thermal transport in multiwall carbon nanotube buckypapers. *Phys. Lett. A* **374**, 4144–4151 (2010).
- Hou, J., Wang, X. & Guo, J. Thermal characterization of micro/nanoscale conductive and non-conductive wires based on optical heating and electrical thermal sensing. *J. Phys. D: Appl. Phys.* **39**, 3362–3370 (2006).
- Hou, J., Wang, X. & Zhang, L. Thermal characterization of submicron polyacrylonitrile fibers based on optical heating and electrical thermal sensing. *Appl. Phys. Lett.* **89**, 152504 (2006).
- Akoshima, M. *et al.* Thermal diffusivity of single-walled carbon nanotube forest measured by laser flash method. *Jpn. J. Appl. Phys.* **48**, 05EC07 (2009).
- Jiang, K. *et al.* Superaligned carbon nanotube arrays, films, and yarns: A road to applications. *Adv. Mater.* **23**, 1154–1161 (2011).
- Kosky, P. G., Maylotte, D. H. & Gallo, J. P. Angstrom methods applied to simultaneous measurements of thermal diffusivity and heat transfer coefficients: Part 1, theory. *Int. Comm. Heat Mass Transfer* **26**, 1051–1059 (1999).
- Bodzenta, J. *et al.* Measurement of the thermal diffusivity of dental filling materials using modified Angstrom's method. *Dent. Mater.* **22**, 617–621 (2006).
- Kakac, S. & Yener, Y. *Heat Conduction*. 3rd Edition. (Taylor & Francis Inc., Bristol PA, 1992).
- Hideyuki, K. E. A. Anisotropic thermal-diffusivity measurements by a new laser-spot-heating technique. *Meas. Sci. Technol.* **12**, 2074–2080 (2001).
- Min, S., Blumm, J. & Lindemann, A. A new laser flash system for measurement of the thermophysical properties. *Thermochim. Acta* **455**, 46–49 (2007).
- Prasher, R. Thermal boundary resistance and thermal conductivity of multiwalled carbon nanotubes. *Phys. Rev. B* **77**, 75424 (2008).
- Yang, J. K. *et al.* Contact thermal resistance between individual multiwall carbon nanotubes. *Appl. Phys. Lett.* **96**, 023109 (2010).
- Zhang, G., Liu, C. & Fan, S. Temperature dependence of thermal boundary resistances between multiwalled carbon nanotubes and some typical counterpart materials. *ACS Nano* **6**, 3057–3062 (2012).
- Borca-Tasciuc, T. *et al.* Anisotropic thermal diffusivity of aligned multiwall carbon nanotube arrays. *J. Appl. Phys.* **98**, 54309 (2005).

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Author contributions

G.Z. contributes to carrying out the experiments and drafting the manuscript. C.L. contributes to designing this research and analyzing the results. S.F. contributes to providing supports for the work running.

Additional information

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