

# 1 One-directional thermal transport in densely aligned 2 single-wall carbon nanotube films

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12 ABSTRACT: Individual carbon nanotubes (CNTs) possess extremely high thermal conductivities.  
13 However, the thermal conductivities and their anisotropy of macroscopic assemblies of CNTs have  
14 so far remained small. Here, we report results of directional thermal transport measurements on a  
15 nearly-perfectly aligned CNT film fabricated via controlled vacuum filtration. We found the

1 thermal conductivity to be  $43 \pm 2.2 \text{ W m}^{-1} \text{ K}^{-1}$  with a record-high thermal anisotropy of 500. From  
2 the temperature dependence of the thermal conductivity and its agreement with the atomistic  
3 phonon transport calculation, we conclude that the effect of intertube thermal resistance on heat  
4 conduction in the alignment direction is negligible because of the large contact area between CNTs.  
5 These observations thus represent ideal unidirectional thermal transport, i.e., the thermal  
6 conductivity of the film is determined solely by that of individual CNTs.

7

8 Thermal management in electronics is becoming more and more important as the degree  
9 of device miniaturization has reached a truly nanometer scale where the level of power dissipation  
10 is also extreme. Therefore, thermally conducting electronic nanomaterials are strongly required  
11 for more efficient heat dissipation. Carbon nanotubes (CNTs) are one of the most promising  
12 candidates, since individual CNTs have exhibited thermal conductivities ( $\kappa$ ) over  $10^3 \text{ W m}^{-1} \text{ K}^{-1}$   
13 <sup>1-5</sup>. There have also been many thermal conductivity studies of CNT assemblies. In particular,  $\kappa$   
14 has been measured for aligned CNT samples prepared either by direct chemical vapor deposition  
15 growth<sup>6-16</sup> or post-processing of synthesized CNTs, such as mechanical processing<sup>17-22</sup>, direct  
16 spinning<sup>23</sup>, and magnetic alignment<sup>24-26</sup>. However, the  $\kappa$  values reported for aligned CNT  
17 materials have so far been limited to tens to hundreds of  $\text{W m}^{-1} \text{ K}^{-1}$ , significantly lower than those  
18 for individual CNTs. This drastic difference has been attributed to structural issues such as low  
19 volume fractions (0.5 ~ 50 %), high defect densities, and low degrees of alignment, which make  
20 the intertube thermal resistance limit the film thermal conductivity. Therefore, it is essential to  
21 eliminate these structural deficiencies to utilize the high  $\kappa$  of individual CNTs in aligned CNT  
22 assemblies.

1           In addition to the general need for materials with high thermal conductivity, there is also a  
2 specific demand for materials with anisotropic thermal conductivity that can direct heat flow only  
3 in a certain direction. For example, when spreading the heat from a chip on a circuit board, such  
4 directional heat flow can prevent heat-sensitive components from being damaged by excess heat  
5 conducted from heat-generating parts<sup>27</sup>. CNTs are clearly a good candidate because of their  
6 uniquely one-dimensional structure. However, unexpectedly, the thermal anisotropy of aligned  
7 CNT assemblies has not exceeded 100 in previous reports<sup>6-9,16-20,24,25</sup>.

8           Here, we demonstrate a record-high value of thermal conductivity anisotropy in  
9 macroscopic films of aligned and packed CNTs prepared by the recently developed controlled  
10 vacuum filtration (CVF) method<sup>28,29</sup>. Using the T-type and time-domain thermoreflectance  
11 (TDTR) methods, we obtained a thermal anisotropy of 500 at room temperature (R.T.), which is  
12 the largest value obtained among all previously studied macroscopic CNT assemblies<sup>6-9,16-20,24,25</sup>.  
13 Although the  $\kappa$  in the alignment direction ( $\kappa_{\parallel}$ ) is lower than the highest reported values<sup>6-26</sup>,  
14 further theoretical analysis reveals that the  $\kappa_{\parallel}$  is determined only by the  $\kappa$  of the constituent  
15 CNTs and is not limited by intertube thermal resistance.

16           The CNT used in this study was unsorted arc-discharge CNT purchased from Carbon  
17 Solutions, Inc., and the highly aligned CNT films (Film 1) were prepared by the CVF method<sup>28</sup>.  
18 In addition, a poorly aligned CNT film (Film 2) and a randomly aligned CNT film (Film 3) were  
19 also prepared for comparison. Film 2 was prepared by filtrating the CNT dispersion with additional  
20 6 mM of NaCl at a filtration speed four times higher than that in the case of Film 1. These changes  
21 alter the interaction between the CNTs and the filter membrane<sup>28</sup> and reduce the degree of  
22 alignment. Film 3 was prepared by filtration of a CNT dispersion without any additives or filtration

1 control.  $\kappa_{\parallel}$  for each sample was measured by the T-type method<sup>1,30</sup> (Figure S1). The ~200-nm-  
2 thick Films 1 and 2 were supported by the polyethylene terephthalate (PET) substrates, and their  
3  $\kappa_{\parallel}$  were calculated by subtracting the thermal conductance of the PET substrate from the thermal  
4 conductance of the CNT/PET (CNT and PET). Film 3 was ~30  $\mu\text{m}$  thick and measured as a self-  
5 standing film. The cross-plane  $\kappa$  ( $\kappa_{\perp}$ ) of Film 1 was measured using TDTR<sup>31,32</sup> after coating the  
6 surface with a ~100-nm-thick Al transducer film. The details of our TDTR setup are described  
7 elsewhere<sup>33</sup>.

8 Top-view Scanning Electron Microscope (SEM) images of each sample are shown in  
9 Figure 1. Film 1 (Figure 1a) had uniformly aligned CNTs, as in previous reports<sup>28,29,34–39</sup>. The  
10 alignment of Film 2 (Figure 1b and 1c) is much weaker. The focused SEM image in Figure 1b  
11 shows that the CNTs are oriented rather randomly but there are parts with higher density due to  
12 the local moderate alignment that gives rise to the different color contrast. This is more evident in  
13 the broad view (Figure 1c) where white lines indicate partial ordering in Film 2. The morphology  
14 of Film 3 (Figure 1d) was clearly different from that of the other two aligned samples; most CNTs  
15 existed as large bundles with a diameter of ~100 nm, and they formed a sparse network structure.  
16 The alignment degrees of Films 1 and 2 were evaluated by measuring the reduced linear dichroism  
17 ( $LD'$ ) with a 660 nm laser beam<sup>38</sup>.  $LD'$  was 0.68 for Film 1 and 0.040 for Film 2, so the alignment  
18 degree of Film 2 was less than one tenth of that of Film 1 (Table 1). The non-zero  $LD'$  value of  
19 Film 2 is also consistent with the ordering of CNTs seen in Figure 1c.

20 The  $\kappa$  measurements of the three films were conducted over a temperature range from 50  
21 to 300 K. First,  $\kappa$  values of different samples at R.T. were compared to see any thermal property  
22 differences caused by the morphological differences, and are summarized in Table 1. The  $\kappa$  of

1 Film 1 in the alignment direction ( $\kappa_{||,1} = 43 \pm 2.2 \text{ W m}^{-1} \text{ K}^{-1}$ ) at R.T. was higher than those of Film  
2 2 ( $\kappa_{||,2} = 28 \pm 1.2 \text{ W m}^{-1} \text{ K}^{-1}$ ) and Film 3 ( $\kappa_{||,3} = 14 \pm 2.8 \text{ W m}^{-1} \text{ K}^{-1}$ ). On the other hand, the  $\kappa$  of  
3 Film 1 in the perpendicular direction ( $\kappa_{\perp,1}$ ) was as low as  $0.085 \pm 0.017 \text{ W m}^{-1} \text{ K}^{-1}$ , which is three  
4 orders of magnitude smaller than  $\kappa_{||,1}$ . This reveals that Film 1 had an extremely large thermal  
5 anisotropy ( $\kappa_{||,1} / \kappa_{\perp,1}$ ) of 500, which is the largest reported value among aligned CNT films<sup>6-9,16-</sup>  
6 <sup>20,24,25</sup>. The highest and second-highest value of  $\kappa_{||,1}$  and  $\kappa_{||,2}$  show the  $\kappa_{||}$  improvement of the film  
7 by the constituent CNT alignment, and the details of which will be discussed later. While the  
8 randomly aligned Film 3 showed lowest  $\kappa_{||}$ , its structure seen in Fig.1d contains seemingly  
9 aggregated CNT chunks. Therefore, it is difficult to separate the effect of aggregation from that of  
10 alignment. Therefore, the case of Film 3 is shown only to compare the absolute thermal  
11 conductivity value of conventional CNT mat with those of the aligned ones, and thus, the detailed  
12 heat conduction mechanism in Film 3 will not be discussed in this paper.

13 It is known that the  $\kappa$  of CNT bundles are lower than those of individual CNTs due to the  
14 quenching of low-frequency phonon modes and small thermal conductance between CNTs<sup>5,19,41,42</sup>.  
15 However, despite the nearly perfect CNT alignment in Film 1, its  $\kappa_{||,1}$  value at R.T. is still one  
16 order of magnitude smaller than the reported  $\kappa$  of a single CNT bundle ( $\kappa_{||, \text{bundle}}$ )<sup>41-43</sup>. To  
17 understand the reason, the temperature dependence of  $\kappa_{||,1}$  was examined in detail. First, the  
18 temperature dependence of  $\kappa_{||,1}$  was compared with that of  $\kappa_{||, \text{bundle}}$  measured in Ref. <sup>43</sup>. As shown  
19 in Figure 2, the profiles normalized by the R.T. value show good agreement. In Ref. <sup>43</sup>, CNT-CNT  
20 contact thermal resistance had negligible effects on  $\kappa_{||, \text{bundle}}$  because both the bundle itself and the  
21 constituent CNTs were  $1 \mu\text{m}$  long; namely, all the CNTs seamlessly connected the two thermostats

1 that suspended the bundle. Therefore, considering that the internal thermal conductance grows  
2 linearly with temperature<sup>4</sup> and the intertube thermal conductance depends weakly on  
3 temperature<sup>40,43</sup> in the range from 150 to 300 K, the agreement in temperature dependence between  
4  $\kappa_{||,1}$  and  $\kappa_{||, \text{bundle}}$  suggests that the intertube thermal resistance has a limited effect on  $\kappa_{||,1}$ .

5 To further verify this suggestion, the internal thermal conductance of a CNT in the axial  
6 direction ( $G_{||}$ ) and intertube thermal conductance at an aligned CNT-CNT contact ( $g$ ) were  
7 calculated using the atomistic Green's function (AGF) method<sup>45</sup>. The variables used for the models  
8 and calculations below are summarized in Table 2. Note that since the CNT length is shorter than  
9 the average phonon mean free path<sup>46-48</sup>, the phonon transport can be considered ballistic. As the  
10 average diameter ( $d$ ) of consisting CNT of Film 1 is  $\sim 1.4$  nm, a hexagonal unit cell consisting of  
11 (10,10) single-wall CNTs ( $d=1.36$  nm) was used as a representative atomic scale model for the  
12 calculations. While Film 1 contains both metallic and semiconducting CNTs, the calculation result  
13 with metallic (10,10) CNTs is valid for the comparison because heat transport in CNTs is  
14 dominated by phonons than by electrons<sup>49,50</sup>. As shown in Figure S2a and S2b, the prepared cells  
15 for the alignment and perpendicular directions, respectively, were repeated twice in the section  
16 between the two leads. Periodic boundary conditions were applied in the directions of the cross  
17 section (Figure S3c and S3d). The interatomic interactions within and between CNTs were  
18 modeled by the Tersoff<sup>51</sup> and Lennard-Jones potentials<sup>52</sup>, respectively; the potential parameters  
19 are described in the references.

20 The effective value of  $\kappa_{||,1}$  ( $\kappa_{||,1, \text{eff}}$ ) was estimated by the following equation based on a  
21 model shown in Figure 4a:

$$\frac{L}{\kappa_{||,1,\text{eff}}} = \frac{1}{G_{||}} + \frac{1}{g} \frac{A}{S} \quad \dots(1)$$

Here,  $L$ ,  $A$ , and  $S$  represent the length, bottom area, and side area of a constituent CNT when CNTs are viewed as a honeycomb structure. The intertube term ( $\frac{1}{g} \frac{A}{S}$ ) includes the area ratio  $\frac{A}{S}$  as  $G_{||}$  and  $g$  both correspond to the thermal conductance per unit area while the cross-sectional areas of heat conduction through CNTs and between CNTs are different. The temperature dependence of the calculated  $\kappa_{||,1,\text{eff}}$  is plotted in Figure 2, which agrees well with the experimental results. Here, the intertube term is around three orders of magnitude smaller than the internal term ( $\frac{1}{G_{||}}$ ), indicating that the temperature dependence of  $\kappa_{||,1,\text{eff}}$  reflects only that of internal thermal conductance. Therefore, from Eq. (1),  $\kappa_{||,1,\text{eff}}$  can be approximated as

$$\kappa_{||,1,\text{eff}} = LG_{||} \quad \dots(2).$$

This equation can be further transformed to

$$\kappa_{||,1,\text{eff}} = AK_{\text{ind}} \quad \dots(3)$$

where  $K_{\text{ind}}$  represents  $\kappa$  of each CNT constituting the Film 1. This shows that, when phonon transport is ballistic (i.e.,  $G_{||}$  is constant),  $\kappa_{||,1,\text{eff}}$  is determined only by the length of the constituent CNTs, or in other words, only by  $K_{\text{ind}}$ . This explains the small observed  $\kappa_{||,1}$  ( $=43 \text{ W m}^{-1} \text{ K}^{-1}$ ); it is merely because the  $K_{\text{ind}}$  of constituent CNTs are small due to their shorter length (around 200 nm) than those in the bundle in Ref. <sup>43</sup> (around 1  $\mu\text{m}$ , giving  $\kappa_{||,\text{bundle}} = 200.2 \text{ W m}^{-1} \text{ K}^{-1}$ ), not because of the intertube thermal resistance. Note that it makes sense that the five-fold difference in the length

1 results in the five-fold difference in  $\kappa$  as thermal conductivity increases linearly with the length  
2 when heat conduction is ballistic<sup>53</sup>.

3 On the other hand, the intertube thermal resistance dominates the thermal conductivity of  
4 Film 1 in the cross-plane direction ( $\kappa_{\perp,1}$ ). This can be confirmed by the good agreement in the  
5 temperature dependences of experimentally measured  $\kappa_{\perp,1}$  and calculated  $g$  (Figure 3). Here, a  
6 quadratic increase at low temperatures and weak dependence at higher temperatures are observed,  
7 which is consistent with the temperature dependence of experimentally observed intertube thermal  
8 conductance between two CNTs<sup>44</sup>. Furthermore, the actual value of the intertube thermal  
9 conductance ( $g_{\text{exp}}$ ) at R.T. was estimated from the experimental result ( $\kappa_{\perp,1}$ ) using a simple model  
10 shown in Figure 4b. In the calculation, the internal thermal conductance of a CNT in the  
11 perpendicular direction ( $G_{\perp}$ ) was assumed to be much greater than  $g_{\text{exp}}$ . Based on the model,  $g_{\text{exp}}$   
12 was calculated to be  $1.5 \times 10^{-8} \text{ m}^2 \text{ K W}^{-1}$ , which is close to  $g$  ( $1.1 \times 10^{-8} \text{ m}^2 \text{ K W}^{-1}$ ) and the other  
13 reported values calculated by molecular dynamics (MD) simulations<sup>54-56</sup>. This confirms that  $\kappa_{\perp,1}$   
14 is mainly determined by the intertube thermal resistance.

15 It is also worth noting that the temperature dependence of  $\kappa_{\parallel,2}$  is different from that of  $\kappa_{\parallel,1}$   
16 (Figure 2).  $\kappa_{\parallel,2}$  increases sharply in the low temperature regime, while it becomes almost constant  
17 above 180 K. It is tempting here to discuss the result in terms of the peak temperature shift as the  
18 peak temperature of CNT materials is known to appear between 300 to 400 K, resulting from the  
19 competition between the increase in heat capacity and decrease in the phonon mean free paths due  
20 to Umklapp scattering with increasing temperature. However, as the constituent CNT material of



1 Film 2 is exactly the same as that of Film 1, it is unlikely that their Umklapp scattering rates or the  
2 heat capacities significantly differ.

3 Instead, this temperature dependence of  $\kappa_{||,2}$  can be explained by the stronger role of  
4 intertube thermal resistance at CNT-CNT cross contacts in Film 2 as shown in the simple model  
5 (Figure 4c). With this model, the effective  $\kappa_{||}$  of Film 2 ( $\kappa_{||,2,\text{eff}}$ ) can be estimated by the following  
6 equation:

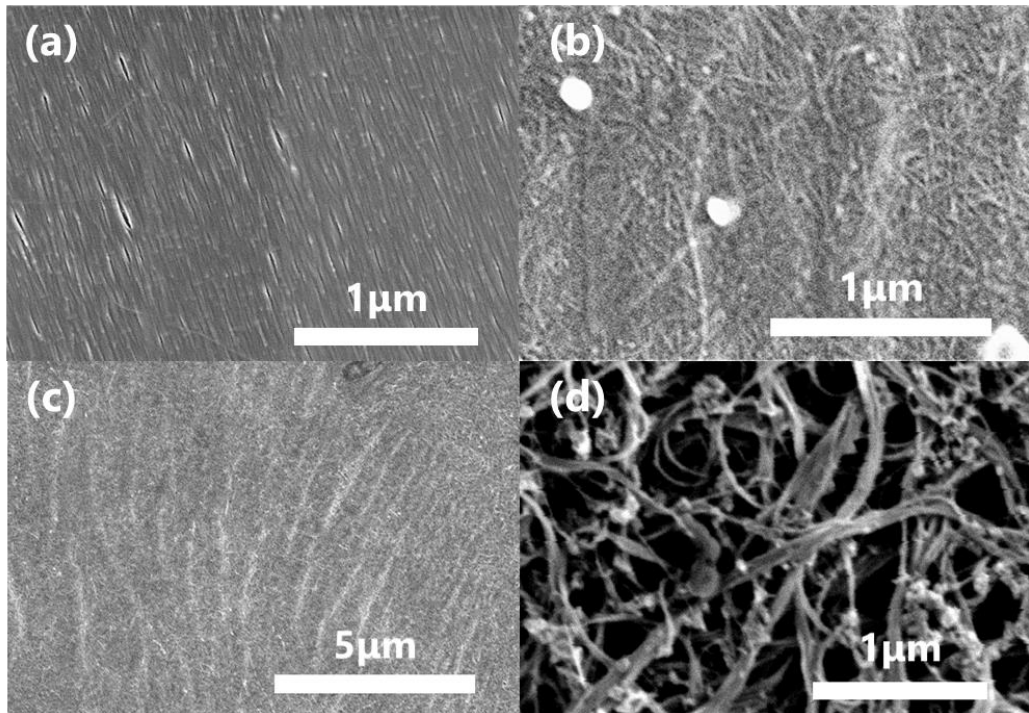
$$7 \quad \frac{L}{\kappa_{||,2,\text{eff}}} = \frac{1}{G_{||}} + \frac{1}{g'} \quad \dots(4).$$

8 Here,  $g'$  represents intertube thermal conductance at a CNT-CNT cross contact. Since the contact  
9 area is as small as the bottom area of CNT, the area ratio  $\frac{A}{S}$  is not included in Eq. (4). According  
10 to the several reports clarifying the actual value of  $g'$  from simulations, it generally ranges from  
11  $10^8$  to  $10^9 \text{ W K}^{-1} \text{ m}^{-2}$ <sup>40,56</sup>, which is one to two orders of magnitude smaller than  $G_{||}$  ( $10^{10} \text{ W K}^{-1} \text{ m}^{-2}$   
12 @ R.T.). Therefore, in contrast to the case of  $\kappa_{||,1,\text{eff}}$ , the intertube term  $\frac{1}{g'}$  is dominant in Eq.  
13 (4), and the temperature dependence of  $\kappa_{||,2,\text{eff}}$  is expected to follow that of  $g'$ . The temperature  
14 dependence of  $g'$  has been investigated by both simulations<sup>57</sup> and experiments<sup>44</sup>, where  $g'$  showed  
15 a quadratic increase at low temperatures while the temperature dependence became very weak  
16 above 150 K, which is consistent with the behavior of  $\kappa_{||,2}$  shown in Figure 2. This indicates that  
17 the difference in temperature dependence between  $\kappa_{||,1}$  and  $\kappa_{||,2}$  arises from the difference in  
18 leading mechanism of thermal resistance.

1            In conclusion, the  $\kappa$  of a highly aligned CNT film synthesized by the CVF method was  
2 measured both in the alignment direction and perpendicular direction. The  $\kappa$  value at R.T. was  $43$   
3  $\pm 2.2 \text{ W m}^{-1} \text{ K}^{-1}$  and  $0.085 \pm 0.017 \text{ W m}^{-1} \text{ K}^{-1}$  in the alignment and perpendicular directions,  
4 respectively, yielding a thermal anisotropy of 500, the highest ever reported. Further analysis of  
5 the temperature dependence of  $\kappa_{||}$  revealed that the effect of intertube thermal resistance, which is  
6 known to be large in pervious CNT films with weaker alignment, has a negligible influence on the  
7  $\kappa_{||}$  owing to the large intertube contact area realized by the nearly-perfect alignment, and  $\kappa_{||}$  is  
8 determined only by  $\kappa$  of the constituent CNT length. This also suggests that the  $\kappa_{||}$  can be even  
9 greater with longer constituent CNTs.

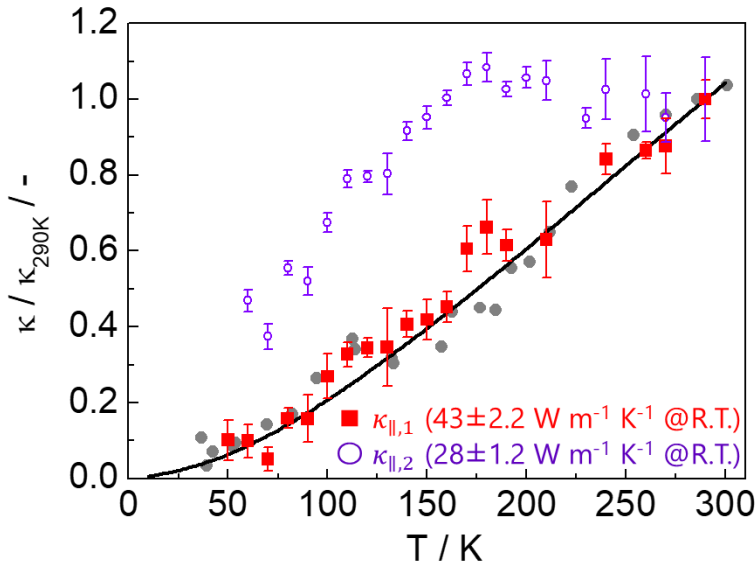
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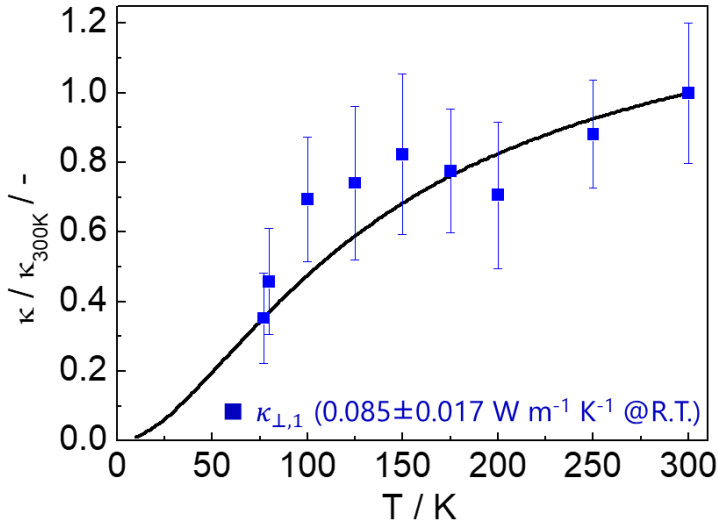


12

1 **Figure 1.** Scanning electron microscopy (SEM) images: (a) Film 1 (highly aligned film prepared  
 2 by CVF method), (b) Film 2 (poorly aligned film prepared by CVF method with NaCl addition),  
 3 (c) Film 2 with lower magnification than (b) and (d) Film 3 (randomly aligned film prepared by  
 4 filtration of a CNT dispersion without any additives or filtration control.).

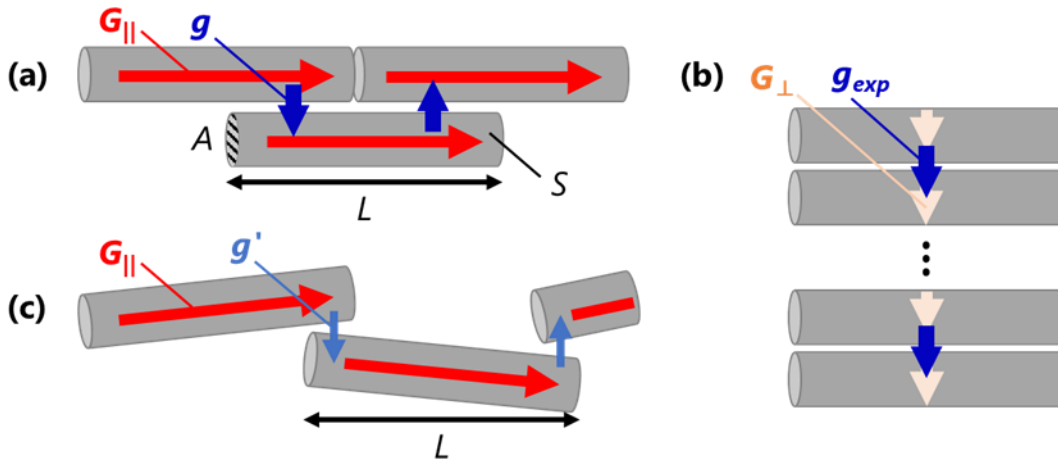


5  
 6 **Figure 2.** Temperature dependence of thermal conductivity. ■: Film 1 (alignment direction), ●:  
 7 single CNT bundle<sup>43</sup>, ○: Film 2 (alignment direction), solid line: simulated effective thermal  
 8 conductivity of highly aligned CNT film. The values of thermal conductivity are normalized by  
 9 the R.T. value for each material.



1

2 **Figure 3.** ■: Normalized thermal conductivity of Film 1 in the perpendicular direction in the  
 3 temperature range of 77 to 300 K, solid line: normalized simulated thermal conductance of highly  
 4 aligned CNT in the perpendicular direction.



5

6 **Figure 4.** (a) Simulation models to estimate the effective thermal conductivity of Film 1 in the  
 7 alignment direction.  $L$ ,  $A$  and  $S$  represent length, bottom area and side area of a constituent CNTs.  
 8  $G_{||}$  represents thermal conductance of internal CNT and  $g$  and  $g'$  represent intertube thermal

1 conductance at parallel/cross contact, respectively. (b) Simulation model for the calculation of  
 2 intertube thermal resistance at CNT-CNT contact. (c) Simulation models to estimate the effective  
 3 thermal conductivity of Film 2.

4

	Film 1 (Highly aligned)	Film 2 (Poorly aligned)	Film 3 (Randomly aligned)
$LD'$	0.68	0.040	0
$\kappa_{\parallel}$	$43 \pm 2.2 \text{ W m}^{-1} \text{ K}^{-1}$	$28 \pm 1.2 \text{ W m}^{-1} \text{ K}^{-1}$	$14 \pm 2.8 \text{ W m}^{-1} \text{ K}^{-1}$
$\kappa_{\perp}$	$0.085 \pm 0.017 \text{ W m}^{-1} \text{ K}^{-1}$	-	-

5

6 **Table 1.**  $LD'$  and  $\kappa$  at R.T. of the three films studied.

Variable	Explanation	Source
$G_{\parallel}$	Internal thermal conductance of a CNT in the axial direction	AGF calculation
$G_{\perp}$	Internal thermal conductance of a CNT in the perpendicular direction	-
$g$	Intertube thermal conductance at an aligned CNT-CNT contact	AGF calculation
$g_{\text{exp}}$	Actual value of the intertube thermal conductance at R.T.	Model in Fig.4b
$g'$	Intertube thermal conductance at a CNT-CNT cross contact	Ref. 40,56
$\kappa_{\parallel,1,\text{eff}}$	The effective value of $\kappa_{\parallel,1}$	Model in Fig.4a
$\kappa_{\parallel,2,\text{eff}}$	The effective value of $\kappa_{\parallel,2}$	Model in Fig.4c
$\kappa_{\text{ind}}$	$\kappa$ of each CNT constituting the Film 1	-
$\kappa_{\parallel,\text{bundle}}$	$\kappa$ of CNT bundle	Ref. 43

7 **Table 2.** List of the variables used in the calculation and their details.

8

1 SUPPLEMENTARY MATERIAL

2 See supplementary material for additional information regarding the synthesis method for  
3 CNT films, experimental setup, theoretical equation for thermal measurement, details for AGF  
4 calculation and absolute values of experiment/simulation data.

5 ACKNOWLEDGMENT

6 This research was supported by JSPS KAKENHI Grant Numbers 19H00744. N.K., W.G.,  
7 and J.K. acknowledge support by the Basic Energy Science (BES) program of the U.S.  
8 Department of Energy through Grant No. DE-FG02-06ER46308 (for preparation of aligned  
9 carbon nanotube films), the U.S. National Science Foundation through Grant No. ECCS-  
10 1708315 (for optical measurements), and the Robert A. Welch Foundation through Grant No. C-  
11 1509 (for structural characterization measurements). We thank M. Ouchi (Iwase Group, Waseda  
12 University) for providing technical support with the processing of measurement sample with  
13 laser plotter and G. Timothy Noe II and Kevin Tian (Rice University) for proofreading the  
14 manuscript.

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