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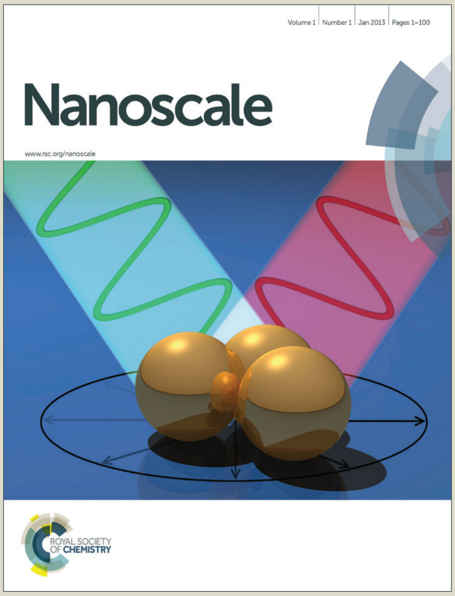

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ARTICLE

A Heat Flux Modulator of Carbon Nanotubes

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For the heat flux modulator, its most difficult problem is that the main carriers named 'phonons' have little response to the external fields. Among the exsiting articles, most of them were theoretical works and the material systems for the theoretical calculations are artificial lattices. In this paper, we made a heat modulator with the ultrathin buckypaper whic was made of multi-layer carbon nanotube sheets overlapped together, and achieved an on/off ratio whose value was 1.41 using an implending block in experiments without special optimizations and when the temperatures of the two sides we on appropriate value, we could even see the negative heat flux. Intuitively, the heat flux was tuned by the gap betwee the buckypaper and the impending gate, and we have observed that there was heat transferred to the impending bl The structure of the modulator is similar with the CNT transistor of the contactless gate, so the micro modulator is easy t manufacture in the future.

Introduction

In recent years, along with the development of materials preparation methods, the advance of thermal detection techniques and thermal simulation soft wares, people have gotten much knowledge about the heat transport on solids. Especially in the nanomaterials, thin films and interfaces of different materials, people have found many novel thermal properties. Researchers want to utilize these properties to demonstrate special thermal devices.

Following the development path of electric devices, many researchers invest their efforts on the non-linear thermal devices.[1-9] The most known non-linear electrical devices are diodes and transistors. They greatly promote our modern smart life. In these devices, the resistances are not constant and they are controlled by the magnitude and the direction of the applied voltage. The relation between the temperature and the heat flux is similar to the relation between the voltage and the electric current. Corresponding thermal devices are thermal diodes and thermal transistors. Relative to the non-linear electric devices, their biggest challenge is that the main carriers named 'phonons' have little response to the external fields. So researchers proposed unconventional structures to affect the heat flux.

Based on recent articles, there are several methods to get the thermal diodes which can archive the heat rectification, i.e., positive thermal resistance is smaller than the reverse thermal resistance, such as two bulk materials with different temperature-dependent thermal conductivities,[10] non-uniform mass loading[3] and phase transitions.[8,11] And researchers hav observed the thermal rectification in the experiments.[3,9] Bu for the thermal three-terminal modulator corresponding to the transistors whose heat flux could be controlled by anothe signal, there is little process both in theories and experiments. Referring to the electric transistors which control the curren with tuning the number of the carriers, the researchers firstly proposed models of the thermal modulators based o adjusting the behaviour of the phonons. Baowen Li et al. designed a theoretical model of the thermal modulator base on the phenomenon of the negative differential thermal resistance. And they pointed out the negative differen thermal resistance could take place in nonlinear lattices. Bu up to now, all the works were focused on the simulation.[12] P. J. van Zwol et al.[13] proposed another model of the thermal modulator which tuned the photon flux to modulate the hea flux. They utilized the near field radiation whose intensity much larger than the intensity of the blackbody radiation with the phase-change materials to affect the heat flux. The theoretical calculations showed that when the phase change there would be a large heat contrast, and they experimentall observed that the phase transition of VO_2 significantly affecte radiative heat transfer in near field.[13] What's more their ide takes a new direction that one could use the non-phonon method to modulate the heat flux. We are able to put our eye on looking for other behaviours which have effect to the heat transfer.

Carbon nanotube (CNT) is an ideal one-dimensional material with superior thermal, electrical and mechanical properties. I the past two decades a number of studies showed CNT had a good nature on heat transfer.[14-17] Recently buckypapers[18-2 or CNT sheets[18,21] which were drawn out from super-aligned C

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arrays have shown a huge application potential, they have been applied on flexible touch screens,[22] loudspeakers,[23] CNT bending actuators,[24] lithium ion batteries,[25] organic solar cells,[26] etc. For the CNT sheets, the heat dissipation through the surface gradually becomes stronger with the increment of their size.

For the general transistors, the gate electrode was usually contact with the conductive channel. But for the new transistor made of the CNT, they are not contact each other because of the manufacturing process,[27,28] and this way modulated the carriers with non-contact manner also have been used to characterize the electrical properties of the nano-materials.[29-31]

In our recent experiments, we found that a structure similar to the non-contact gate transistor could be used to adjust the heat flux. Specially, when the buckypaper was spanned between two aluminium blocks and it was heated by the joule heating, its temperature could be altered by changing the gap between it and the impending block (Fig. 1(a)), the temperature change was stable and repeatable. Based on this phenomenon, in this paper, we made a heat flux modulator. By adjusting the gap between the buckypaper and an impending block just like the gate electrode in the electric transistor, the heat flux along the buckypaper can be modulated. When the temperatures of the two sides were on appropriate value, we could even see the negative heat flux.

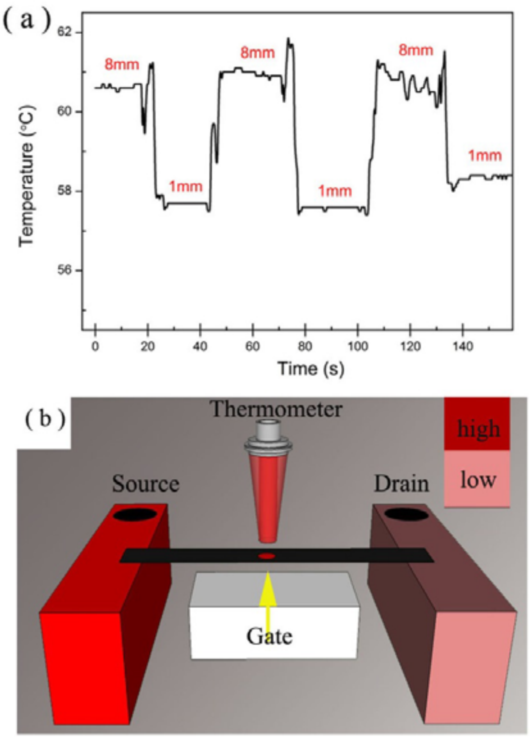

Fig. 1 (a) The temperature response with different gaps between the buckypaper and the impending block. 1mm and 8mm were the values of the gap, respectively. (b) The structure of thermal modulator. A strip of buckypaper with 4mm in width and 15mm in length was spanned between two aluminium blocks, and another aluminium block was used as the impending gate to tune the heat flux. Two infrared lasers maintained the constant temperature on the two sides ($T_s>T_D$), and an Optris LS infrared thermometer measured the temperature along the buckypaper.

Experimental

The super-aligned CNT arrays were grown by the chemical vapor deposition method.. All the CNTs were multi-walled CNTs, and their diameters were range from 10nm to 20nm. The transparent CNT sheets were drawn out from it. The sheets were loosened and the apparent thickness of the single-layer CNT sheets was about several micrometers, but after densified using ethanol the thickness could be tens of nanometers. The buckypaper was gotten from 1000-layer CNT sheets overlapped together. Its thickness, density and porosity was 70um, 0.52 g/cm^3 and 67%, respectively. The gap between the CNT bundle was range from 130nm to 250nm. The axial thermal conductivity of the buckypaper at room temperature was about 96W/(m.K). More information about the sheets and the buckypaper, *e. g.* growth process, structure information, special nature, could be found in Ref 21.

We found when the buckypaper was spanned between two aluminium blocks and it was heated by the joule heating, its temperature could be altered by changing the gap between it and an impending block (Fig. 1(a)), and the altered temperature was stable and repeatable.

Based on the above phenomenon, we made a thermal modulator of the buckypaper (Fig. 1(b)). A strip of buckypaper was spanned between two aluminium blocks and cut into 4 mm in width by the laser. The direction of the CNT made of buckypaper was along the longitudinal direction of the sample. Alcohols and silver paste were both applied on the buckypaper/block interface to decrease the interfacial thermal resistance. The distance of the two blocks was 15mm. Another aluminium block was used as the impending gate to tune the gap between the buckypaper and the surroundings. The gap was 1mm and 8mm whether the block was put under the buckypaper respectively. In order to maintain a stable external environment, the entire testing process was conducted in a closed chamber. Analogous to the electric field effect transistor, we named the three blocks as ‘Source’, ‘Drain’, and ‘Gate’, respectively. In our experiments, the source and drain blocks were kept on the stationary temperature by the heated laser. We employed an Optris LS infrared thermometer driven by a displacement station whose temperature and spatial resolution were 0.1 K and 1 mm to measure the temperature along the buckypaper and the emissivity of the buckypaper was set to 0.95 by the calibration of the thermocouple.

Results and discussion

We need to note that we only tuned the gaps between the impending block and the buckypaper. To insure the impending gate did not come into contact with the buckypaper, the minimum gap between the gate block and the buckypaper was controlled as 1 mm. In the case that only source end was heated, the working states of the modulator were shown in Fig. 2(a). When the gate block approached the buckypaper, the temperature curve along the buckypaper came down and became flat on the right side. This showed there was less heat transferred to the drain side. We repeated the tests altered the temperature of the source side, the change trend of the temperature curves was same. For the condition in Fig. 2(a), we named the state that there was less heat transferred to the drain side as “gated” state, and named the other as “ungated” state. We applied linear fitting for the temperature of the last five millimetres to calculate the heat flux to the drain side on different states, respectively. The on/off ratio of the heat flux was 1.41. In the case that both source and drain ends were at elevated temperatures, more unusual thermal phenomenon could be observed. When the temperatures of the two sides were on appropriate value, we could even see the reverse heat flux of the drain side (Fig. 2(b)). This meant that in such case the heat did not flow from high temperature to the low temperature end as usual. This phenomenon could not be seen on other heat modulation models. An application of this phenomenon we can imagine is the heat feedback network to control the temperature of the drain side. We emphasize one point that our phenomenon is not conflict to the second law of thermodynamics, because the heat could be transferred to the surroundings whose temperature is lower than the source side and drain side.

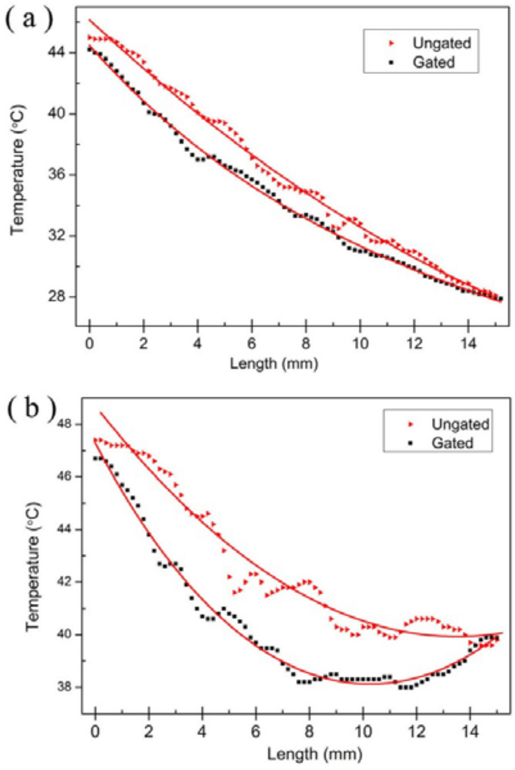

Fig. 2 The temperature curves along the buckypaper under the conditions that (a) only the source side was heated or (b) both the source and drain sides were appropriately heated. The gap between the buckypaper and the impending gate were 1mm and 8mm under the ‘Gated’ and ‘Ungated’ conditions, respectively.

By above results, we can visually see the working states of the modulator. Then we further explored the phenomenon. Firstly, we changed the gap between the buckypaper and the impending gate, substituted the aluminium block with the plastic foam as the gate under the joule heating, respectively. On the same time, we measured the temperature on the midpoint of the sample, whose value was the biggest along the buckypaper and had a large response to the environme change. Then we replaced the buckypaper with the aluminiu foil or the nickel-foam foil respectively, which wer conventional heat transferred materials.

On the condition of changing the gap, in order to use the sam aluminium block as the impending block, firstly, both the aluminium blocks on the two sides increased 7 mm, then w adjusted the gap of the buckypaper and the impending block from 1 mm to 8 mm by adding 1 mm-thick standard glas sheet under the impending block one by one. We found the relation between the temperature and the gap was not subjectively linear (Fig. 3(a)). There was a sudden drop whe the gap was about 2 mm. This distinctly showed th modulation only happened on close distance.

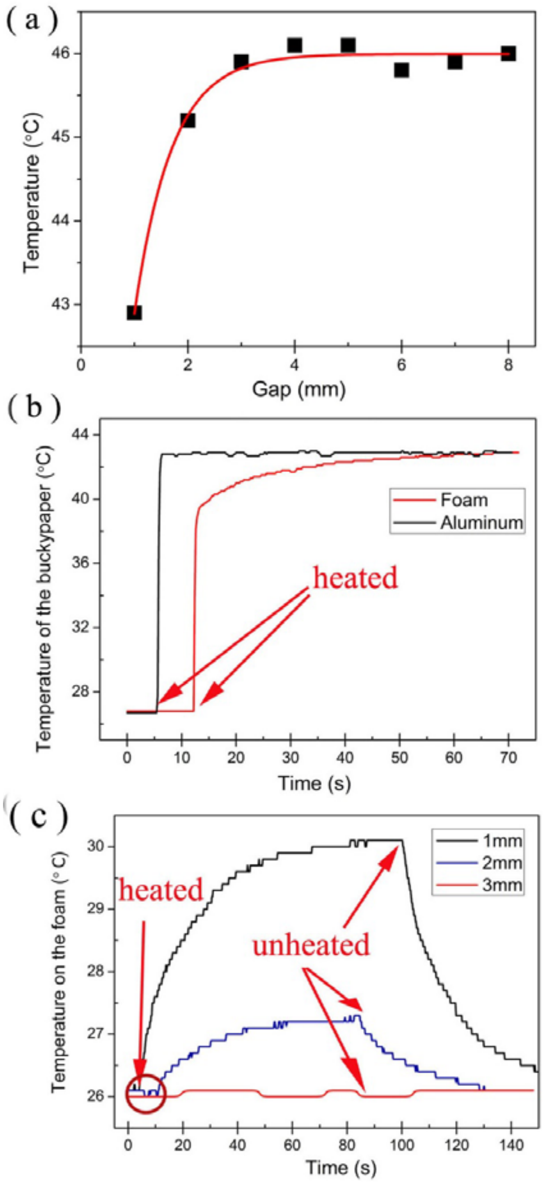

Fig. 3 (a) The relation between the temperature of the buckypaper and the gap (b) Temperature response of the buckypaper on different gate materials. (c) Temperature response of the form which was put under the buckypaper.

When we changed the gate block with the same size foam, we found the temperature response of the buckypaper had an obvious difference (Fig. 3(b)). When the gate block was aluminium, the buckypaper got balanced quickly, and when the block was the plastic foam, the buckypaper got balanced gradually. Further, we covered a piece of about 60 um-thickness buckypaper whose heat capacity was small on the foam to verify whether there was heat transferred to the foam on different gaps. The results were showed in Fig. 3(c). The results clearly showed there was heat transferred to the gate block on 1 mm or 2mm gaps, but the gap was 3 mm, there was no heat transferred to the foam. These showed there was heat transferred to the impending block only on close distance. On the other hand, the phenomenon in Fig. 3(b) also could be explained with the heat transfer between the buckypaper and the impending block. For the aluminium block, its thermal capacity was large, its temperature had little change in the experiments, the response time only related to the thermal capacity of the buckypaper which was very small, so its response time was short. But for the foam, owing of its not very large thermal capacity, there would be a heating process which could be observed in Fig. 3(b), the temperature of the foam affected the transferred heat, so the temperature of the buckypaper changed gradually. We need to point out that the balanced temperature of buckypaper should be higher in theory when the gate was foam, but we didn't observe this phenomenon. The possible reason is that the sizes were not strictly the same for the two impending blocks.

We also replaced the buckypaper with the aluminium foil and the nickel foam, respectively. Since the resistances of them were very small, we heated the block on the left side to supply a heat flux and measured the temperature at the midpoint from 1mm to 8mm of the gap. The instrument of temperature measurement was K-type thermocouple whose resolution was 0.1 K. The thickness of the aluminium foil and the nickel foam were 70um and 80um, respectively. The density, porosity and the thermal conductively of nickel foam was 0.91g/cm^3, 89.7% and 4.8 W/(m.K), respectively. The pore size of the nickel foam was range from 30um to 120um. The properties of the aluminum foil were the same with the aluminum block. Other structure information was the same with the buckypaper. However, after several experiments we didn't observe the change of temperature.

We are not sure the physical mechanism under this phenomenon. But we are sure that there is heat transferred to the impending block and it play an important role for the heat flux modulation. The behaviour between the transferred heat and the gap was similar to a variable thermal resistance tuned by the gap. Based on this, we give a phenomenological principle of the thermal modulator. The schematic of our views is shown in Fig. (4). There is a thermal channel between the source and drain (R1 and R2, R1, R2 mean the thermal resistance).There is another variable thermal channel between the source and gate tuned by the gap, and we define it as R3. When R3 is purposively changed by the gap, owing of its thermal shunt, the heat flux to the drain can be modulated.

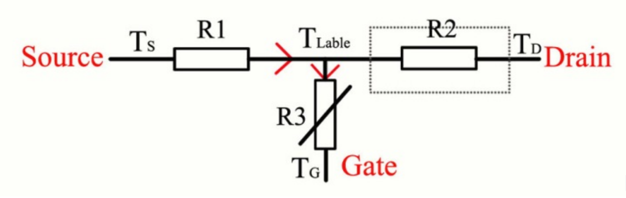

Fig. 4 Schematic of the modulator. R1 and R2 are constant thermal resistances; R3 is a variable thermal resistance. When R3 is purposively changed, owing of its thermal shunt, the heat flux to the drain can be modulated.

Next we compare the results in this article with the previous scientific results. In this paper, the most important phenomenon is that the transferred heat has a steep rise only when the gap is less than 2 mm. For this enhancement of the transferred heat on close distance, there were reports mentioned it previously.[8,32-38] However, after carefully analyzing them, we find our results have significant difference to their results.

Most of the articles related to the enhancement of heat flux at close distance are about the radiation on near field.[8,3[illegible]] When the space of the two objects on different temperatures is less than about 10 um, the radiation between them is much larger than the blackbody radiation on far field. Multiple precise experiments have observed this phenomenon.[34,35] But this phenomenon is only happened when the space is less than 10um. On the other hand, for the CNT, there was also report mentioned the enhancement of transferred heat at close distance. On the research of the micro thermoacoustic chip of the CNT sheet which only need to put CNT sheets on two electrodes, Wei yang et al.[38] found that when the groove depth was less than a value, there was a sudden and large heat dissipated to the substrates. They used the thermal wave to explain this phenomenon. The maximum groove depth that had effect to the transferred heat was less than about 100 um, what's more, it was changed as the frequency of the current. Overall, from above compare we can see our results have similar to their reports, but is apparently not the same. Our results are also the behaviour modulated by the gap, and had a steep jump on the near gap. But the transition gap which was about 2 mm in our experiments was much larger.

Up to now we are not sure the physical mechanism under this phenomenon. But we can be sure that there is heat transferred to the impending block and this modulation has a great relationship with the thermal properties of CNT. We will further study it, *e.g.* further reduce the gap by micromachining process.

Conclusions

In conclusions, we observed the heat flux enhancement between the buckypaper and the impending block on the close gap and made a heat modulator with buckypaper. In this work, we utilized the transferred heat between the buckypaper and the impending object as the gate signal to indirectly modulate the heat flux along the buckypaper. When the space between the buckypaper and the impending gate was tuned on close gap and far gap respectively, the transferred heat between them had a remarkable change, and we achieved an on/off ratio whose value was 1.41 in experiment. We have observed that there is heat transferred to the impending block, and we can be sure that this modulation has a great relationship with the thermal properties of CNT. The structure of the modulator is similar with the CNT transistor of the contactless gate, so the micro modulator is easy to manufacture. The idea indirectly modulating the heat flux along the buckypaper may take a new perspective to the study of heat modulation. In the future, we will continue to study the heat transfer enhancement on close gap and find the large heat contrast ways as the gate.

Acknowledgments

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