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2012

Loh, G. C., Teo, E. H. T., & Tay, B. K. (2012). Thermal rectification reversal in carbon nanotubes. *Journal of Applied Physics*, 112(10), 103515-.

<https://hdl.handle.net/10356/98249>

<https://doi.org/10.1063/1.4766391>

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Citation: *J. Appl. Phys.* **112**, 103515 (2012); doi: 10.1063/1.4766391

View online: <http://dx.doi.org/10.1063/1.4766391>

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Thermal rectification reversal in carbon nanotubes

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(Received 19 May 2012; accepted 20 October 2012; published online 21 November 2012)

In principle, rectifying phonon and electron flows appear similar, whereby more energy is transported in one direction than the opposite one. However, their physical mechanisms are inherently different. By using molecular dynamics simulations, this study reports on a few interesting aspects of thermal rectification in carbon nanotubes: (1) The dependence of the rectification ratio on the structural symmetry (represented by the position of vacancy clusters) of the nanotube and more importantly (2) a reversal in the rectifying direction as the normalized temperature difference of the heat baths is increased. The flux-mediated diffuse mismatch model is extended to explain the reversal phenomenon—initially with a simplifying assumption that the transmission coefficients at the vacancy/scatterer are identical in bidirectional phonon transport, and then with a moderating factor to distinguish between both coefficients. It is noted that in both cases, the conditions for thermal rectification reversal are attainable and thus explain the results of the simulations. © 2012 American Institute of Physics. [<http://dx.doi.org/10.1063/1.4766391>]

I. INTRODUCTION

Rectification is a transport phenomenon whereby the transfer of carriers in one particular direction is at a larger rate than that in the opposite direction. From the early electrolytic rectifiers,¹ plasma-type rectifiers,² and vacuum tubes³ to the modern solid state rectifiers,^{4,5} electronic rectification has been a well-examined field and has many applications in direct current power transmission systems. On the other hand, a thermal rectifier is a close analog of the electronic rectifier. While electronic rectification is defined by the dissimilitude of the magnitude of electron current in both directions, the energy carriers that are asymmetrically transported in the rectifying material/device during thermal rectification can be either electrons or phonons,⁶ depending on the dominant type of thermal carriers in the material.

II. THERMAL RECTIFICATION

The first account of thermal rectification was made by Starr,⁷ who has observed asymmetric thermal conduction in copper oxide. Later Marucha and his coworkers⁸ demonstrated the direction-dependent thermal conductivity of a monocrystalline nonhomogeneous gallium arsenide (GaAs) sample. A few other studies explored the existence of thermal rectification in dissimilar bulk materials with different dependencies on temperature.^{9–11} Hudson¹² proposed that the rectifying effect is not necessarily due to dissimilar thermal conductivities in the two opposing directions, but rather due to interfacial thermal strain or warping.^{13–17} Other models of thermal rectification were formulated, which include (1) metal/insulator effect,¹⁸ (2) interfacial thermal potential barrier,¹⁹ (3) molecular mechanisms,^{20–23} (4) asymmetric nanostructure geometry,^{24–33} (5) phonon mismatch,³⁴ (6) one-dimensional anharmonic lattices,^{35–40} and (7) quantum

thermal rectification.⁴¹ A thorough review of these mechanisms and models was presented by Roberts.⁴²

Indeed thermal rectification is an interesting occurrence in many materials. But of what relevance and importance is it to technological advancement? In other words, does it have any realistic use? Its practicality is exemplified by recent reports of the excellent electrical performance^{43–48} of graphene and carbon nanotubes, triggering their stellar rise as potential materials in nanoelectronic devices. This is especially crucial and timely as silicon is reaching its performance limits and replacement materials have to be found. Other than the high electrical conductivity, their superior intrinsic thermal conductance^{49–55} enables thermal energy to be excavated quickly. Indeed the need for efficient thermal management is increasingly dire with more heat generation as device feature sizes are downscaled to accommodate a greater device packing density. In addition to thermal conductance, rectification is another essential aspect of thermal management, with the variation of the rate of thermal flow with direction, thermal energy can be directed to heat sinks and away from vital electronic components.

III. OBJECTIVES

This work on the phenomenon of thermal rectification is unique—it is a first observation and study of the reversal of the direction of thermal rectification. Both molecular dynamics (MD) simulations and theoretical formulation were performed to delineate this effect, and they were shown to be complementary. By building upon an existing model of thermal boundary resistance, namely the flux-mediated diffuse mismatch model (FMDMM),⁵⁶ this effect is explained and justified in an asymmetrically structured carbon nanotube (CNT). The asymmetry in the CNT was created by vacancy defects at one end of the tube, as in Refs. 32 and 33. Essentially, this paper aims to highlight the possible complication of the reversal of thermal rectification when structural defects such as vacancies are introduced to induce rectification.

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FIG. 1. Schematic of a carbon nanotube with a single vacancy cluster.

IV. SIMULATION DETAILS

Molecular dynamics simulations were carried out in this study. In the carbon nanotube with 1 vacancy cluster, 387 atoms were arranged in hexagonal rings and aligned in a tubular form to represent a (4,4) armchair single-walled carbon nanotube. The carbon nanotubes with 2 vacancy clusters consist of 374 atoms, and the clusters are positioned equidistant from either tube end, and opposite to one another in the same radial segment of the nanotube. Both types of carbon nanotubes are essentially similar in axial length (length ~ 61.6 Å). The single-vacancy-cluster nanotube is shown in Figure 1. The covalent carbon-carbon interactions were expressed by the Tersoff-Brenner interatomic potential.⁵⁷ The velocity Verlet algorithm was adopted to predict the next positions and velocities, with a timestep of 0.5 fs. The extreme-most ring of atoms in the nanotubes were fixated and the dynamics restricted by an adiabatic approximation. Nosé-Hoover^{58,59} thermostats were used at the ends of the system (exclusive of the “frozen” atoms) to create a temperature difference. The cold and hot thermostat temperatures were predetermined in the simulations, and an equilibration time of 2 ns is allowed for the atomic system to reach a steady-state. The thermal flux through the nanotube is defined to be the thermal energy passing through a unit area per unit time. By averaging the energy input into and out of the nanotube hot and cold thermostats, respectively, the thermal flux can be calculated.

V. RELATIONSHIP BETWEEN SCATTERER-ASYMMETRY-LOADING RATIO AND RECTIFICATION RATIO

To investigate the effect of the degree of structural asymmetry of the CNT on the extent of thermal rectification, we vary the position of the vacancy clusters in the nanotube. The structural asymmetry is defined by a scatterer-asymmetry-loading ratio η

$$\eta = \frac{l_{\text{scatt}}}{l_{0.5}}, \quad (1)$$

where l_{scatt} is the distance of the scatterer (in this context the vacancy cluster) from the midpoint along the axial direction of the nanotube, and $l_{0.5}$ is half the length of the nanotube. The thermal rectification ratio R is given by

$$R = \frac{J^+ - J^-}{J^-}, \quad (2)$$

where J^+ is the higher thermal flux from one end of the nanotube to the other, and J^- is the thermal flux of lower

magnitude. It is noted that these definitions of J^+ and J^- are different from those in other studies. In the latter, each of these fluxes is taken to be in a certain direction, while J^+ is still the larger flux. On the other hand, this work examines the reversal of thermal rectification, in which the larger flux does not have a fixed direction under different parameters. Therefore J^+ and J^- are not defined in directional terms, but rather distinguished by their magnitude. Furthermore, the hot and cold heat baths are defined to be

$$T_H = T_0(1 + \Delta), \quad (3)$$

$$T_C = T_0(1 - \Delta), \quad (4)$$

in which T_0 is the mean temperature of the nanotube and Δ is the normalized temperature difference.

Figure 2 shows that at a Δ of 0.667 and a T_0 of 300 K, as the scatterer-asymmetry-loading ratio increases from 0.491 to 0.593 for both vacancy types, the rectification ratio rises in magnitude. The rectification ratio remains relatively constant from $\eta = 0.389$ to 0.491, while it increases significantly from $\eta = 0.491$ to 0.593. In both cases of vacancy type (and in later presented data), our definition of a positive rectification ratio refers to a J^+ flowing from a non-defected end of the nanotube to a defected end, and a J^- in the opposite direction. The negative ratio is defined vice-versa. Succinctly, as the vacancy cluster is shifted away from the central position along the nanotube, i.e., the carbon nanotube becomes more asymmetric in its structure, it undergoes greater thermal rectification. However interestingly, a nanotube with a double vacancy cluster records a lower rectification ratio at all values of η .

VI. REVERSAL OF THERMAL RECTIFICATION

Fixing the cold bath temperature T_C at 100 K and changing the normalized temperature difference Δ , the change in rectification ratio is recorded (Figure 3). The rectification ratio decreases from a positive value to a negative one. The first point on the left is taken at a hot bath temperature of 500 K while the rightmost is at a hot bath temperature of 3000 K.

FIG. 2. Relationship between rectification ratio and scatterer-loading-asymmetry ratio for 1 vacancy and 2 vacancies, at a normalized temperature difference of 0.667 and a mean temperature of 300 K.

FIG. 3. Relationship between rectification ratio and normalized temperature difference, with a fixed cold bath temperature of 100 K.

This reversal of thermal rectification, or a switch in direction of J^+ and J^- , suggests that the rectifying direction is not absolute. The stronger thermal flux can reverse its direction towards or away from the vacancy clusters (Figure 4). With a constant cold bath temperature and a varying hot bath temperature T_H , the normalized temperature difference is changed. This is fundamentally analogous to pseudo-realistic situations whereby the heat production sites (hot end) in nanoelectronic devices have a non-constant temperature, while the base plate is the cold end of an infinite heat sink.

VII. FLUX-MEDIATED DIFFUSE MISMATCH MODEL

Following a previous work on the FMDMM,⁵⁶ the phenomenon of thermal rectification reversal is mathematically analysed and described in details. First, we define medium 1 (M1) to be the region of the nanotube from the non-defected

FIG. 4. Schematic of the phenomenon of thermal rectification reversal; stronger thermal flux (a) towards vacancy clusters and (b) away from vacancy clusters.

end to the defect/scatterer (vacancy clusters) and medium 2 (M2) to be the region from the defected end to the defect/scatterer. This is illustrated in Figure 5. $l_{(1)}$, $l_{(2)}$, $l_{(3)}$, and $l_{(4)}$ are regions of investigation and are small compared to the length of the nanotube. M2 is defined to be at the extreme end of the nanotube, and the length of M2 is much smaller than that of M1, i.e., $l_{M2} \ll l_{M1}$. The red arrows refer to the phonon flux flowing from the adjacent heat bath towards the other end, the blue arrows refer to the reflected phonon flux, and the green arrows refer to the transmitted phonon flux. Two scenarios will be described below.

A. Equal transmission coefficients

In this first case, an assumption is made such that the transmission coefficient from a non-defected end to a defected end is equal to that from a defected end to a non-defected end

$$\alpha_{12,(1)(2)} = \alpha_{12,(3)(4)}, \quad (5)$$

whereby $\alpha_{ij,(a)(b)}$ is the transmission coefficient while the subscripts ij refers to the flux from medium i to medium j , and $(a)(b)$ refers to the respective case of investigation, e.g., (1)(2) corresponds to regions (1) and (2), as portrayed in Figure 5(a).

By the nature of diffuse scattering,^{60,61} $\alpha_{12}(\omega, T) + r_{12}(\omega, T) = 1$, and $r_{12}(\omega, T) = \alpha_{21}(\omega, T)$, where $r_{12}(\omega, T)$ is the reflection probability. From the flux-mediated diffuse mismatch model, the flux at the four regions of investigation is as follow:

$$(1) \quad J_H - \beta_1^2 r_{12} J_H - \beta_1 \alpha_{21} J_C = [1 - \beta_1^2 (1 - \alpha_{12})] J_H + \beta_1 (\alpha_{12} - 1) J_C, \quad (6a)$$

$$(2) \quad -J_C + r_{21} J_C + \beta_1 \alpha_{12} J_H = \beta_1 \alpha_{12} J_H + (\alpha_{12} - 1) J_C, \quad (6b)$$

FIG. 5. Schematic to explain the phenomenon of thermal rectification reversal using FMDMM; (a) cold end closer to scatterer and (b) hot end closer to scatterer.

$$(3) -J_C + \beta_1^2 r_{12} J_C + \beta_1 \alpha_{21} J_H = -\beta_1 (\alpha_{12} - 1) J_H - [1 - \beta_1^2 (1 - \alpha_{12})] J_C, \quad (6c)$$

$$(4) J_H - r_{21} J_H - \beta_1 \alpha_{12} J_C = -(\alpha_{12} - 1) J_H - \beta_1 \alpha_{12} J_C, \quad (6d)$$

where J_H is the flux from the hot bath, J_C is the flux from the cold bath, r_{ij} refers to the reflection coefficient from medium i to medium j , and β_1 is the attenuating coefficient in medium 1, and a larger value represents less attenuation. It is assumed that $l_{(2)}$ is small enough such that the flux does not attenuate from the adjacent heat bath to the scatterer. The attenuation arises because phonons encounter Umklapp events before they reach the scatterer. This major assumption of FMDMM⁵⁶ indicates that the flux from one end is attenuated by a factor of β at the interface (in this case a defect). In expressions (6a) and (6c), the second term contains a square of β_1 because of the doubly attenuation of the flux as it flows to and from the heat bath and the scatterer.

To explore the possibility in which the flux in the configuration in Figure 5(a) is less than that in Figure 5(b), we start by comparing expressions (6a) and (6c) and assuming

$$\text{for } \beta_1 (\alpha_{12} - 1) < -\beta_1 [\beta_1 (\alpha_{12} - 1)] - 1, \quad \alpha_{12} < 1 - \frac{1}{\beta_1 (\beta_1 + 1)}, \quad (7)$$

$$\text{for } 1 + \beta_1 [\beta_1 (\alpha_{12} - 1)] < -\beta_1 (\alpha_{12} - 1), \quad \alpha_{12} < 1 - \frac{1}{\beta_1 (\beta_1 + 1)}. \quad (8)$$

Therefore if

$$\alpha_{12} < 1 - \frac{1}{\beta_1 (\beta_1 + 1)},$$

the magnitude of the thermal flux at region 1 is smaller than that at region 3. Similarly, comparing expressions (6b) and (6d),

$$\text{for } \beta_1 \alpha_{12} < 1 - \alpha_{12}, \quad \alpha_{12} < \frac{1}{\beta_1 + 1}, \quad (9)$$

$$\text{for } \alpha_{12} - 1 < -\beta_1 \alpha_{12}, \quad \alpha_{12} < \frac{1}{\beta_1 + 1}. \quad (10)$$

Thus if

$$\alpha_{12} < \frac{1}{\beta_1 + 1},$$

the magnitude of the thermal flux at region 2 is smaller than that at region 4. Akin to the averaging of the thermal flux into and out of the heat baths in molecular dynamics simula-

tions, the sum of the fluxes in regions 1 and 2 is compared to that in regions 3 and 4. As

$$1 - \frac{1}{\beta_1 (\beta_1 + 1)} - \frac{1}{\beta_1 + 1} = 1 - \frac{1}{\beta_1} > 0$$

and if the condition

$$\alpha_{12} < \frac{1}{\beta_1 + 1} \quad (11)$$

is fulfilled, the sum of the fluxes in regions 1 and 2 is less than that in regions 3 and 4. Otherwise, the reverse is true. This connotes a possible occurrence of thermal rectification reversal. When phonon Umklapp processes become more prominent, β_1 decreases. The range of values whereby condition (11) is satisfied extends, hence the probability of thermal rectification reversal increases. This rationalizes the reversal of rectifying direction in the 100–3000 K case, relative to the 100–500 K case (Figure 3), as U-processes are more pronounced in the former.

B. Dissimilar transmission coefficients

However the above model is true only provided the transmission coefficients $\alpha_{12,(1)(2)}$ and $\alpha_{12,(3)(4)}$ are equal. It is highly probable that the components $\beta_1 (\sum_j \int_{\omega_c} \hbar \omega \nu_{1,j} g_{1,j} f_1 d\omega)$ and $\beta_2 (\sum_j \int_{\omega_c} \hbar \omega \nu_{2,j} g_{2,j} f_2 d\omega)$ which constitute the transmission coefficient⁵⁶ are disparate in both cases of Figure 5, and therefore the transmission coefficients $\alpha_{12,(1)(2)}$ and $\alpha_{12,(3)(4)}$ are dissimilar. This difference can be represented by a moderation of $\alpha_{12,(1)(2)}$ by a factor of γ_M , with values of γ_M less than or more than 1, depending on which transmission coefficient is larger. This factor is a parameter that embodies changes to phonon velocities and populations. Cognate to the variations of diffuse mismatch model, including FMDMM, $\alpha_{12,(1)(2)} = \alpha_{21,(3)(4)}$. The fluxes at the four regions are

$$(1) \left[1 - \beta_1^2 \left(1 - \frac{\alpha_{12}}{\gamma_M} \right) \right] J_H + \beta_1 \left(\frac{\alpha_{12}}{\gamma_M} - 1 \right) J_C, \quad (12a)$$

$$(2) \beta_1 \frac{\alpha_{12}}{\gamma_M} J_H + \left(\frac{\alpha_{12}}{\gamma_M} - 1 \right) J_C, \quad (12b)$$

$$(3) -\beta_1 \left(\frac{\alpha_{12} - 1}{\gamma_M} \right) J_H - \left[1 - \beta_1^2 \left(\frac{1 - \alpha_{12}}{\gamma_M} \right) \right] J_C, \quad (12c)$$

$$(4) -\left(\frac{\alpha_{12} - 1}{\gamma_M} \right) J_H - \beta_1 \left(1 - \frac{1 - \alpha_{12}}{\gamma_M} \right) J_C. \quad (12d)$$

Comparing expressions (12a) and (12c),

$$\text{for } \beta_1 \left(\frac{\alpha_{12}}{\gamma_M} - 1 \right) < -\beta_1 \left[\beta_1 \left(\frac{\alpha_{12} - 1}{\gamma_M} \right) \right] - 1, \quad \alpha_{12} < \frac{(\beta_1 - 1)\gamma_M + \beta_1^2}{\beta_1 (\beta_1 + 1)}, \quad (13)$$

$$\text{for } 1 - \beta_1^2 \left(1 - \frac{\alpha_{12}}{\gamma_M}\right) < -\beta_1 \left(\frac{\alpha_{12} - 1}{\gamma_M}\right),$$

$$\alpha_{12} < \frac{(\beta_1^2 - 1)\gamma_M + \beta_1}{\beta_1(\beta_1 + 1)}, \quad (14)$$

as

$$\frac{(\beta_1 - 1)\gamma_M + \beta_1^2}{\beta_1(\beta_1 + 1)} - \frac{(\beta_1^2 - 1)\gamma_M + \beta_1}{\beta_1(\beta_1 + 1)} = \frac{\beta_1(1 - \beta_1)(\gamma_M - 1)}{\beta_1(\beta_1 + 1)}.$$

The above term can be (1) positive if $\gamma_M > 1$ or (2) negative if $\gamma_M < 1$. In the first case, if the condition

$$\alpha_{12} < \frac{(\beta_1^2 - 1)\gamma_M + \beta_1}{\beta_1(\beta_1 + 1)} \quad (15)$$

is fulfilled, the flux at region 1 is less than that at region 3. On the other hand, in the second case, if the condition

$$\alpha_{12} < \frac{(\beta_1 - 1)\gamma_M + \beta_1^2}{\beta_1(\beta_1 + 1)} \quad (16)$$

is satisfied, region 1 has a smaller flux. Conversely for both cases, the flux at region 3 is greater.

Next, comparing expressions (12b) and (12d),

$$\text{for } \beta_1 \frac{\alpha_{12}}{\gamma_M} < -\left(\frac{\alpha_{12} - 1}{\gamma_M}\right), \quad \alpha_{12} < \frac{1}{\beta_1 + 1}, \quad (17)$$

$$\text{for } \frac{\alpha_{12}}{\gamma_M} - 1 < -\beta_1 \left(1 - \frac{1 - \alpha_{12}}{\gamma_M}\right),$$

$$\alpha_{12} < \frac{\beta_1 + \gamma_M(1 - \beta_1)}{\beta_1 + 1}. \quad (18)$$

Again, the magnitude of these equalities is compared,

$$\frac{1}{\beta_1 + 1} - \frac{\beta_1 + \gamma_M(1 - \beta_1)}{\beta_1 + 1} = \frac{(\beta_1 - 1)(\gamma_M - 1)}{\beta_1 + 1}.$$

As before, a value of $\gamma_M > 1$ gives a smaller flux at region 2 (with the above expression negative) when

$$\alpha_{12} < \frac{1}{\beta_1 + 1}. \quad (19)$$

A value of $\gamma_M < 1$ sets the equality to be

$$\alpha_{12} < \frac{\beta_1 + \gamma_M(1 - \beta_1)}{\beta_1 + 1}. \quad (20)$$

When $\gamma_M > 1$, with Eqs. (15) and (19),

$$\frac{(\beta_1^2 - 1)\gamma_M + \beta_1}{\beta_1(\beta_1 + 1)} - \frac{1}{\beta_1 + 1} = \gamma_M \left(1 - \frac{1}{\beta_1}\right), \quad (21)$$

which is always negative. Thus a combination of the two inequalities results in the condition of

$$\alpha_{12} < \frac{(\beta_1^2 - 1)\gamma_M + \beta_1}{\beta_1(\beta_1 + 1)}, \quad (22)$$

which gives a sum of the flux at regions 1 and 2 to be lower than that at regions 3 and 4. Since the transmission coefficient is always positive, the term has to be greater than zero

$$\frac{(\beta_1^2 - 1)\gamma_M + \beta_1}{\beta_1(\beta_1 + 1)} \geq 0, \quad x \leq \frac{\beta_1}{1 - \beta_1^2}. \quad (23)$$

With $\gamma_M > 1$, the additional condition applies

$$\frac{\beta_1}{1 - \beta_1^2} > 1,$$

$$\beta_1 > 0.618.$$

For the case of $\gamma_M < 1$, the mathematical difference of Eqs. (16) and (20) gives the same eventual negative expression of (21). Hence

$$\alpha_{12} < \frac{(\beta_1 - 1)\gamma_M + \beta_1^2}{\beta_1(\beta_1 + 1)} \quad (24)$$

results in a smaller flux in the scenario in Figure 5(a). To sum it up, if $\gamma_M > 1$, condition (22) with $\beta_1 > 0.618$ gives a negative rectification ratio; if $\gamma_M < 1$, condition (24) is the determinant.

The above rigorous analysis suggests the dependence of the rectifying direction on a few variables—the attenuation coefficient β_1 , the moderating factor γ_M , and the transmission coefficient α_{12} . The attenuation coefficient relates to phonon dynamics along the nanotube, while the moderating factor and transmission coefficient represent the interfacial phononic conditions at the scatterer/defect. In the case that β_1 decreases due to momentum-destroying Umklapp events, there will be greater dissimilitude between the transmission coefficients, since medium 2 is taken to be small. If $\gamma_M > 1$, that dissimilitude is depicted by an increase in γ_M . The range of values in Eq. (22)

$$\frac{(\beta_1^2 - 1)\gamma_M + \beta_1}{\beta_1(\beta_1 + 1)} = \frac{1}{\beta_1 + 1} + \frac{(\beta_1 - 1)\gamma_M}{\beta_1}$$

may extend or contract; hence, the probability of thermal rectification reversal is dependent on the exact values of β_1 and γ_M .

If $\gamma_M < 1$, a decrease of γ_M signifies a larger discrepancy between the transmission coefficients. With a smaller β_1 , the change in the inequality range of Eq. (24)

$$\frac{(\beta_1 - 1)\gamma_M + \beta_1^2}{\beta_1(\beta_1 + 1)} = 1 + \frac{2\gamma_M - 1}{\beta_1 + 1} - \frac{\gamma_M}{\beta_1}$$

hinges on the values of β_1 , γ_M , and α_{12} , which dictate the rectifying direction, i.e., the occurrence of thermal rectification reversal.

The conditions for both cases of γ_M are attainable within the limits of the variables, e.g., $0 \leq \beta_1 \leq 1$ and $0 \leq \alpha_{12} \leq 1$.

VIII. CONCLUSION

To sustain the electrical performance in nanoelectronic applications, efficacious thermal management is imperative. Not only do we need good thermal-conducting materials at critical hotspots, it is vital that thermal dissipation is directed appropriately. This paper utilizes a dual approach to study the phenomenon of thermal rectification in carbon nanotubes, and a unique effect of the reversal of rectification under some conditions. Molecular dynamics simulations demonstrate the correlation between the position of vacancy defects (which defines the asymmetry of the structure) and the rectification ratio, and dissociate the direct dependence of the rectification ratio on the size of the defects. The reversal of rectifying direction is observed as the normalized temperature difference of the heat baths is varied. A mathematical approach, an extension of the flux-mediated diffuse mismatch model,⁵⁶ gives an account of phononic parameters and their range of values under which thermal rectification is possible (and vice-versa).

ACKNOWLEDGMENTS

The codes used in this work are developed from a code provided by the Maruyama Group, the University of Tokyo. G. C. Loh was supported by the Nanyang Research Scholarship, and the work funded by the MOE Tier 2 Fund.

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