Physics-Based Thermal Conductivity Model for Metallic Single-Walled Carbon Nanotube Interconnects

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***Abstract*—In this letter, a closed-form analytical model for temperature-dependent longitudinal diffusive lattice thermal conductivity (κ) of a metallic single-walled carbon nanotube (SWCNT) has been addressed. Based on the Debye theory, the second-order three-phonon Umklapp, mass difference (MD), and boundary scatterings have been incorporated to formulate κ in both low- and high-temperature regimes. It is proposed that κ at low temperature (T) follows the T^3 law and is independent of the second-order three-phonon Umklapp and MD scatterings. The form factor due to MD scattering also plays a key role in the significant variation of κ in addition to the SWCNT length. The present diameter-independent model of κ agrees well with the available experimental data on suspended intrinsic metallic SWCNTs over a wide range of temperature and can be carried forward for electrothermal analyses of CNT-based interconnects.**

***Index Terms*—Interconnects, nanotubes, phonons, thermal conductivity.**

I. Introduction

SINGLE-WALLED carbon nanotube (SWCNT) bundles have recently gained the potential to overcome electrothermal management issues of copper interconnect [1]. Although there are few recent progress in the study of electrothermal transport on both substrate and suspended intrinsic metallic SWCNT interconnects [2]–[4], phonon-assisted thermal conductivity is assumed to be either temperature independent or possesses an empirical dependency [1]–[4]. Unlike aluminum and copper which are conventional interconnect materials, the experimental observations and molecular dynamic simulations on the metallic SWCNT exhibit a wide variation of the thermal conductivity with temperatures ranging from hundreds to thousands of $\mathrm{Wm^{-1}K^{-1}}$ [5]. However, from these studies, the possible effects of various important scatterings on the thermal conductivity of the metallic SWCNTs which is a phonon-dominated phenomenon are not clear [4]. Due to these facts, to carry out an efficient electrothermal analysis of integrated circuits incorporating these emerging interconnects requires a closed-form analytical model of temperature and length-dependent thermal conductivity of this material.

In this letter, a physics-based model for diffusive longitudinal phonon-assisted thermal conductivity (κ) of metallic SWCNTs has been addressed. The Debye lattice theory has been used to formulate κ based on three major phonon scattering phenomena, namely, the second-order three-phonon Umklapp, mass difference (MD), and boundary scatterings, and to demonstrate that not only the length but also the form factor or the strength of MD scattering play a key role in the wide variation of κ. A closed-form analytical solution of diameter-independent κ at both low and high temperatures has been modeled and compared with available experimental data by other groups. This model stands valid for interconnects whose length is more than the phonon mean free path which is in the range of 180 nm at room temperature [6].

II. Model Development

The realization of the second-order three-phonon process scattering rate $\tau_U^{-1} = (32/27)\gamma^4(k_BT/Mv_g^2)^2\omega_B$ [7], the MD scattering rate $\tau_M^{-1} = (V_0\Gamma_m/4\pi v_g^3)\omega^4$ [8], and the purely diffusive boundary phonon scattering rate $\tau_B^{-1} = (v_g/L)$ [6] in nanotubes leads to the resultant scattering rate (τ_R^{-1}) by Matthiessen's rule: $\tau_R^{-1} = \tau_U^{-1} + \tau_M^{-1} + \tau_B^{-1}$. The parameters γ, ω_B, M, $V_0 (= h_t R_0^2)$, Γ_m, and v_g are the Gruneisen parameter, phonon branch frequency at the zone boundary, mass of the carbon atom, volume per atom, form factor, and average phonon group velocity, respectively. T, ω, L, h_t, and R_0 are the temperature, phonon frequency, SWCNT length, effective wall thickness, and the in-plane interatomic distance, respectively. With the aforementioned definitions, the phonon-assisted κ can now be written following the Callaway's model [8] as

$$\kappa = \frac{k_B}{2\pi^2 v_g}\left(\frac{k_BT}{\hbar}\right)^3 \int_0^{\frac{\theta_D}{T}} \frac{\tau_R x^4 e^x}{(e^x-1)^2}dx \tag{1}$$

in which $\tau_R = (Dx^4 + ET^2 + (v_g/L))^{-1}$, with $D = (V_0\Gamma_m/4\pi v_g^3)(k_BT/\hbar)^4$, $E = (32/27)\gamma^4(k_B/Mv_g^2)^2\omega_B$, $x = \hbar\omega/k_BT$, and θ_D is the Debye temperature. The integrand in (1) can be easily solved now at two temperature regimes viz., $T \to 0$ and $T \to \infty$. In the first case, (1) reduces to

$$\kappa_{\mathrm{Low}} = A(T)\left(\frac{4\pi^4}{15}\right) \tag{2}$$

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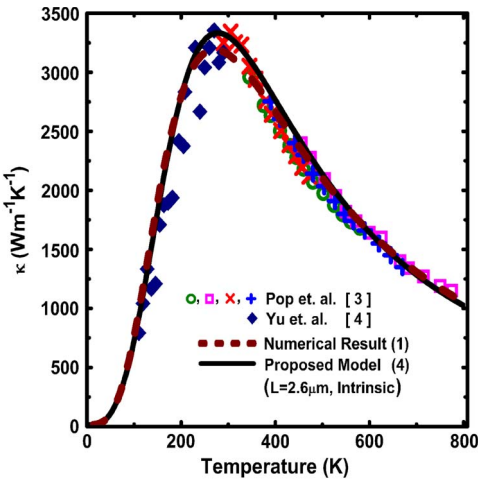

Fig. 1. κ as a function of temperature for intrinsic metallic SWCNT. The solid and dash curves correspond to the present model (4) and the numerical result of (1). The symbols are the experimental data from [3] and [4].

where, $A(T) = (k_B/2\pi^2 v_g)(k_B T/\hbar)^3 (ET^2 + (v_g/L))^{-1}$. In the second case, (1) assumes the form

$$\kappa_{\text{High}} = \frac{k_B}{2\pi^2 v_g}\left(\frac{k_B T}{\hbar}\right)^3 \left[4\sqrt{2}(aD^3)^{\frac{1}{4}}\right]^{-1} B(T, \Gamma_m) \quad (3)$$

where $B(T,\Gamma_m) = \ln[(\sqrt{D}(\theta_D/T)^2 + \sqrt{a} - \sqrt{2}(aD)^{1/4} \times (\theta_D/T))/(\sqrt{D}(\theta_D/T)^2 + \sqrt{a} + \sqrt{2}(aD)^{1/4}(\theta_D/T))] + 2\tan^{-1}[\sqrt{2}\theta_D(D/a)^{1/4}/T(1-(D/a)^{1/2}(\theta_D/T)^2)]$, in which $a = ET^2 + (v_g/L)$. In the case of intrinsic SWCNTs, $\kappa_{\text{High}} = (1/3)A(T)(\theta_D/T)^3$. Thus, using (2) and (3), κ can then be modeled as

$$\kappa^{-1} = \kappa_{\text{Low}}^{-1} + \kappa_{\text{High}}^{-1}. \quad (4)$$

Fig. 1 exhibits the variation of κ as a function of temperature for the spectrum constants $\theta_D = 1000$ K [9], $v_g = 10^4$ ms^{-1} [4], $h_t = 0.335$ nm [10], $R_0 = 0.14$ nm, and $\gamma = 1.24$ [11] for intrinsic metallic SWCNTs having a length of 2.6 μm.

It can be seen that the relative error between the present diameter-independent model (4) and the numerical result of (1) near both the temperature limits vanishes, while around room temperature, this error is less than 3%. In general, the value of the function $x^2 e^x/(e^x - 1)^2$ at high temperatures can be approximated to unity, and to correlate with the experimental data, the parameter ω_B has been taken to be 28 GHz. The approach used here may be also applicable to semiconducting SWCNTs only when the effect of acoustic electron–phonon $(e-p)$ scattering is ignored. Careful experimental measurements can depict whether the $e-p$ scattering rate should be considered or not as compared with the Umklapp and normal phonon scattering processes in semiconducting tubes for the determination of overall thermal conductivity. However, in the case of low-temperature zone, the present model can serve for both metallic and semiconducting SWCNTs. One of the fundamental assumptions underlying this derivation is the negligence of the phonon dispersion relation for the determination of phonon group velocity for a particular mode in the Umklapp process. Although this is important, τ_U is normally derived ignoring this consideration [4], [12]. It is worth noting that this assumption is valid only for long-wavelength phonons (acoustic), due to the small group velocity optical phonons that are not effective in transporting heat energy. It is not worthwhile to try to calculate κ for the precise phonon frequency spectrum and dispersion curve of a solid material. To obtain a closed-form expression, one should use the assumptions of the Debye theory, where v_g is used to replace the precise frequency-dependent phonon velocity for all the phonon branches together with the same phonon velocities for all polarizations.

III. Results and Discussions

A consistent result is exhibited by the present diameter-independent model as shown in Fig. 1 when compared with the experimental data by Pop *et al.* [3] and Yu *et al.* [4] for suspended intrinsic SWCNTs. The diameter dependency on κ comes through τ_B^{-1} which is given by $2v_g/L_{\text{eff}}F$ [13], in which F is a geometric factor and L_{eff} is a function of lateral and longitudinal lengths. For the present case, where the phonon transport is purely diffusive, τ_B^{-1} will mainly depend on the axial length of the tube [6], and hence, κ becomes independent of diameter. However, if the SWCNT is in contact with other tubes or substrate, the phonons may suffer both transmission and/or reflection along the diametrical axis also. In such a case, the SWCNT and the other tubes or substrate may act as a total composite material, where lateral dimension of both the individual material counts, and hence, to the first approximation, τ_B^{-1} should be modified. Thus, using these methods and principles, we propose a diameter-independent model of κ for suspended SWCNTs. In other way round for the present case, it is the thermal conductance (G) which should depend on the diameter and not κ. Thus, in the equation, $\kappa = GL/A$, where A is the cross-sectional area of the tube; the diameter is taken as an adjustable parameter to extract the value of the κ from the known values of G [4], so that it complies with the experimental data of [3] and our model around 300 K. In Fig. 1, we find that the low-temperature data of [4] also matches with our model if the diameter is 2.4 nm, which is also well within the spectrum of 1–3 nm as stated in [4].

It is to be noted that in the lower temperature region, the curves tend to match the T^3 law. However, for a scattering-less transmission, κ can also exhibit a linear T-dependence [14]. The physical condition for such a case is the adiabatic contact between the system and the heat baths, making the phonon transmission-coefficient unity together with the assumption of an approximate linear response of the difference in the temperature of the heat baths. In case of purely diffusive phonon transport, the effect of second-order, three-phonon, and mass-difference scatterings does not tend to dominate at low temperatures. However, as T increases, the effect of second-order three-phonon process dominates over the boundary scattering, and thus, κ follows a $1/T^2$ law.

Fig. 2 shows the variations of κ with temperature and length as functions of different form factors, respectively. From Fig. 2(a), it is seen that, at room temperature, κ increases from 3500 to 4800 Wm^{-1}K^{-1} for a length variation of 3–12 μm, respectively, for intrinsic SWCNT. Fig. 2(b) shows the variation

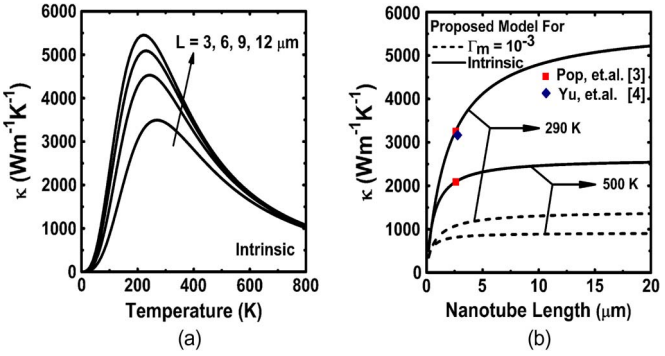

Fig. 2. κ as a function of (a) temperature for different lengths and (b) length for different temperatures. The symbols are the experimental data from [3] and [4].

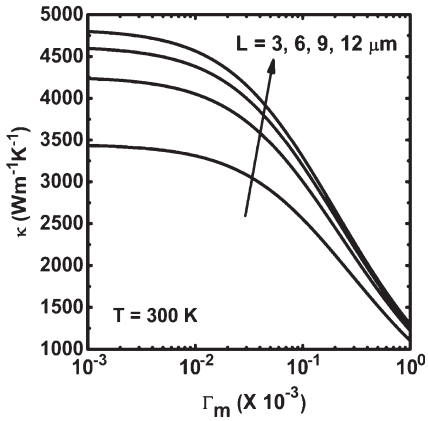

Fig. 3. κ as a function of Γ_m for different nanotube lengths at room temperature.

of κ as a function of nanotube length for an intrinsic case and $\Gamma_m = 10^{-3}$ at different temperatures; this has been compared to the experimental data of Pop *et al.* [3] and Yu *et al.* [4]. Note that it takes longer length to saturate κ for the intrinsic case.

Fig. 3 shows the effect of Γ_m on κ at room temperature for different SWCNT lengths. It appears that κ decreases with an increase in Γ_m and can reach around 1000–1500 $\mathrm{Wm^{-1}K^{-1}}$ at 300 K for a length scale of 3–12 μm at $\Gamma_m = 10^{-3}$. As Γ_m decreases, κ reaches to their respective intrinsic value. However, as Γ_m increases, the phonons are strongly scattered more than the zone-boundary process; thus, κ tends to be independent of nanotube length. Also, when the impurity level is large and all phonon modes are strongly scattered by resistive processes, then the normal-scattering time becomes higher than the resultant scattering time. This effect has been ignored in this present model for an analytical solution. However, this is usually treated as a correction term and can generally be ignored [8].

At this point, it is to be noted that although the electrothermal transport analyses of an individual metallic SWCNT on oxide have been recently reported [2], the heat transport mechanism of their bundles as interconnects is not yet fully understood. It can be stated that κ is one of the key parameters to analyze the characteristic diffusive length, which in turn decides whether the interconnects are thermally long or short [15]. Also, apart from the formulation of κ, the lateral heat transport should also be modeled accurately in order to carry out a realistic electrothermal analysis of SWCNT interconnects.

IV. Conclusion

Using the second-order three-phonon Umklapp process, an analytical solution of κ in metallic SWCNTs has been formulated as a function of temperature, length, and strength of the MD scattering. At lower temperature, κ is dominated by the boundary scattering, whereas as temperature increases, it is the second-order three-phonon process which dominates κ. The present theoretical model agrees well with the experimental data at both low- and high-temperature zones, and the error is less than 3% at room temperature.

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