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Barbalinardo et al. Reply

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Barbalinardo *et al.* Reply: Comment [1] shows that the thermal conductivity (κ) of a (10,0) carbon nanotube (CNT), obtained by inversion of the linearized Boltzmann transport equation (BTE), may not converge when the third-order interatomic force constants (IFC) are computed analytically. Bruns *et al.* [1,2] showed that the lifetimes $\tau_{\mathrm{RTA}}(q)$ of the acoustic branches, computed in the relaxation time approximation (RTA), obey precise power laws for $q \to 0$, which are violated by the numerical IFCs in our Letter [3]. κ in Ref. [1] is much larger than that in Ref. [3], and does not converge even when 400 q-points are used to integrate the first Brillouin zone. These new results are correct and challenge the agreement between BTE (with classical statistics) and molecular dynamics (MD) proposed in Ref. [3], and the statement that κ is finite in the bulk limit.

Here we address the accuracy of finite-differences (FD) calculations, the convergence of κ for infinitely long CNTs, and $\kappa(L)$ for CNTs of length L. Bruns *et al.* shows that the long-wavelength trends of τ_{RTA} are disrupted by numerical errors introduced in the FD calculation of the IFCs resulting in nonsymmetrized force constant matrices. We computed IFCs by FD with displacements dx ranging from 10^{-6} to 10^{-3} Å, and the correct trends for τ_{RTA} of the flexural (FA), longitudinal (LA) and torsional (TW) acoustic modes at low q are recovered for $dx > 4 \times 10^{-5}$. Figure 1 shows that $\tau_{\mathrm{RTA}}(q)$ from numerical IFCs with $dx = 10^{-4}$ Å are nearly indistinguishable from those obtained from analytical IFCs (in Ref. [3] we used $dx > 4 \times 10^{-6}$ Å). This result is useful for future works for those cases in which it is not possible to compute IFCs analytically.

τ_{RTA} provides a compelling test of the IFC's accuracy but cannot be used to calculate κ of CNTs due to the predominance of hydrodynamic effects in low-dimensional materials [4]. We computed the lifetimes of the acoustic modes by directly inverting the scattering matrix (τ_{inv}), using both analytical and numerical IFCs [Fig. 1]. Because of the hydrodynamic effects, the values and low-q trends of τ_{inv} and τ_{RTA} are different. Fitting τ_{inv} for $q \to 0$ one may infer whether κ converges or diverges in the infinite-size limit. The modes that may lead to divergence are FA and TW, as their τ_{inv} diverge for $q \to 0$. The group velocity of the FA modes is proportional to q, and that of TW is constant; thus the onset of κ divergence is either $\tau_{\mathrm{FA}} \propto 1/q^3$ or $\tau_{\mathrm{TW}} \propto 1/q$. τ_{TW} has a similar low-q behavior as in RTA calculations, $\tau_{\mathrm{inv,TW}} \propto 1/q^{0.5}$. $\tau_{\mathrm{inv,FA}}$ from analytical IFCs diverges as $1/q^3$ or steeper, suggesting divergent κ. Conversely, using numerical IFCs we get a slower than $1/q^3$ trend, which would lead to finite κ. This calculation may be more representative of systems at finite temperature, possibly reconciling BTE with convergent κ in MD simulations. Calculations with finite temperature IFC renormalization and thicker q-point sampling will be necessary to prove the convergence of κ in CNTs.

Finally, we computed finite-length $\kappa(L)$ with analytical IFCs. We find that $\kappa(L)$ remains in agreement with our former calculations [3] up to $L = 1$ mm while it gets significantly larger for larger lengths (Fig. 2).

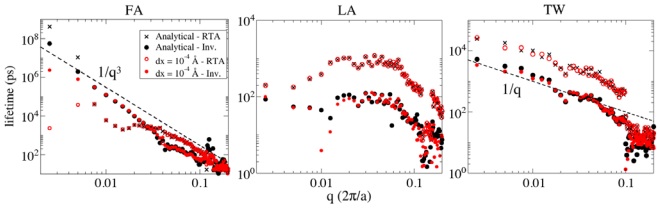

FIG. 1. Lifetimes of the FA, LA, and TW acoustic modes computed with quantum statistics. All the calculations are done by integrating the BTE over a uniform grid of 401 q points.

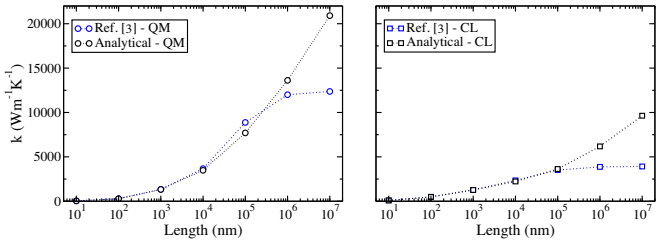

FIG. 2. Thermal conductivity as a function of length computed with analytical and the numerical IFCs from Ref. [3].

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