Full Band Monte Carlo Simulation of Thermal Transport Across Lateral Interface Between 2D Materials

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***Abstract*—We have presented a stochastic Monte Carlo algorithm parameterized with ab initio calculations to investigate the thermal transport across lateral interfaces between two-dimensional materials, with a focus on hexagonal boron nitride and Graphene heterostructures. Ab initio parameters are incorporated into the Monte Carlo simulation, enhancing the accuracy of thermal properties. The interface thermal conductance, temperature profiles, response times, and modal contributions of phonon modes in all transport regimes are theoretically studied. This research significantly contributes to our understanding of thermal properties at the nanoscale, providing essential insights for electronics and energy management applications.**

Keywords— Monte Carlo method, density functional theory, 2D materials, lateral heterostructure, phonon transport

I. Introduction

In-plane heterostructures based on two-dimensional (2D) materials are emerging as promising candidates for various applications due to their unique electronic, mechanical, and thermal properties [1,2]. Amon those, 2D hexagonal boron nitride (h-BN), whose structure is very similar to graphene, is gaining importance revealing a wide range of applications [3,4]. In nanostructures, heat transport plays an essential role because it influences the performance, reliability, and efficiency as their dimensions are miniaturized to the nanoscale. Therefore, understanding thermal properties can lead to significant advancements not only in electronics and but also in energy management. Despite significant progress in thermal transport modeling beyond Fourier's law, a rigorous analysis of the nanostructures that fully captures nonlinear transport properties is still lacking. In other words, a comprehensive understanding of thermal transport across interface between 2D materials, particularly at lateral interfaces, has not been completely achieved in order to assess their performance potential [5,6].

Among several numerical methods to investigate the thermal properties such as the Molecular Dynamics (MD) [7,8] and Non Equilibrium Green's function approach (NEGF) [9,10], a particle Monte Carlo (MC) method is very efficient and intuitive since it stochastically solves the Boltzmann transport equation (BTE) for phonons [11,12]. It is relevant for all transport regimes even in the presence of interfaces and is capable of incorporating complex scattering terms in a wide temperature range.

In this work, we present a full-band Monte Carlo approach for phonon transport [13] and investigate the interface thermal conductance of in-plane heterostructure between 2D h-BN/Graphene based on ab initio parameters. The interface thermal conductance (ITC), temperature profiles, response times and modal contributions of phonon modes are analyzed in all types of phonon transport regimes.

II. Models

A. Full-band description of 2D h-BN

Phonon dispersions and scattering rates including isotope effect in the full Brillouin zone (see Fig. 1) have been computed via *ab initio* methods in the framework of the density functional theory (DFT) by using Quantum ESPRESSO software [14]. The term 'full-band' refers here to the inclusion of all phonon modes in the entire Brillouin zone. Ab initio methods enable the predictive power of quantum mechanics into the Monte Carlo simulation without using any empirical parameters. Calculated phonon dispersions for 2D h-BN have excellent agreement with experimental results [15]. The phonon scattering rates, i.e. the inverse of the phonon lifetimes, were calculated within the temperature range from 0 to 1000 K [16]. In contrast to bulk materials, momentum-conserving Normal scattering dominate over Umklapp scattering at all temperatures [17]. Furthermore, a strong phonon-isotope scattering is observed due to a substantial boron isotope mixture [18].

B. Monte Carlo simulation for phonons

To solve the BTE for phonons and describe the time evolution of the phonon distribution function, we used an in-house MC simulator. The trajectories of numerous simulated phonons are randomly chosen having different wave vectors and modes. After each free flight, phonons are scattered by

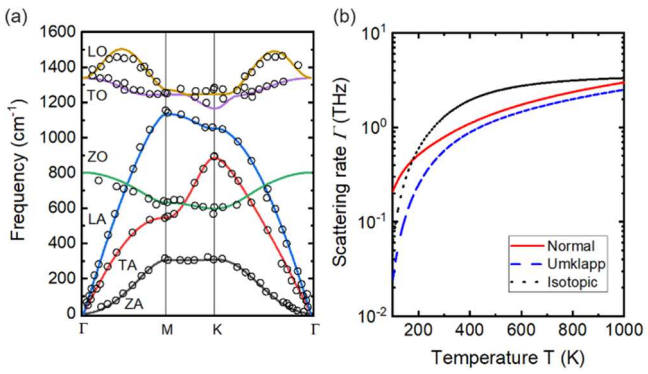

Fig. 1. (a) Calculated phonon dispersions for 2D h-BN along the high symmetry paths and experimental data [15]. (b) Phonon scattering rates of Normal, Umklapp and isotopic processes as a function of temperature computed via DFPT.

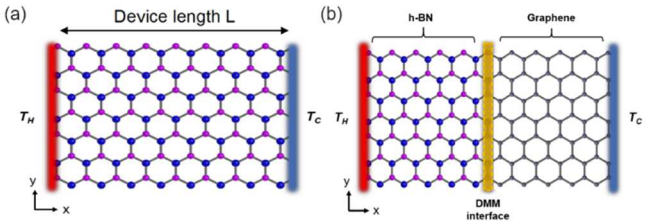

Fig. 2. Schema of the simulated (a) h-BN nanofilm and (b) h-BN/Graphene lateral heterostructure. Red/blue faces for hot/cold thermostats with T_H = 302 K, T_C = 298 K, respectively. Yellow zone indicates the DMM interface.

either a temperature-dependent phonon scattering or a collision on the boundaries. The implementation and the post-processing methods are presented in detail in [13].

Phonon transmission across the interface is implemented by using a full-band version of the Diffusive Mismatch Model (DMM) [19]. This semi-transparent interface assumes that all phonons at the interface undergo diffusive scattering thus losing the memory of their initial state, but conserving the phonon energy to satisfy the energy conservation.

The standard pseudo-temperature T at each position in the device are brought up to date from summing over the phonon distribution according to the equilibrium Bose-Einstein statistics. However, this standard temperature definition is unsuitable to treat the heat transfer through interface where it is expected to occur in strong out-of-equilibrium conditions. The directional temperatures T^+ and T^- are respectively defined in association with the phonon sub-populations possessing positive and negative velocities [20]. Using directional temperatures T^+ and T^- instead of standard pseudo-temperatures T allows for an accurate description of non-equilibrium conditions, thereby enabling the precise calculation of the ITC for the lateral heterostructure with an interface [19].

C. Investigated structures

Fig. 2 illustrates investigated homogeneous and heterogeneous 2D planes with a distance L between the two thermostats (hot and cold) connected at their respective edges. In the h-BN/Graphene lateral heterostructure, the diffusive interface is located in the middle of the structure (x=L/2) joining different 2D materials. The thermal flux is oriented to flow along the x-direction due to the positioning of thermostats at either end of the x-axis. Concurrently, it is assumed to extend infinitely in the y-direction for all structures. A finite rectangular mesh, comprised of cells with a length equal to or less than 1 nm, is used along the x-axis to numerically calculate the thermal properties of the devices.

III. Results

A. Thermal conductivity of 2D h-BN nanofilms

First, we examine the length-dependent thermal conductivity of 2D h-BN nanofilms at room temperature, as shown in Fig. 3. The Monte Carlo simulation results (red crosses) are compared with several analytical models detailed extensively in [19] and experimental values [21,22]. In ultra-short films, which are on the order of magnitude of the phonon mean free path, the thermal conductivity converges to the ballistic model (dotted line), derived from Landauer's formalism, that is linearly dependent on the device length L. As the length of the device increases, the calculated thermal conductivities move towards the diffusive value (dashed line) displaying an asymptotic behavior and align closely with the

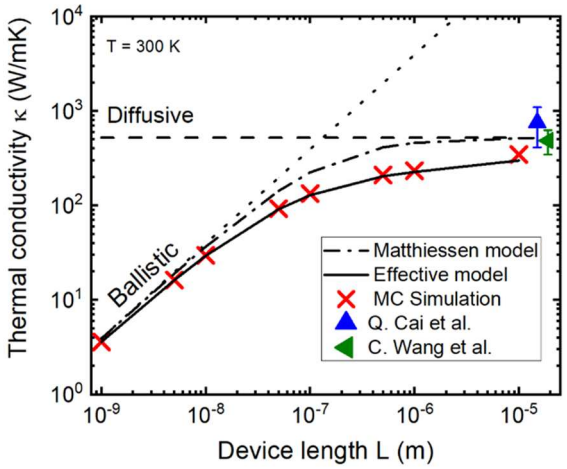

Fig. 3. Thermal conductivity as a function of the device length for 2D h-BN nanofilms at 300 K. Crosses represent the MC results. Lines stand for several analytical model results. Exp : Q. Cai et al. [21], C. Wang et al. [22].

experimental measurements for mono (blue triangle) and bilayer h-BN (green triangle).

As expected, the Matthiessen model (point-dashed line), which merely aggregates the ballistic and diffusive thermal resistances (the inverse of thermal conductivity), fails to accurately characterize the transition regime, also referred to as the quasi-ballistic regime. Moreover, the effective model (solid line), already validated for bulk silicon [13], demonstrates its utility in 2D device systems. Consequently, the MC results show good agreement across all phonon transport regimes, i.e. ballistic, quasi-ballistic, and diffusive, without using any fitting parameters.

B. 2D h-BN/Graphene lateral heterostructure

Fig. 4(a) displays the temperature profiles of the standard pseudo-temperature T (black), as well as the directional temperatures T^+ (red) and T^- (blue) in a 100 nm h-BN/Graphene heterostructure calculated by a MC simulation. In the heterostructure depicted in Fig. 2(b), the 2D h-BN is in contact with the hot thermostat, while the graphene touches the cold thermostat. All temperatures exhibit a sharp drop at the interface, indicating a significant thermal gradient.

The interface thermal conductance is commonly computed by dividing the heat flux density Q by the difference in the temperatures on each side of the interface, that is:

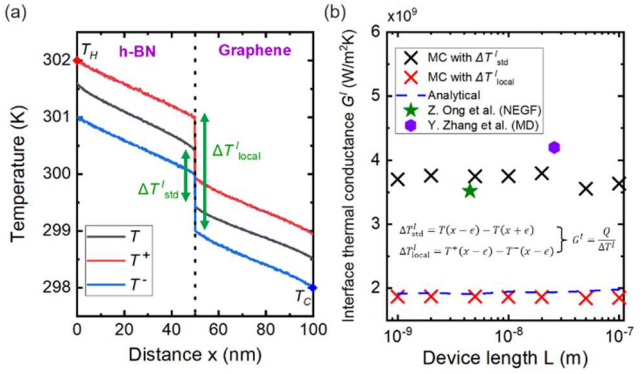

Fig. 4. (a) Temperature profiles of T, T^+ and T^- for a length h-BN/Graphene heterostructure with L = 100 nm. (b) Interface thermal conductance as a function of the device length calculated from MC results for 2 ITC definitions with other simulation methods (NEGF [23] and MD [24]).

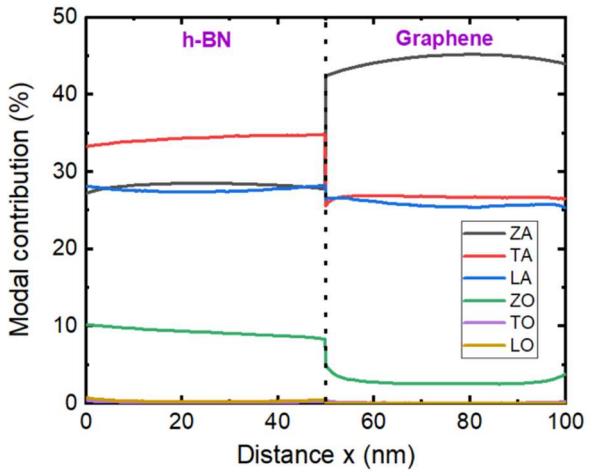

Fig. 5. Modal contributions of six phonon modes to the total heat flux density in a 100 nm long h-BN/Graphene heterostructure.

$$G^I = \frac{Q}{\Delta T^I} \tag{1}$$

According to the choice of temperatures, there are two different temperature drop at interface:

$$\Delta T^I_{\text{std}} = T(x-\epsilon) - T(x+\epsilon) \tag{2}$$

$$\Delta T^I_{\text{local}} = T^+(x-\epsilon) - T^-(x+\epsilon) \tag{3}$$

where ϵ is an infinitely short distance. Thus, these two quantities lead to the different interface thermal conductances plotted in Fig. 4(b). The ITC results obtained using the standard temperature difference ΔT^I_{std} (black crosses) are twice as large as those based on the directional temperatures $\Delta T^I_{\text{local}}$ (red crosses) for all device lengths. The results using $\Delta T^I_{\text{local}}$ are well matched with anaytical model [19] (blue dashed line) because the directional temperatures T^+ and T^- can consider the phonons that actually interact with the interface. Other simulation results using the ΔT^I_{std} for the ITC calculation showed similarly erroneous results far from the anaytical model [23,24].

Fig. 5 shows the modal contribution of the six phonon modes to the total heat flux density along the x-axis, distinguishing each h-BN and Graphene region in a 100 nm long h-BN/Graphene lateral heterostructure. The sum of the contributions of all modes is equal to 100 percent due to the conservation of the total heat flux. The modes are significantly perturbed when passing through the DMM interface, showing a pronounced curvature, except for the transverse and longitudinal optical ones because of their low group velocities. These local indicators of the transport regime reveal a strong non-equilibrium transport regime over a few nanometers around the interface.

Finally, in Fig. 6, the transient thermal response is investigated by analyzing the time evolution of both heat flux density and temperature at four distinct locations (x = 2.5 nm and 25 nm on the h-BN side, and x = 75 nm and 97.5 nm on the graphene side) within a 100 nm h-BN/Graphene heterostructure. The time step duration was set to 1 picoseconds (ps) with different thermostat temperatures (T_H = 400 K and T_C = 300 K) to fully capture the transient thermal response. On the h-BN side connected to a hot thermostat, both temperature and flux increase rapidly within

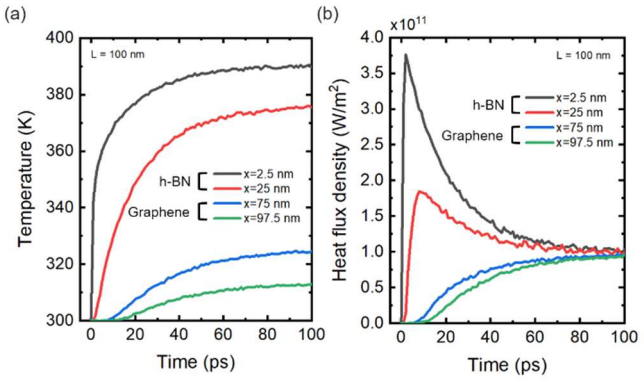

Fig. 6. The evolution of (a) temperature and (b) heat flux density as a function of time for h-BN/Graphene heterostructure of length 100 nm at four different positions along the x-axis.

a few ps, while the reaction on the other side is notably slower. Subsequently, the entire system stabilizes within approximately 100 ps maintaining constant values. Utilizing these results, the delay and the characteristic time of the thermal interface can be accurately estimated.

IV. Conclusion

A full-band Monte Carlo method, combined with ab initio parameters, has been presented to study the heat transport in two-dimensional materials, particularly h-BN/Graphene lateral heterostructures.

Firstly, in 2D h-BN nanofilms, the simulation results are in good agreement with analytical models and experimental measurements, ensuring coverage of all transport regimes. In h-BN/Graphene heterostructures, we have calculated the interface thermal conductance based on directional temperatures, consistently with the analytical formula. Furthermore, we have revealed the presence of a pronounced non-equilibrium transport regime within a few nanometers around the interface by examining the phonon mode contributions.

Finally, this research could serve as a robust foundation for further studies of complex nanostructures based on 2D materials.

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References

[1] J. Igo, M. Gabel, Z. G. Yu, L. Yang, and Y. Gu, "Photodefined In-Plane Heterostructures in Two-Dimensional In2Se3 Nanolayers for Ultrathin Photodiodes," *ACS Appl Nano Mater*, vol. 2, no. 10, pp. 6774–6782, Oct. 2019.

[2] J. Liang, X. Zhu, M. Chen, X. Duan, D. Li, and A. Pan, "Controlled Growth of Two-Dimensional Heterostructures: In-Plane Epitaxy or Vertical Stack," *Acc Mater Res*, vol. 3, no. 10, pp. 999–1010, Oct. 2022.

[3] X. K. Chen, Y. J. Zeng, and K. Q. Chen, "Thermal Transport in Two-Dimensional Heterostructures," *Front Mater*, vol. 7, p. 427, Dec. 2020.

[4] M. H. Kalantari and X. Zhang, “Thermal Transport in 2D Materials,” *Nanomaterials*, vol. 13, no. 1, p. 117, Dec. 2022.
[5] Q. V. Yu, K. Watanabe, T. Taniguchi, and L. H. Li, “Interfacial thermal conductance between atomically thin boron nitride and graphene,” *Nanoscale*, vol. 15, no. 1, pp. 122–126, Dec. 2023.
[6] J. Song *et al.*, “Effect of strain and defects on the thermal conductance of the graphene/hexagonal boron nitride interface,” *Physical Chemistry Chemical Physics*, vol. 22, no. 20, pp. 11537–11545, May 2020.
[7] E. S. Landry and A. J. H. McGaughey, “Thermal boundary resistance predictions from molecular dynamics simulations and theoretical calculations,” *Phys Rev B Condens Matter Mater Phys*, vol. 80, no. 16, p. 165304, Oct. 2009.
[8] S. Merabia and K. Termentzidis, “Thermal conductance at the interface between crystals using equilibrium and nonequilibrium molecular dynamics,” *Phys Rev B Condens Matter Mater Phys*, vol. 86, no. 9, p. 094303, Sep. 2012.
[9] N. Mingo, “Anharmonic phonon flow through molecular-sized junctions,” *Phys Rev B*, vol. 74, no. 12, p. 125402, Sep. 2006.
[10] S. Sadasivam, U. V. Waghmare, and T. S. Fisher, “Phonon-eigenspectrum-based formulation of the atomistic Green’s function method,” *Phys Rev B*, vol. 96, no. 17, p. 174302, Nov. 2017.
[11] J. P. M. Péraud and N. G. Hadjiconstantinou, “Efficient simulation of multidimensional phonon transport using energy-based variance-reduced Monte Carlo formulations,” *Phys Rev B Condens Matter Mater Phys*, vol. 84, no. 20, Nov. 2011.
[12] D. Lacroix, K. Joulain, and D. Lemonnier, “Monte Carlo transient phonon transport in silicon and germanium at nanoscales,” *Phys Rev B*, vol. 72, no. 6, p. 064305, Aug. 2005.
[13] B. Davier *et al.*, “Heat transfer in rough nanofilms and nanowires using full band ab initio Monte Carlo simulation,” *Journal of Physics Condensed Matter*, vol. 30, no. 49, Nov. 2018.
[14] P. Giannozzi *et al.*, “Advanced capabilities for materials modelling with Quantum ESPRESSO,” *Journal of Physics Condensed Matter*, vol. 29, no. 46, Oct. 2017.
[15] J. Serrano *et al.*, “Vibrational properties of hexagonal boron nitride: Inelastic X-ray scattering and Ab Initio calculations,” *Phys Rev Lett*, vol. 98, no. 9, Mar. 2007.
[16] L. Paulatto, F. Mauri, and M. Lazzeri, “Anharmonic properties from a generalized third-order ab initio approach: Theory and applications to graphite and graphene,” *Phys Rev B Condens Matter Mater Phys*, vol. 87, no. 21, Jun. 2013.
[17] A. Cepellotti, G. Fugallo, L. Paulatto, M. Lazzeri, F. Mauri, and N. Marzari, “Phonon hydrodynamics in two-dimensional materials,” *Nat Commun*, vol. 6, 2015.
[18] L. Lindsay and D. A. Broido, “Enhanced thermal conductivity and isotope effect in single-layer hexagonal boron nitride,” *Phys Rev B Condens Matter Mater Phys*, vol. 84, no. 15, Oct. 2011.
[19] B. Davier, P. Dollfus, N. D. Le, S. Volz, J. Shiomi, and J. Saint-Martin, “Revisiting thermal conductivity and interface conductance at the nanoscale,” *Int J Heat Mass Transf*, vol. 183, p. 122056, Feb. 2022.
[20] J. Maassen and M. Lundstrom, “Modeling ballistic effects in frequency-dependent transient thermal transport using diffusion equations,” *J Appl Phys*, vol. 119, no. 9, p. 95102, Mar. 2016.
[21] Q. Cai *et al.*, “High thermal conductivity of high-quality monolayer boron nitride and its thermal expansion,” *Sci Adv*, vol. 5, no. 6, Jun. 2019.
[22] C. Wang, J. Guo, L. Dong, A. Aiyiti, X. Xu, and B. Li, “Superior thermal conductivity in suspended bilayer hexagonal boron nitride,” *Sci Rep*, vol. 6, May 2016.
[23] Z. Y. Ong and G. Zhang, “Efficient approach for modeling phonon transmission probability in nanoscale interfacial thermal transport,” *Phys Rev B Condens Matter Mater Phys*, vol. 91, no. 17, May 2015.
[24] Y. Zhang, Q. X. Pei, C. M. Wang, C. Yang, and Y. W. Zhang, “Interfacial Thermal Conductance and Thermal Rectification of Hexagonal BCnN/Graphene In-Plane Heterojunctions,” *Journal of Physical Chemistry C*, vol. 122, no. 39, pp. 22783–22789, Oct. 2018.