Coherent thermal transport in isotopic graphene superlattices: a NEGF study

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ABSTRACT

We developed an in-house *ab initio* 2D Non-Equilibrium Green's Function (NEGF) code to study phonon transport in 2D materials. Based on this code, coherent phonon transport in isotopic ($^{12}C/^{13}C$) graphene superlattices is investigated.

INTRODUCTION

Nanoscale phononic crystals based on 2D materials provide a new opportunity to tune thermal properties using the wave nature of phonons. Constructive (resp. destructive) interferences could indeed induce high (resp. low) thermal conductivities [1].
In that context, atomistic Non-Equilibrium Green's Function (NEGF) approach is well suited, since it intrinsically describes the wave nature of thermal phonons by solving the atomic dynamical equation.

MODEL AND DISCUSSIONS

We extended our in-house NEGF code [2] to study 2D materials-based superlattices. The code solves the dynamical equation using the Retarded Green's function of phonons:

$$G^R(\omega,q_z) = [\omega^2 I - \tilde{\phi}(q_z) - \Sigma^R(\omega,q_z)]^{-1} \quad , \quad (1)$$

where ω denotes the phonon frequencies, I is the identity matrix, and q_z the wave vector along the z-axis, perpendicular to the transport direction. Σ^R represents the self-energies of the contacts while $\tilde{\phi}$ corresponds to the harmonic matrix, computed from the open-source packages Quantum ESPRESSO (QE) and PHONOPY (Figure 1).

Based on this method, we investigate the thermal properties of $^{12}C/^{13}C$ isotopic-superlattices of graphene (see Fig. 2). Since our initial NEGF code requires an orthogonal cell, we consider an orthorhombic unit cell whose resulting *ab initio* bandstructure is shown in Figure 3. Those results have been validated against phonon bandstructure considering a hexagonal unit cell (not shown).
Fig. 4, shows the thermal conductivity ratio γ ($=K_{isotope}/K_{pure}$) as a function of the central region length N for two superlattice (SL) period lengths M, where $K_{isotope}$ and K_{pure} are the thermal conductivity of SL and of pure ^{12}C graphene, respectively. We see that for a given N, *i)* the thermal conductivity of the SL is lower than the one of a pure ^{12}C graphene structure. The smaller the period, the more the thermal conductivity decreases. This expected trend is due to the interface scattering [2].

On the other hand, Figure 5 shows the ratio γ as a function of the SL period length M for three different central regions lengths N. It decreases with the SL period length until a critical value M=8. This is a signature of the wave-nature of phonon which overcomes the interface scattering when the SL period length is smaller than the dominant phonon wavelength. Similar phenomenon has also been found in carbon nanoribbons.

Finally, Figure 6 shows the spectral heat flows of pure graphene structure and SL at the critical length (M=8), where the interface scattering is maximum. We see that the heat flow decrease in SL is significant at 20 THz, which corresponds to a high-density mode in the bandstructure of Fig. 2.

Additional coherent effects will be discussed considering graded [2] and Golomb ruler SL.

REFERENCES

[1] Z. Zhang, *et al., Coherent thermal transport in nano-phononic crystals: An overview*, APL Materials **9** (8), 081102 (2021).
[2] Y. Guo, *et al., Thermal conductivity minimum of graded superlattices due to phonon localization*, APL Materials **9** (9), 091104 (2021).

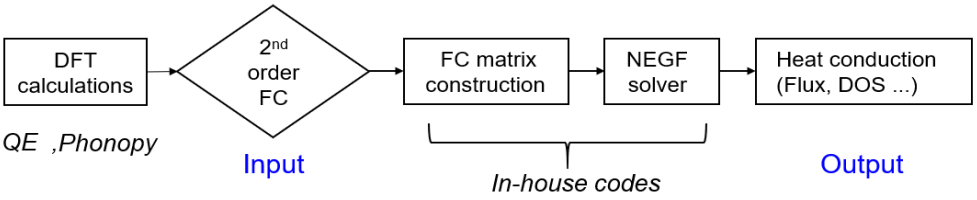

Fig. 1. Flowchart illustrating the structure of the 2D NEGF transport code.

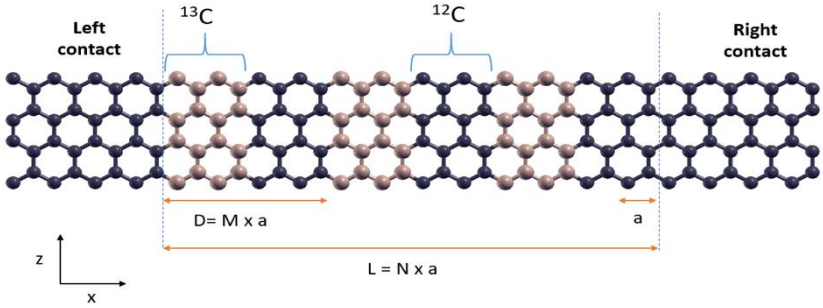

Fig. 2. Schematic of the considered graphene superlattice. The black and brown atoms represent the carbon 12 and 13 respectively, L is the length of the device, D is the period and a is the unit cell's lattice parameter.

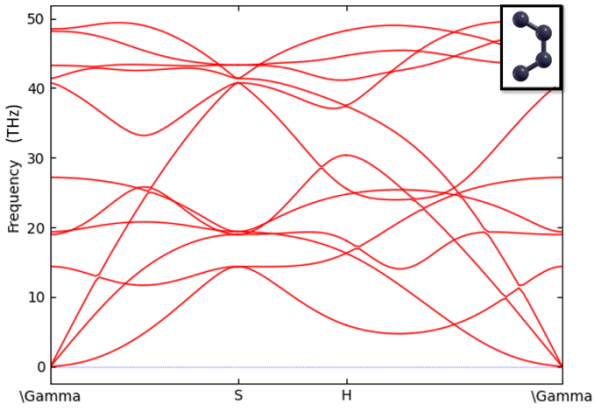

Fig. 3. DFT results of phonon bandstructure of graphene with an orthorhombic unit cell (shown in the top right corner).

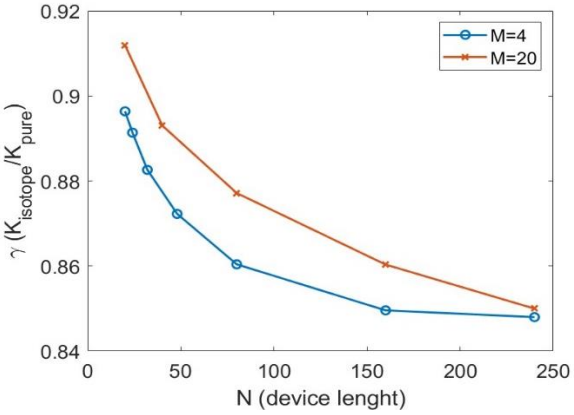

Fig. 4. Thermal conductivity ratio γ as a function of the device's length N for two superlattice period lengths M.

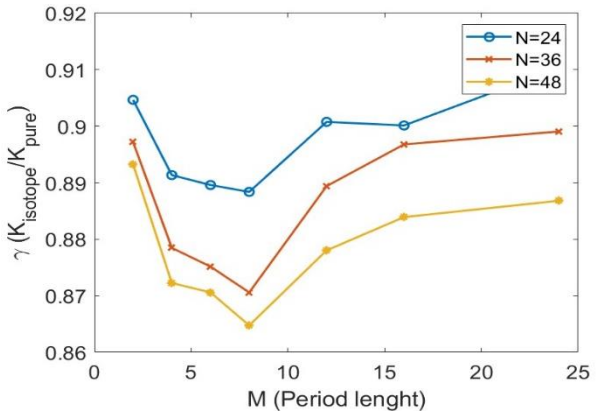

Fig. 5. Thermal conductivity ratio γ as a function of the superlattice period length M for three device lengths N.

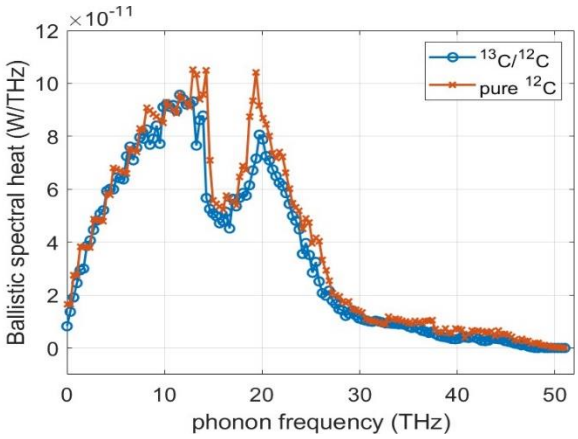

Fig. 6. Spectral heat flow comparison between pure graphene and the isotope superlattice with N=24 and M=8.