Interflake Thermal Conductance of Edge-passivated Graphene

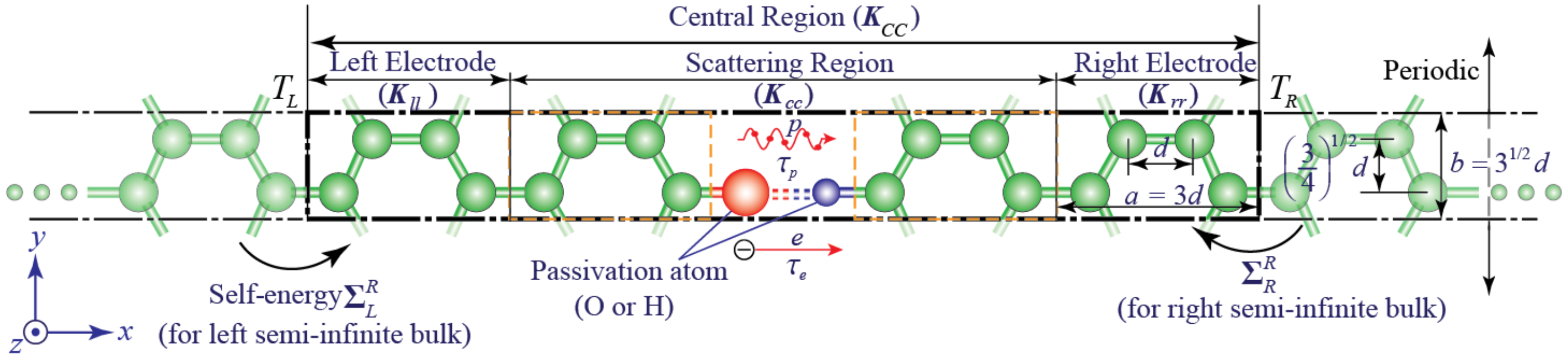
Based on the quantum-junction transmission/Green's function formalism and the dynamical matrix/DFT, we find the phonon wave features result in bimodal resonant transmission in the interflake conductance of H or O edge-passivated graphene. The low-frequency mode is due to the weak interaction between the flakes, while the high-frequency mode depends on the passivated species and brings the temperature dependence. The phonon transport polarized in the transport directions is dominant because of the asymmetric charge distribution of . . .C-O−H-C. . . and this contributes to the conductance. Thermal conductance decreases due to the passivation junctions, and the electronic thermal conductance becomes negligible except for the O−H junction at high temperatures.

Motivation

- Graphene-based composites are promising for superior electrical and thermal transport properties.
- The bottleneck in transport is in the interflake resistance, and edge passivation (e.g., oxidation) is not avoidable in non-vacuum use of graphene.
- Low-dimensional phonon and electron energy transport are of fundamental interest.

Objective

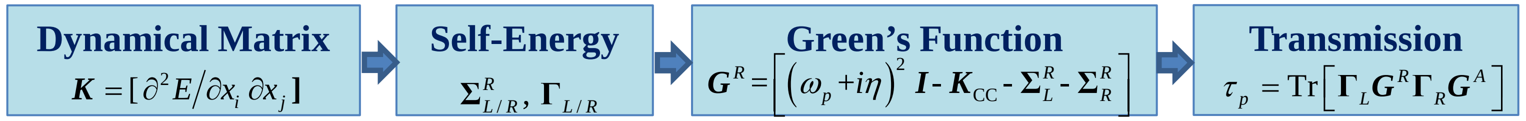
- Formulate and calculate phonon and electron transport across the junction of edge-passivated graphene flakes, using quantum and semi-classical treatments.
- Understand the carrier scattering and transmission across the passivation junctions in the ballistic regime.



The edge-passivated graphene-flake junction used for the thermal transport calculations.

Phonon Transport

1) Green's function formalism with the dynamical matrix from the DFT- Quantum and wave features



Dynamical Matrix $\boldsymbol{K} = [\partial^2 E / \partial x_i \partial x_j]$

Self-Energy $\Sigma^R_{L/R}, \Gamma_{L/R}$

Green's Function $\boldsymbol{G}^R = \left[(\omega_p + i\eta)^2 \boldsymbol{I} - \boldsymbol{K}_{\rm CC} - \Sigma_L^R - \Sigma_R^R \right]$

Transmission $\tau_p = {\rm Tr}\left[\Gamma_L \boldsymbol{G}^R \Gamma_R \boldsymbol{G}^A \right]$

2) Semiclassical treatment – AMM [acoustic mismatch model, $\tau_{p,L/R,\rm AMM} = (4Z_{p,L}Z_{p,R})/(Z_{p,L}+Z_{p,R})^2$]

With the multiple interfaces and impedance (Z_p) proportional to D_p^{-1}, $\tau_{p,\rm AMM} = \prod_{i=1}^{17} \tau_{p,i/i+1}$

3) Phonon Conductance (G_p)

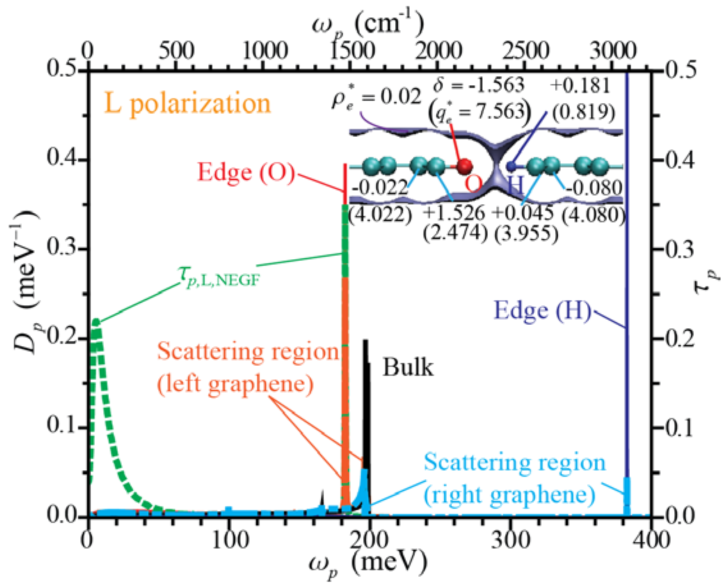
- 1D with the Landauer formula. Transport in the x direction ($k_y = 0$) is dominant.

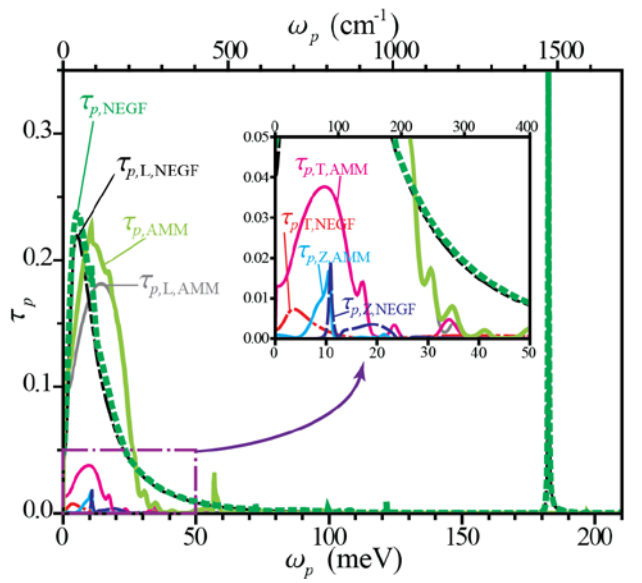
$$G_{p,\rm 1D}(\kappa_y^*)\,[\rm W/K] = \int_0^\infty (\mathrm{d}\omega_p / 2\pi)\ \hbar\omega_p \tau_p(\kappa_y^*, \omega_p) \left[\partial f_p^o(\omega_p, T) / \partial T \right]$$

- k points are sampled in the y direction for 2D, $G_{p,\rm 2D}\,[\rm W/m\text{-}K] = \frac{1}{b}\int_{-\pi}^{\pi} \mathrm{d}\kappa_y^* G_{p,\rm 1D}(\kappa_y^*)$.

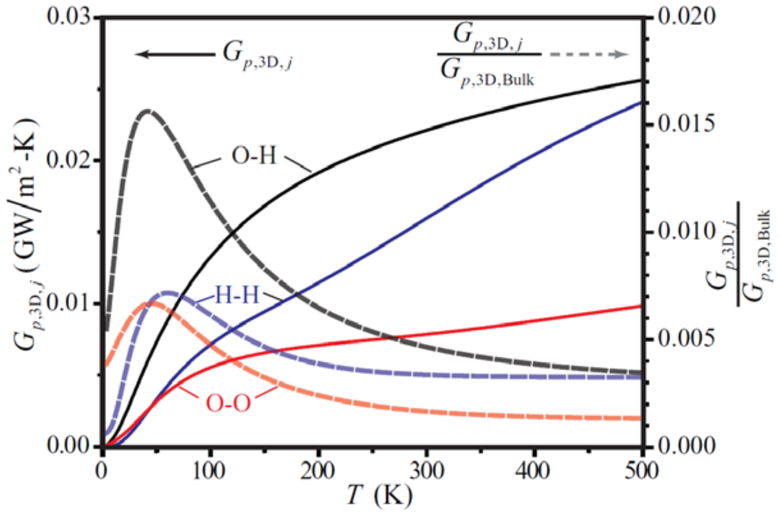
Electron Transport - using the Green's function formalism with the Hamiltonian and overlap matrix

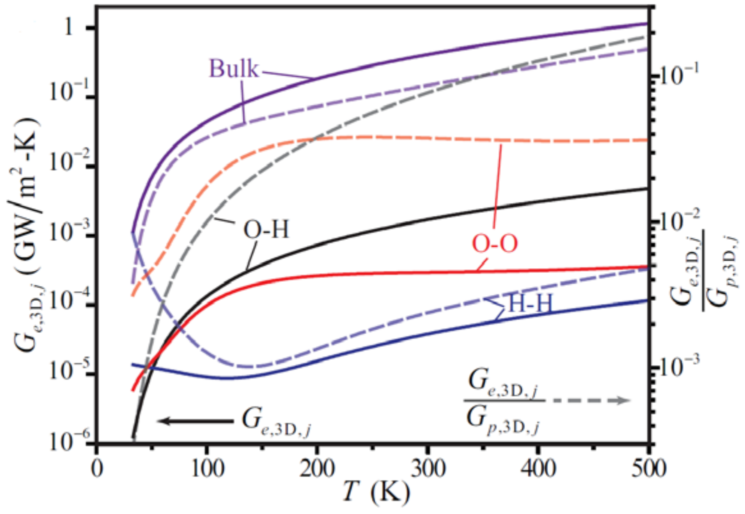
- Electronic Thermal Conductance $G_{e,\rm 2D}\,[\rm W/m\text{-}K] = \frac{1}{bT}\left(K_2 - \frac{K_1^2}{K_0} \right)$, where $K_n = \frac{1}{\pi\hbar}\int dE_e (E_e - E_{\rm F})^n \tilde{\tau}_e(E) \left[-\frac{\partial f_e^o(E_e, T)}{\partial E_e} \right]$

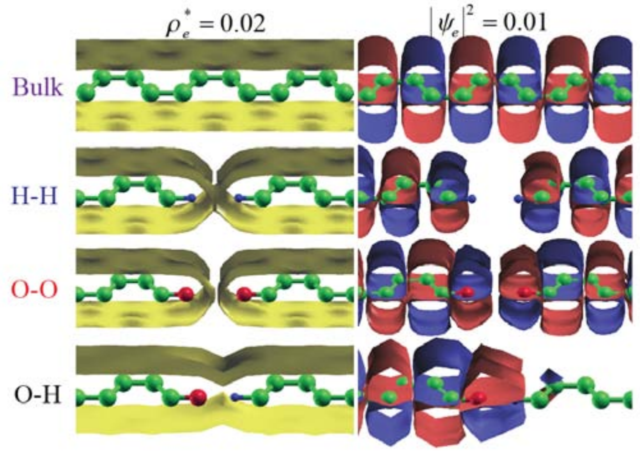




- τ_p is the largest in the L polarization, and is bimodal with the wide low-ω_p peaks and the sharp peaks at the edge-resonant frequencies.
- The AMM agrees well with the NEGF at low ω_p's, but does not show the high ω_p tunneling.







- Phonon conductance decreases due to the passivation junctions while the O-H junction has the largest.
- The electronic thermal conductance is obtained from the NEGF for electrons and found to be negligible except for the O-H junction at high temperatures.
- The low charge density presents between the flakes and the localized orbital exists near the passivated edge for the symmetric junctions.