

Pressure Dependence of Thermal Boundary Conductance at 2D-Substrate Interfaces

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Two-dimensional (2D) materials such as graphene and transition metal dichalcogenides (TMDs) offer transformative potential for scaled nanoelectronics, but severe thermal dissipation bottlenecks arise at van der Waals (vdW) interfaces with dielectric substrates. Gate-induced electrostatic pressure (P_{elec}) provides a promising contact-free mechanism to actively modulate thermal boundary conductance (TBC) across these interfaces without inducing mechanical damage [1]. In this study, we present a comprehensive multiscale numerical framework coupling Density Functional Perturbation Theory (DFPT) calculations with phonon transport modeling to elucidate the physical mechanisms governing pressure-enhanced TBC across 2D-substrate interfaces.

Interfacial vdW interactions between 2D sheets and amorphous SiO_2 substrates were calculated from first principles using Density Functional Theory (DFT) implemented in VASP, then parameterized using a modified Buckingham potential fitted to DFT energy-distance curves, yielding precise vdW spring coupling constants $K_s(h)$. Our model incorporates localized surface roughness statistics (RMS height Δ_{rms} , auto-correlation length L_{cor} , and average surface slope of the roughness $\Delta_{\text{rms}}/L_{\text{cor}}$), energy minimization, and full flexural phonon density of states. On flat substrates, TBC enhancement (dG/dP) is driven exclusively by vdW bond compression, yielding pressure sensitivities of $dG/dP = 0.016 \text{ ms}^{-1}\text{K}^{-1}$ for unencapsulated graphene/ SiO_2 and $0.025 \text{ ms}^{-1}\text{K}^{-1}$ when encapsulated, eliminating internal flexural phonon bottlenecks [1].

When realistic substrate roughness is introduced, partial delamination occurs at zero external pressure, suppressing initial conductance G_0 . Applied electrostatic fields draw suspended regions toward the substrate, expanding the real contact area $\Delta A/\Delta P$ and simultaneously stiffening interfacial vdW bonds $\Delta K_s/\Delta P$ at contact peaks. Consequently, dG/dP exhibits a non-monotonic dependence on surface slope, peaking sharply at $\Delta_{\text{rms}}/L_{\text{cor}} \approx 0.1$, where contact area expansion and bond compression each contribute $\sim 50\%$ to overall TBC enhancement [2]. Beyond this threshold, severe delamination reduces G_0 , suppressing pressure sensitivity. Furthermore, stronger interfacial adhesion increases G_0 and consequently dG/dP [3], whereas stiffer 2D materials (higher bending stiffness D_{bend}) resist conforming to rough surfaces, creating air gaps that lower TBC [4]. Gate-induced carrier densities up to $2 \times 10^{13} \text{ cm}^{-2}$ enhance flexural (ZA) phonon repopulation via electron-phonon coupling ($\Gamma_{\text{el-ph}}$), adding $\sim 4\%$ to total TBC [5]. These findings establish key predictive guidelines for active thermal management in 2D electronics.

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Supplementary Information

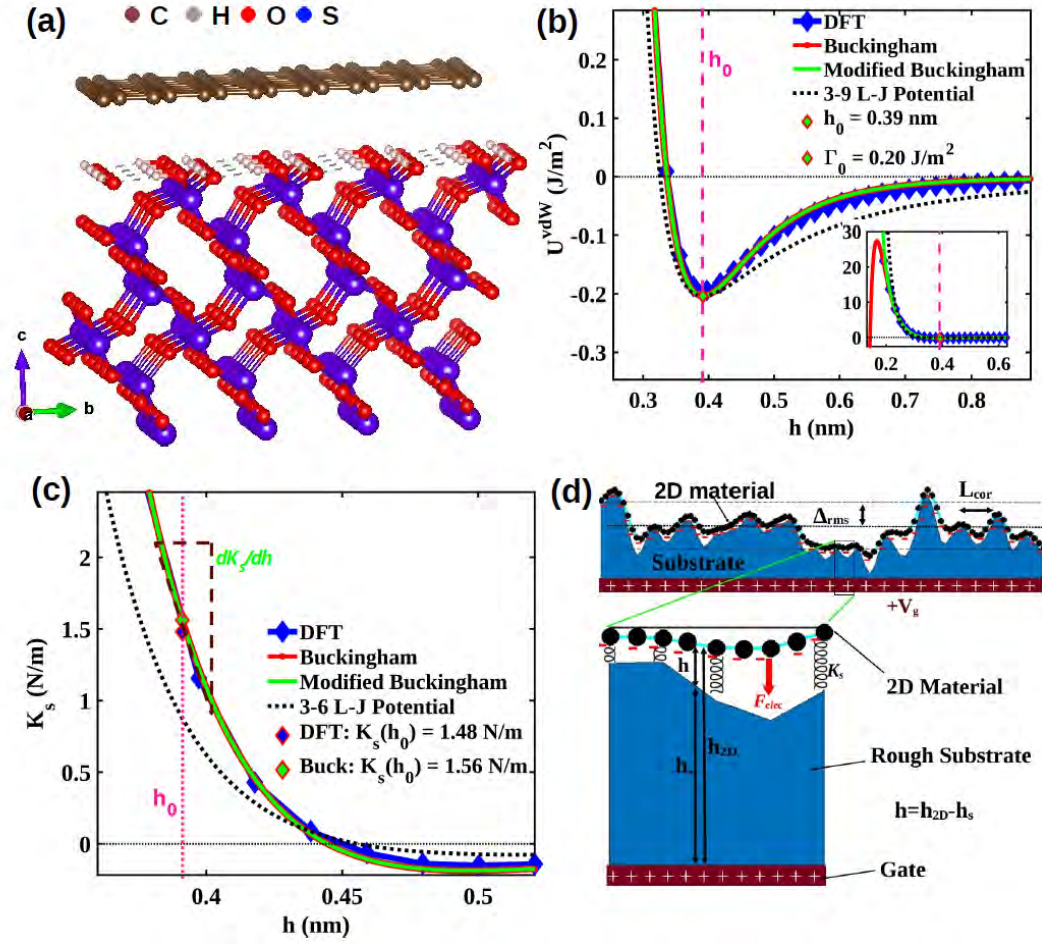


Figure 1: (a) atomic positions in the graphene/SiO₂ supercell for DFT calculations, b) Parameterization of interfacial van der Waals (vdW) adhesion energy $U^{\text{vdW}}(h)$ and (c) spring coupling constant $K_s(h)$ using a modified Buckingham potential fitted to DFPT calculations. (d) schematic of the calculation setup for thermal boundary conductance including atomic-scale surface roughness of the substrate and partial delamination of the 2D sheet.

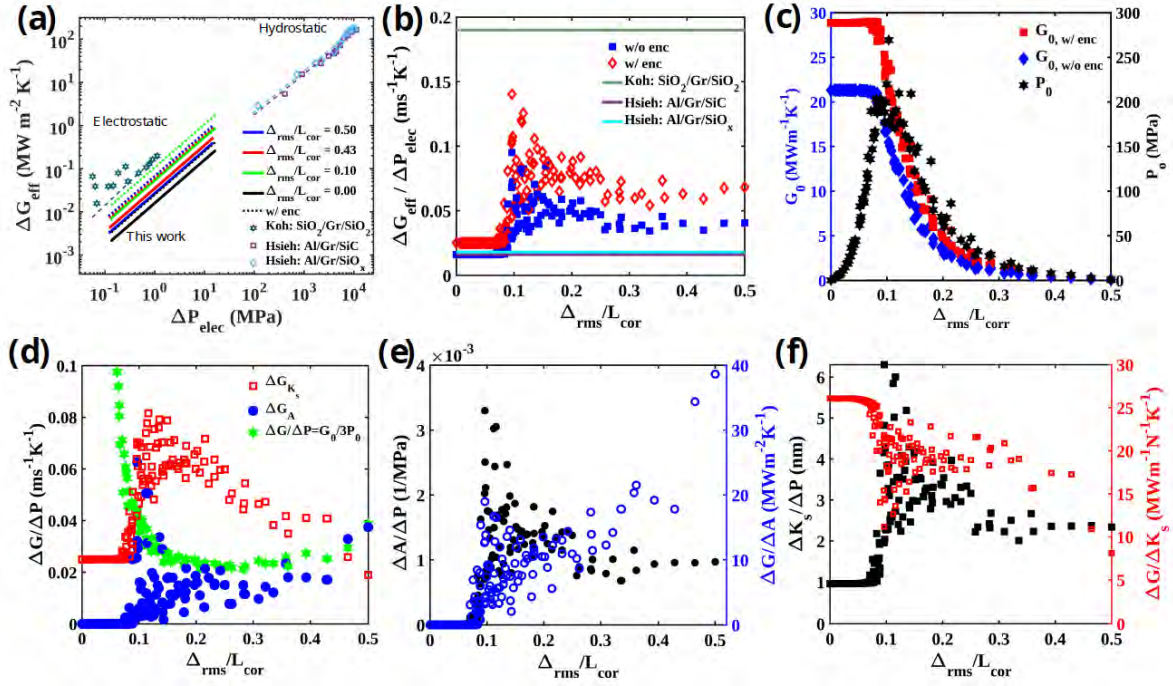


Figure 2: (a) TBC as a function of pressure showing comparison with experimental data from the literature. (b) The ratio of TBC to pressure, $\Delta G/\Delta P$, as a function of surface slope, showing that $\Delta G/\Delta P$ first increases with surface slope, peaks around 0.1 and then decreases. (c) TBC before the application of the electrostatic field, G_0 , and pressure due to the vdW attraction force, P_0 , showing that G_0 decreases with surface slope while P_0 first increases to a peak around a surface slope of 0.1, then decreases. (d) The ratio of TBC enhancement due to improvements in K_s and contact area to the overall TBC, showing that the TBC enhancement at flat interfaces is dominated by K_s , whereas at large surface slope, the increase in contact area becomes more significant.