Silicon-Doped Graphene as a Phonon Bridge: A Route to Threefold Enhancement of Interfacial Thermal Conductance in Graphene–Diamond Heterostructures

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Abstract
The exceptional intrinsic thermal conductivity of graphene makes it an attractive candidate for next-generation thermal management; however, heat dissipation in practical devices is often constrained by interfacial resistance at graphene-substrate junctions [1,2]. Weak coupling and vibrational mismatch across van der Waals interfaces significantly suppress interfacial thermal conductance (ITC), limiting efficient phonon transmission [3]. Here, we investigate the role of heteroatom doping in modulating heat transfer across graphene-diamond (Gr-D) interfaces using a multiscale computational framework combining non-equilibrium molecular dynamics, vibrational density of states analysis, and density functional theory. The baseline ITC of pristine Gr-D at 298 K is found to be ~14 MW m^{-2} K^{-1}, consistent with prior reports [4]. While nitrogen and boron substitutions induce only marginal changes, silicon doping leads to a substantial and concentration-dependent enhancement in ITC, reaching values up to ~57 MW m^{-2} K^{-1}. Spectral analysis indicates that silicon incorporation redistributes out-of-plane phonon modes, increasing overlap with diamond vibrational states across both low- and high-frequency regimes, thereby facilitating more effective phonon transmission. First-principles calculations further reveal strengthened interfacial interactions and localized charge redistribution associated with silicon sites, suggesting partial covalent character that promotes cross-interface vibrational coupling. In contrast, nitrogen and boron primarily perturb the phonon spectrum without significantly altering interfacial bonding. These findings highlight a viable strategy for tailoring interfacial heat transport through targeted dopant selection, providing design guidelines for high-performance graphene-based thermal interfaces in advanced electronic systems.

References

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