

# Tuning the vibrational density of states of van der Waals heterostructures for enhanced thermal boundary conductance

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Heterostructures of two-dimensional (2D) vdW materials have emerged as a platform for novel nano- and opto-electronic applications because they offer numerous stacking combinations that may lead to properties not achievable in individual layers. However, the thermal properties of these heterostructures have received less attention, even though they are essential, as Joule heating can generate hotspots in such devices. In 2D transistor applications, hexagonal boron nitride (hBN) is typically used as an interlayer between the 2D channel and the substrate to enhance electron mobility [1] and contribute to heat spreading. Heat transfer between 2D materials and their substrates is driven by a combination of the overlap of the vibrational density of states (vDOS) of the flexural phonons in the 2D layer and the substrate [2,3,4] (Figure 1) and vdW coupling strength at the interface. When a 2D layer is coupled to a substrate, the interfacial restoring force opens a gap in the vDOS, with the gap frequency determined by the interfacial force constant and the layer's mass density [5], and when multiple layers are added to the structure, the phonon branches become hybridized [6], creating new features in the vDOS. Whether one can control the vDOS of the heterostructure by selecting the composition and the stacking order in the vdWH to enhance heat transfer to the substrate remains an open question.

In this work, we compute phonon dispersions from density functional theory for the constituent monolayers and construct a coupled dynamical matrix for the out-of-plane flexural phonon branches. We also compute the van der Waals spring constants for the interlayer and substrate interactions. We then calculate the layer-projected vDOS to determine how modes originating in the transport layer contribute to motion in the substrate-interface layer and obtain the thermal boundary conductance (TBC) into the substrate. We observe that coupling to the substrate through long-wavelength breathing modes that displace all layers of the vdW heterostructure dictate the TBC.

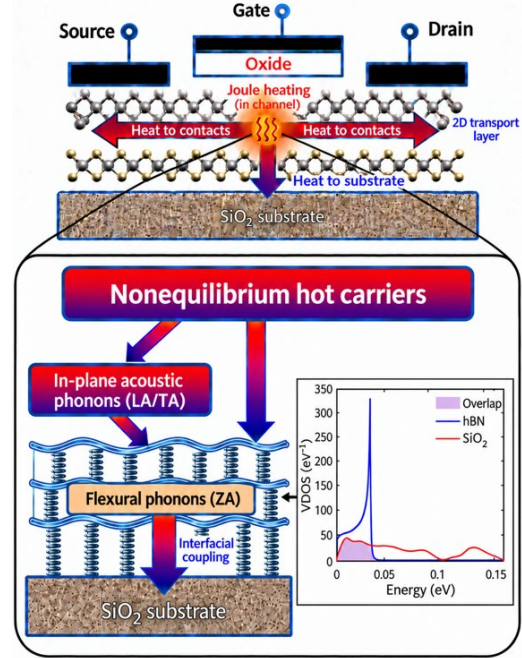


Figure 1: A simplified schematic of thermal transport pathways for heat generated in a transistor. Heat transfer to the substrate is driven by the van der Waals spring constants and overlap between the vibrational density of states.

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## Supplementary Information:

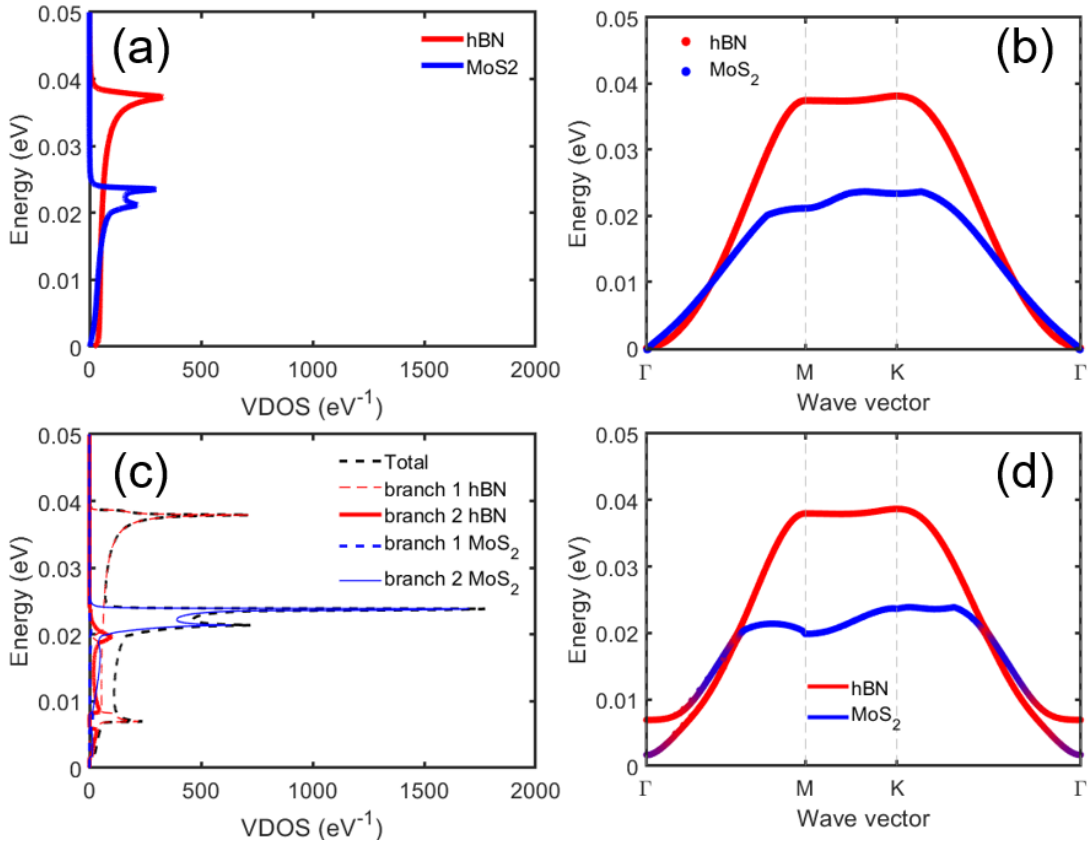


Figure 2: The vibrational density of states (VDOS) for hBN and MoS<sub>2</sub> monolayers are plotted in (a). The corresponding dispersions for the two monolayers are plotted in (b). In (c) the total VDOS of the heterostructure when sandwiched between SiO<sub>2</sub> and AlO<sub>x</sub> is plotted along with projections of the VDOS into the top and bottom layers of the stack. In (d) the corresponding dispersion of the stack is plotted with color weights depicting the layer that the band resides in.

Figure 3 shows a schematic of a van der Waals stack on the left, consisting of layers labeled  $K_0, K_1, K_2, \dots, K_{N-1}, K_N$ . To the right is the dynamical matrix  $\tilde{D}_{mat}$  for the structure, which is a symmetric matrix with the following elements:

$$\tilde{D}_{mat} = \begin{pmatrix} \frac{K_0+K_1}{\rho_1} + \omega_1^2 & -\frac{K_1}{\rho_1} & 0 & \dots & 0 \\ -\frac{K_1}{\rho_2} & \frac{K_1+K_2}{\rho_2} + \omega_2^2 & -\frac{K_2}{\rho_2} & 0 & \dots & 0 \\ 0 & -\frac{K_2}{\rho_3} & \frac{K_2+K_3}{\rho_3} + \omega_3^2 & -\frac{K_3}{\rho_3} & 0 & \dots \\ \vdots & \vdots & \vdots & \ddots & \vdots & \vdots \\ 0 & 0 & 0 & 0 & \dots & -\frac{K_{N-1}}{\rho_N} & \frac{K_{N-1}+K_N}{\rho_N} + \omega_N^2 \end{pmatrix}$$

Figure 3: A schematic of the coupling forces for a van der Waals stack sandwiched between two rigid 3D materials along with the dynamical matrix for the structure which modifies the dispersions of monolayers calculated through density functional theory.