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Zlatan Aksamija, *University of Massachusetts Amherst*

Cameron J Foss, *University of Massachusetts Amherst*

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Quantifying thermal boundary conductance of 2D–3D interfaces

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Cameron J Foss and Zlatan Aksamija

University of Massachusetts Amherst, Amherst, MA 01003, United states of America

E-mail: zlatana@engin.umass.edu

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Abstract

Heat dissipation in next-generation electronics based on two-dimensional (2D) materials is a critical issue in their development and implementation. A potential bottleneck for heat removal in 2D-based devices is the thermal pathway from the 2D layer into its supporting substrate. The choice of substrate, its composition and structure, can strongly impact the thermal boundary conductance (TBC). Here we investigate the temperature-dependent TBC of 42 interfaces formed between a group of six 2D materials and seven crystalline and amorphous substrates. We use first-principles density functional perturbation theory to calculate the full phonon dispersion of the 2D layers and substrates and then input them into our model for phonon transport across the 2D–3D interface. Our results show that the TBC depends on the overlap between the vibrational frequencies and can be varied by nearly two orders of magnitude, from as low as $\sim 0.6 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ (h-BN on diamond) to $\sim 40 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ (h-BN on SiO_2), for the same 2D layer by changing the substrate material. We find that amorphous materials significantly boost the TBC relative to their crystalline counterparts, assuming the two interfaces have the same adhesion, owing to the low-frequency Boson peak feature in their vibrational density of states (vDOS). For crystalline substrates, we further correlate constituent material properties with the calculated TBCs and find that the TBC strongly depends on a combination of the speed of sound, Debye temperature, and density of the substrate as well as the bandwidth of the flexural branch in the 2D material. We conclude that softer substrates with sharp low-frequency features in their vDOS, such as amorphous materials, polymers, and nanoparticles, could have higher TBC, leading to a trade-off between TBC and the thermal conductivity of the substrate.

1. Introduction

With the further decrease of device sizes and dimensionality, the interface resistance between adjacent material domains in common electronic devices surpasses the material-domain resistances, becoming a bottleneck for waste heat removal [1–3]. Further, advanced design complexities increase the number of domains/interfaces in the pathway from heat source to heat management solutions, thus adding additional thermal resistance. Atomically-thin 2D materials have promising potential for next-generation nano- and optoelectronics [4–6]. However, heat dissipation from hot spots in the 2D material to its environment remains a critical concern to the design of 2D-based devices [6–9]. Thermal currents flowing in a 2D material can either dissipate through source/drain contacts, as in a transistor configuration, and/or through a supporting substrate via the vdW

interaction [10]. When a 2D material is supported by a substrate, the interfacial area formed between the 2D material and the substrate is typically larger than the lateral source/drain contact interface area [7, 11]; thus, it is expected that thermal resistance at the interface between the 2D layer and substrate will play a critical role in thermal management of 2D-based devices. The role of the interface is particularly prominent when a high thermal conductivity substrate, such as diamond or aluminum nitride, is used [12]. Interface heat transfer is further hampered by the fact that interactions of atoms on the two sides of the interface are governed by relatively weak van der Waals (vdW) forces, which are known for reduced interface thermal transport relative to covalently bonded interfaces [9, 13, 14].

For these reasons, it is imperative for reliable 2D-device performance that the thermal boundary conductance (TBC) between 2D materials and

various substrates be well characterized and that new solutions for boosting the TBC are proposed. Consequently, numerous measurements and calculations of 2D–3D TBC have been performed; we review these in section 2. Despite these advances, measured and calculated TBC values are scattered over a range spanning two orders of magnitude and there is not yet a roadmap between material structure (crystal and amorphous) and properties and the TBC of 2D–3D interfaces. Nor does a clear picture yet emerge of the ideal substrate, from the thermal point of view, for each 2D material. In this work, we investigate the TBC between six 2D-materials (graphene, h-BN, MoS₂, MoSe₂, WS₂, and WSe₂) and seven substrates (SiO₂, GaN, AlN, SiC, diamond, crystalline Al₂O₃, and amorphous AlO_x), and draw connections between the constituent material properties and their resulting TBCs. We assume all interfaces have the same adhesion energy (van der Waals spring coupling) throughout to highlight the effects of the substrate properties on the TBC. Our results suggest that (i) due to the large contributions of low-frequency excitations, amorphous materials exhibit much higher TBC than crystalline materials; (ii) the TBC can change by nearly two orders of magnitude based on choice of substrate; and (iii) the TBC for crystalline materials shows a strong correlation with the inverse of the product of 2D atomic mass (m_{2D}), substrate sound velocity (v_{sub}), substrate Debye temperature squared (Θ_{sub}^2), substrate mass density (ρ_{sub}), and 2D flexural phonon bandwidth (BW_{ZA}).

2. Background

The TBC between 2D and 3D materials has been studied experimentally through several methods including Raman spectroscopy [8, 9, 15–17], differential 3- ω [18, 19], pump-probe [20–22], time- and frequency-domain thermoreflectance (FDTR/TDTR) [23–25], and, more recently, electrical thermometry platforms [12, 26]. Graphene on silicon dioxide (Gr/SiO₂) has been studied frequently and most measurements (w/ and w/o metallic coating-layers) [12, 21, 23, 24, 26] report TBC in the range 20–27 MW · m^{−2} · K^{−1}, although values as high as 50 and 83.33 MW · m^{−2} · K^{−1} have been demonstrated by pump-probe (bare Gr/SiO₂) [20] and 3- ω (Au/SiO₂/Gr/SiO₂) [18] measurements, respectively. Similarly, Au/Ti-coated graphene on amorphous aluminum oxide (AlO_x) was shown to be within the same range at 21 MW · m^{−2} · K^{−1} by electrical thermometry measurements [12].

Graphene-on-wurtzite structures have also been investigated; three-layer (3L) graphene on silicon carbide (Gr/SiC) was revealed to have a TBC of 1.9×10^{-2} MW · m^{−2} · K^{−1} [27] by Raman thermometry with electrical Joule heating, and the TBC of single-layer graphene on aluminum nitride (Gr/AlN) was measured to be 34 MW · m^{−2} · K^{−1} [12]. An electrical thermometry platform was also used to measure

the TBC of Gr/Sapphire (crystal Al₂O₃) and Gr/diamond interfaces with metal (Au/Ti) encapsulation [12]. There, the authors report TBCs of 34 MW · m^{−2} · K^{−1} for Au/Ti/Gr/Al₂O₃ and 6 MW · m^{−2} · K^{−1} for Au/Ti/Gr/diamond stacks. Thermal transport across a graphene-semiconductor interface without a coating-layer was measured by Raman spectroscopy and found to have a TBC of 28.01 MW · m^{−2} · K^{−1} (Gr/Si) [28], which was followed up by a pump-probe study [22] of an Al/Gr/Si stack where the TBC of the Gr/Si interface was reported to be 62.5 MW · m^{−2} · K^{−1}. The TBC of graphene-metal interfaces were also recently studied by TDTR measurements where the TBC was demonstrated to be in the range 20–30 MW · m^{−2} · K^{−1} for Gr/Cu and Gr/Al interfaces [25].

Investigations into the TBC of 2D/3D interfaces with 2D materials beyond graphene have garnered attention only recently. Metal-coated (Au/Cr) single- and few-layer boron nitride on SiO₂ was measured by a 3- ω method and reported to have a TBC of 62.5 MW · m^{−2} · K^{−1} (for 1 l) which was shown to monotonically decrease to 29.4 MW · m^{−2} · K^{−1} in 12.8-nm-thick h-BN [19]. Molybdenum disulfide (MoS₂) on dielectric, semiconducting, and metallic substrates have also been investigated predominantly by Raman spectroscopy. Initial Raman thermometry measurements for bare MoS₂/SiO₂ interfaces revealed low TBC at 0.44 [29] and 2 MW · m^{−2} · K^{−1} [30]. However, follow-up Raman experiments with electrical [9] and optical [15] heating sources have shown slightly higher TBC values at 15 ± 4 MW · m^{−2} · K^{−1}, yet these values are still lower than previously reported Gr/SiO₂ interfaces. Most recent electrical thermometry measurements [26] of mechanically exfoliated/transferred and direct-CVD grown Au/Ti/MoS₂/SiO₂ have demonstrated even higher TBC at 20.3 and 33.5 MW · m^{−2} · K^{−1}, respectively.

Single-layer MoS₂/AlN interfaces were probed by Raman thermometry with optical heating [15] where the TBC was detailed to be similar to the identical device with SiO₂ as the substrate (15 ± 4 MW · m^{−2} · K^{−1}). Bare seven-layer MoS₂ on silicon (7L-MoS₂/Si) was studied by a photon-heating Raman-probe method [17] where the TBC was reported to be 0.974 MW · m^{−2} · K^{−1}. In the same work, the TBC was shown to increase from 0.974 to 68.8 MW · m^{−2} · K^{−1} with increasing MoS₂ film thickness from 7 to 75 layers; we note that this trend contradicts the aforementioned [19] trend seen for Au/Cr/BN/SiO₂. The TBC between MoS₂ and gold was probed by Raman thermometry [29] and found to be very low at 0.09 MW · m^{−2} · K^{−1}. A new method that uses an electron beam as a heating source and metal contacts as temperature sensors was implemented to study the thermal interface between MoS₂ and platinum (Pt) [31] where the authors report a value of 0.416 MW · m^{−2} · K^{−1} for the MoS₂/Pt interface. Experimental investigations into the thermal boundary conductance of interfaces involving other 2D materials have received far less

attention. An electronic-heating Raman-probe platform was used to measure the TBC of few-layer WSe_2 on SiO_2 with an AlO_x coating layer [16] where the TBC was revealed to be $\sim 16 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$.

While experimental measurements provide lumped-value TBCs that cannot be readily separated into their comprising parts. For instance, it was demonstrated that measurements with coating layers may not necessarily provide quantitative information on bare 2D–3D interfaces due to changes in the vibrational spectra of the 2D layer with encapsulation [26]. Furthermore, trends seen for thickness dependence of TBC [17–19] have yet to be corroborated across experimental techniques/studies. To address these questions, there have been several efforts to theoretically model thermal transport across a 2D–3D interface by molecular dynamics (MD) [13, 32–36] and various first-principles-based semi-classical (FP-SC) models [11, 16, 26, 37–40]. A theoretical model for heat transfer between weakly coupled systems was developed in [37] where the TBC of a Gr/ SiO_2 was predicted to be $25 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$. This model was subsequently modified [11] to account for dampening of flexural (out-of-plane) acoustic phonons, the primary heat carriers across a 2D–3D interface [36]. The damping, caused by the coupling and exchange of energy with other intrinsic degrees of freedom (including electrons and in-plane acoustic phonons), resulted in an increase of the TBC to $34.6 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ for bare Gr/ SiO_2 and $105 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ for an oxide/Gr/ SiO_2 stack, in good agreement with previous $3\text{-}\omega$ measurements [18].

Gr/SiC interfaces have also been studied by molecular dynamics and FP-SC modeling where reported TBC values have been shown to be higher than measured values. MD simulations for a Gr/4H-SiC interface [13] revealed the TBC increases from $17.8 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ for a Si-terminated 4H-SiC surface to $23.3 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ for the C-terminated counterpart. The authors attribute the difference to the stronger bonding of C–C bonds over C–Si bonds. Further, in the same work it is shown that making the SiC substrate amorphous increases the TBC beyond the values seen for crystal SiC to $34.9 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$. In another work the TBC of a 6H-SiC/Gr/6H-SiC stack was reported to be nearly three-times higher than the bare (Gr/SiC) interfaces at $90 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ by MD [33]. MD simulations of a Gr/Si interface [35] reported a TBC of $28.4 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ which agrees well with TBC values reported by Raman spectroscopy [28]. Interfaces formed between graphene and polymers were also studied by MD calculations where a rather high TBC of $60 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ was demonstrated [34].

Similar to aforementioned measurements, only recently have 2D–3D interfaces with 2D materials beyond graphene been a focus of theoretical investigation. The TBC of bare and oxide-coated $\text{MoS}_2/\text{SiO}_2$ interfaces were reported in [11] to be 3.1 and $5.07 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$, respectively. These low TBC values

are in fair agreement with early Raman measurements [29, 30]. Later, MD and FP-SC modeling demonstrated higher TBC values at 15.6 (MD) and $33 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ (FP-SC) for a $\text{Ti}/\text{MoS}_2/\text{SiO}_2$ interface [26]. A multilayer TBC model was recently developed [40] and used to study the thickness dependence of a Gr/ SiO_2 interface. The TBC was predicted to increase from 34.48 to $87.71 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ from 1 to 14 layers then saturate to the TBC value of graphite/ SiO_2 . A first-principles-based multilayer TBC model was recently used to study AlO_x encapsulated few-layer WSe_2 on SiO_2 [16], where the TBC displayed a soft peak as a function of 2D-layer thickness, contrary to the monotonic trend seen in $\text{Au}/\text{Cr}/\text{BN}/\text{SiO}_2$ by $3\text{-}\omega$ measurements [19].

3. Theory

Typical parameters that determine thermal boundary conductance at same-dimensional, covalently-bonded interfaces (3D/3D or 2D/2D) are acoustic impedance, vibrational density of states (vDOS), and atomic morphology at the interface [3, 41–44]. When a vdW gap is present, the interactions between the atoms on either side of the interface are governed by weak vdW forces. As a consequence, the TBC drops significantly relative to covalently bonded interfaces [45]. Although vdW forces are weak, they collectively lead to an adhesion energy between 2D and 3D materials, which has been shown to impact the TBC [39]. In addition, there is significant mismatch between the vibrational modes and between the 2D and 3D phase spaces. While the adhesion at 2D/3D interfaces can be somewhat improved through annealing [26] and by choosing a substrate with improved surface features, such as lower surface roughness, the range of adhesion values is likely to have an upper bound, that of the interlayer adhesion in the layered bulk material (for graphene that would be the interlayer adhesion of graphite [46, 47], around $0.2 \text{ J} \cdot \text{m}^{-2}$). On the other hand, the vibrational mismatch can be dramatically reduced by choosing a substrate that is more commensurate with the 2D material, as in the case of graphene on h-BN, or one with vibrational features that more closely resemble those of the 2D material.

In this work, we consider the substrate effects on thermal properties of the 2D-layer and aim to draw conclusions between the constituent materials and their resulting TBC. Namely, the atoms in the 2D layer and surface atoms of the substrate interact via weak van der Waals forces, which can be treated as a spring with coupling constant K_s , analogous to a classical harmonic spring model [37]. The atoms vibrate around their equilibrium positions and thus transfer vibrational energy through the vdW ‘springs’. In addition, due to the coupling between the 2D layer and substrate, out-of-plane flexural acoustic phonons in the 2D material experience an offset ω_0 which is representative of the long-wavelength interactions between the

Figure 1. The full phonon dispersion of graphene (a), h-BN (b), MoS₂ (c), MoSe₂ (d), WS₂ (e), and WSe₂ (f), calculated from first-principles using density functional perturbation theory. The gapped ZA (g-ZA) branch of each 2D layer is highlighted in red and is the primary contributor of thermal transport across the interface [36].

2D-layer and substrate [48, 49]. Note that out-of-plane flexural acoustic phonons have atomic displacements in the z -direction, so they are commonly abbreviated as ZA—analogue to the abbreviations of longitudinal acoustic (LA) and transverse acoustic (TA) phonons. The offset ZA branch has been referred to as the flexural-gapped branch [48]; in this work we will refer to it as the gapped ZA branch (g-ZA).

In our approach, we employ first-principles DFPT simulations as implemented in the Quantum Espresso package [50] to obtain the full phonon spectrum of six 2D materials, shown in figure 1, and four substrates (namely wurtzite GaN, AlN, SiC, and fcc diamond). The DFPT simulation details are described in the appendix. After we obtain the phonon dispersion of each 2D material, we modify it to account for long-wavelength interactions with the substrate which cause gapping in the flexural mode. To do so, we modify the ZA branch frequencies $\omega_{ZA}(\vec{q})$ in the same manner as our previous works [26, 39] as $\tilde{\omega}_{ZA}(\vec{q}) = \sqrt{\omega_{ZA}^2(\vec{q}) + \omega_0^2}$, which ensures that only long-wavelength ZA phonons are gapped. The dispersions of gapped ZA phonons are depicted by red lines in figure 1. We obtain values of ω_0 precisely as the angular frequency of a harmonic oscillator as $\omega_0 = \sqrt{K_a/m_{2D}}$, where K_a is the spring coupling constant between the 2D-layer and substrate and m_{2D} is the mass of the atoms of the 2D-layer in contact with the substrate.

Once the gapping is accounted for, we then calculate the vDOS of each 2D-material as

$$D_{2D}(b, \omega) = \frac{\Omega}{4\pi^2} \int \delta[\omega - \omega_b(\vec{q})] d\vec{q} \quad (1)$$

$$= \frac{\Omega}{4\pi^2} \oint_C L(\omega) dL / |\nabla_{\vec{q}} \omega_b(\vec{q})|. \quad (2)$$

In the above, Ω is the Brillouin zone area which is discretized by a dense set (25,208) of equidistant q -points. The delta function integral is equal to an integral about the constant energy contour C of the quotient between the contour length $L(\omega)$ and the magnitude of phonon group velocity $|\vec{v}_b(\vec{q})| = |\nabla_{\vec{q}} \omega_b(\vec{q})|$. The full vDOS is then obtained by summing over each branch contribution, $D_{2D}(\omega) = \sum_b D_{2D}(b, \omega)$. This is analogous to the Brillouin zone integration method of Gilat and Raubenheimer [51], which we use in calculating the vDOSs of our four 3D crystal substrates. The vDOS of amorphous SiO₂ (a-SiO₂), crystalline Al₂O₃ and amorphous AlO_x are extracted from previously published MD [52], DFPT [53] and first-principles MD [54] works, respectively. Once we have the vDOS of each 2D layer and substrate, we then use them as input to our 2D–3D TBC model.

3.1. 2D–3D thermal boundary conductance

In our model, phonons traveling in the 2D layer transfer thermal energy to the substrate through the vdW bond at a rate given by the substrate scattering rate $\Gamma_S(\omega)$. In this approach, flexural (ZA) phonons of frequency $\omega(\vec{q})$ traveling (in-plane) in the 2D-layer are perturbed by substrate interactions, and in this perturbation the phonon transfers all of its energy $E = \hbar\omega(\vec{q})$ to the substrate. We note that this transfer process is harmonic and conserves energy but it does not conserve momentum due to the mismatch between the phase spaces and the presence of atomic-scale

roughness at the substrate surface which randomizes the component of the phonon momentum parallel to the substrate. The phonon-substrate scattering rate was formulated by Seol *et al* [55] based on Fermi's golden rule as

$$\Gamma_S(\omega) = \frac{\pi}{2} \frac{D_S(\omega)}{m_S m_{2D}} \frac{K_a^2}{\omega^2}, \quad (3)$$

where $D_S(\omega)$ is the substrate vDOS, m_{2D} and m_S are the atomic masses in contact at the interface of the 2D-material and substrate, K_a is the van der Waals (vdW) coupling spring constant, and ω is the phonon vibrational frequency. The vdW spring coupling constant is proportional to the adhesion energy [39] between the 2D layer and substrate, and is generally difficult to quantify experimentally [45]. An upper bound for K_a can be roughly determined as the vdW interlayer coupling of the bulk counterpart of the 2D material which can be as high as roughly 8 N m^{-1} in MoS_2 and WSe_2 [56]. Ong *et al* used DFT to calculate the vdW coupling constant for Gr/SiO₂ and MoS₂/SiO₂ interfaces with H and OH substrate surface termination atoms [11]. There, values of K_a for H (or, OH) termination were reported to be 8.18 (6.45) N m^{-1} and 2.36 (4.27) N m^{-1} for Gr/SiO₂ and MoS₂/SiO₂, respectively.

After calculating the substrate scattering rate, we calculate the TBC from a Landauer formalism as the integral of the product of the modal heat capacity $C(\omega, T)$, 2D vDOS $D_{2D}(\omega)$, and the substrate scattering rate over the phonon spectrum. Assuming the two materials have distinct temperatures with a difference between the 2D layer and the substrate of ΔT , the TBC $G(T)$ is given by

$$G(T) = \int C(\omega, T) D_{2D}(\omega) \Gamma_S(\omega) d\omega. \quad (4)$$

The modal heat capacity is given by

$$C(\omega, T) = \hbar\omega \frac{dN^0(\omega, T)}{dT} \quad (5)$$

$$= k_B \left[\frac{\hbar\omega}{k_B T} \right]^2 \frac{e^{\hbar\omega/k_B T}}{[e^{\hbar\omega/k_B T} - 1]^2}, \quad (6)$$

where $N^0(\omega, T)$ is the equilibrium Bose–Einstein distribution function for phonons, $N^0(\omega, T) = 1/[e^{\hbar\omega/k_B T} - 1]$. A detailed derivation of this model is described in our earlier work [39]. Our model can be related to the Green's function model of Persson *et al* [57] in the weakly-coupled and incoherent limits, with quantitative differences arising from our use of first-principles phonon dispersions.

The TBC we calculate is the inverse of the Kapitza resistance, determined solely by the external resistance between the 2D material and substrate. It assumes the phonons in each material are in local thermal equilibrium at their respective temperature and does not include any additional internal resistance that could arise between electrons and phonons in the 2D mat-

erial [11], or among the phonon branches themselves [58]. We also note that, while we restrict our focus to single-interfaces formed between a 2D material and substrates, this model can be extended to study double-interfaces of 3D–2D–3D stacked systems [26], supported 2D multilayers [16], and the effects of encapsulation [58]. Furthermore, while we only study a modest subset of 2D materials and substrates, this model is general in that it can be used to obtain the TBC between any 2D–3D interface so long as the vDOSs of the constituent materials are known.

4. Results and discussion

In figure 2 we show the temperature dependence of the 42 pairs of 2D-substrate interfaces. Our results show that the TBC can vary by nearly two orders of magnitude from as low as $\approx 0.6 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ (h-BN on diamond) to $\approx 40 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ (hBN on SiO₂) for the same 2D layer by changing only the substrate material. We keep the same coupling constant of $K_a = 2.7 \text{ N m}^{-1}$ throughout to highlight the influence of constituent material properties such as vDOS and atomic mass on the TBC. We arrived at 2.7 N m^{-1} based on our previous work [26] where we used our model to recover experimental measurements of TBC for Ti/Au/Gr/SiO₂ and Au/Ti/MoS₂/SiO₂ by using K_a as a fitting parameter. In that work we extracted values of K_a that were lower than DFT-reported values for Gr/SiO₂ but roughly agreeing with MoS₂/SiO₂ interfaces [11]. We also note that although amorphous substrates can have rougher surface features than crystalline ones, rougher substrate surfaces may not necessarily lead to lower adhesion or vdW coupling. It has been shown that some surface roughness can actually improve interface adhesion [59] and for highly-flexible 2D materials, such as graphene, the 2D-layer can conform to the rough features of the substrate [55, 59]. For these reasons and for the lack of data on vdW coupling constant for interfaces with a wide range of substrates we find that using the same K_a to highlight the effects of material properties is justified.

The temperature dependence of the TBC follows roughly that of the heat capacity of the 2D material, increasing as T^3 at low temperatures and then saturating before reaching the Debye temperature Θ_{2D} of the 2D-material's ZA branch where its heat capacity becomes constant. However, graphene on diamond and Al₂O₃ shows a stronger T-dependence due to their high Debye temperatures. When compared to the crystalline substrates, atomic layers atop amorphous SiO₂ and AlO_x have the highest TBC for all 2D materials, analogous to English *et al* [60], who observed higher TBC at a disordered 3D–3D interfaces. This observation was extended to graphene interfaces with MD simulations [13] by comparing the TBCs of graphene on crystalline and amorphous silicon carbides. We demonstrate it here for a range of 2D layers as well as

Figure 2. Temperature dependence of the thermal boundary conductance (TBC) between our six 2D materials and substrates, exhibiting a roughly cubic trend at low temperatures, followed by saturation near room temperature, approximately following the heat capacity. The vdW coupling strength $K_a = 2.7 \text{ N m}^{-1}$ is kept uniformly across all combinations to facilitate comparisons. Amorphous SiO_2 displays the highest TBC amongst all 2D-layers while diamond and crystalline Al_2O_3 display the lowest.

for other amorphous media. A general trend between the choice in substrate and the TBC arises where the TBC decrease monotonically from SiO_2 - AlO_x - GaN - AlN - SiC - Al_2O_3 -diamond, with amorphous AlO_x significantly outperforming its crystalline counterpart. We note that the values we calculate should be considered representative of the best quality samples and that some experiments may report smaller values due to interface contaminants or variations in sample processing.

Figures 3(a)–(e) show the vDOS of each substrate (black lines) along with the vDOS of the out-of-plane flexural branch of graphene (solid red) and MoS_2 (dotted red). We note here that the large low-frequency peak in the vDOS of amorphous SiO_2 (figure 3(a)) and AlO_x (figure 3(e)) leads to a large substrate scattering rate $\Gamma_s(\omega) \propto D_s(\omega)/\omega^2$; consequently, phonon modes with energies near the ZA gap ω_0 contribute most to TBC [36]. The randomized atomic positions characteristic of amorphous materials destroy long-range order, and with it the higher frequency modes that comprise the van Hove singularities in crystalline materials are redistributed to lower frequencies [61, 62]. This redistribution causes the vDOS of amorphous materials to deviate significantly from the ω^2 dependence, typical of crystalline solids at low frequencies; in contrast, the reduced vDOS $D_s(\omega)/\omega^2$ of amorphous materials exhibits a low-frequency peak that is often termed the *Boson peak* [63, 64]. We identify the presence and location of the Boson peak as the cause of higher TBC. The peak for SiO_2 occurs around 10 meV whereas the peak for Al_2O_3 occurs around 20 meV, and as a result we see slightly larger TBC for most interfaces with SiO_2 as the substrate.

For crystalline substrates, we can again look at the vDOS at low-energies ($<60 \text{ meV}$), as shown in

figure 3, where the vDOS follows $D_{\text{Debye}} = \omega^2/v_{\text{sub}}^3$ until the acoustic branches deviate from their linearity and flatten near the Brillouin zone edge, forming van Hove singularities in the vDOS. We can see the inverse relationship of TBC on the substrate sound velocity as GaN, which has low acoustic sound velocity ($\sim 3200 \text{ m s}^{-1}$), has the highest TBC among the crystalline materials. When we replace GaN with AlN and SiC the substrate sound velocity increases ($\sim 4800 \text{ m s}^{-1}$ and $\sim 5400 \text{ m s}^{-1}$, respectively) and we see a decrease in the low-energy vDOS leading to a decrease in the TBC. The worst substrate for TBC is diamond, whose high sound velocity ($\sim 9500 \text{ m s}^{-1}$) leads to minimal low-energy vDOS and a very low TBC.

To further elucidate the effects of the Boson peak location, we model a generalized amorphous substrate vDOS that follows a Lorentzian squared function for the Boson peak curve [65]

$$D_{BP}(\omega) = \frac{\omega^3}{(\omega^2 - v_0^2)^2}, \quad (7)$$

where v_0 roughly determines where the Boson peak occurs in the energy spectrum. We denote the precise location of the Boson peak for this Lorentzian squared function as $\omega_{BP} = \sqrt{3}v_0$. For comparison, we plot the vDOS of amorphous SiO_2 and AlO_x alongside this generalized vDOS (D_{BP}) for several values of ω_{BP} in figure 4(a). In figure 4(b) we show the TBC between several 2D-layers and the generalized Boson peak vDOS as a function of the Boson peak location ω_{BP} . We see a distinct peak around 4–6 meV which then drops rapidly thereafter. This indicates that substrates whose Boson peak occurs lower in the energy spectrum (roughly between 4–6 meV) will boost the TBC. Further, we see that softer 2D materials, such as the transition metal dichalcogenides (TMDs),

Figure 3. The vibrational density of states (vDOS) of SiO₂ (a), GaN (b), AlN (c), SiC (d), crystalline Al₂O₃ (e), amorphous Al₂O₃ (f), and diamond (f) compared to the vDOS of their respective Debye model approximations (where applicable) as well as the vDOS of the ZA branch of graphene and MoS₂.

Figure 4. The vibrational density of states of amorphous SiO₂ (solid black) and AlO_x (dotted black) shown in (a) alongside a Boson peak model at different values of ν_0 . The thermal boundary conductance between our six 2D-layers and a generalized amorphous substrate with a Boson peak vDOS is plotted as a function of the Boson peak location in (b).

respond better to this trend as their TBC peaks at a higher value than graphene. This highlights that the role of overlap between the vDOS of the 2D-layer and substrate is dominant over the increase of mass of the 2D-material atoms. Most interestingly, hBN atop this generalized substrate shows a peak around $600 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ which far exceeds previously reported TBC values for 2D-oxide and 2D-semiconductor interfaces (both experimentally and theoretically) and is within the range of TBC for graphene-metal [66, 67], 3D-3D vdW [14] and other 3D-3D covalently-bonded interfaces [68, 69].

The superior TBC of hBN on amorphous substrates over other 2D materials can be explained by

its lower atomic mass, which maximizes the substrate scattering rate, in combination with its softer ZA phonon dispersion as compared to graphene (see figures 1(a) and (b)). In hBN, the difference in masses of B and N atoms causes a gap between ZA and ZO phonons at the K point and a flattening of the phonon dispersion of ZA phonons along the M-K path [70]. The flatter dispersion leads to a larger vDOS at low-energies (0–30 meV) that combines with the larger substrate scattering rate, caused by the light monolayer mass and large substrate vDOS of amorphous materials, and maximizes the integrand of equation (4), ultimately boosting the TBC. We suspect that hBAs, having even larger mass difference and a flatter ZA

Figure 5. Scatter plots of the TBC of our six 2D layers and five crystalline substrates as functions of their material properties: (a) ratio of Debye temperatures Θ_{2D}/Θ_{sub} , (b) $1/(m_{2D}m_{sub}\Theta_{sub}^3)$, (c) $1/(m_{2D}m_{sub}v_{sub}^3)$, (d) $1/(m_{2D}v_{sub}^3\rho_{sub})$, (e) $1/(m_{2D}v_{sub}^3\rho_{sub}BW_{ZA})$, and (f) $1/(m_{2D}v_{sub}\Theta_{sub}^2\rho_{sub}BW_{ZA})$. The choice of parameters is determined from a Debye model for the substrate vDOS, as detailed in section A.2.

branch [71], would exhibit even higher TBC on amorphous substrates.

Next, by starting from a Debye approximation for the substrate vDOS, we derive analytical expressions to the TBC that help map material properties to the TBC (see appendix). With these analytical expressions, we map the properties of the materials that constitute the interface to their calculated TBC. We use our 6×5 dataset (the vDOS of amorphous SiO_2 and AlO_x does not follow the ω^2 Debye model, so they are excluded here) to map TBC values to material properties in figures 5(a)–(e). We look at the ratio between Debye temperatures in figure 5(a) which is often used for bulk 3D–3D interfaces [42, 72], and we see a slight trend when looking at certain 2D layers (graphene and h-BN) but overall the trend across different substrates is weak. In figures 5(b) and (c), we plot the values of TBC against two formulations of the denominator of equation (A.5), where we interchange the substrate sound velocity and the substrate Debye temperature as they are related through equation (A.7) and $\hbar\omega_D = k_B\Theta_{sub}$. While we do not see much of a trend in figures 5(b) and (c) starts to show grouping in the top-right corner of the higher TBC interfaces. However, the lower value TBC interfaces are far from the bottom-left corner.

We then recognize that the substrate atomic mass is related to the substrate mass density in figure 5(d) where we see the TBC values for diamond interfaces shift to the left. While we see a shift in the low TBC value interfaces, there is little change for the interfaces involving GaN, AlN, and SiC. In the next panel (figure 5(e)) we go a step beyond the analytical expressions and insert the phonon bandwidth of the flexural

branch (BW_{ZA}) of the 2D-layer into the denominator. The BW_{ZA} is the frequency range of flexural modes, so that a smaller bandwidth implies a flatter dispersion and denser modes at low frequencies where more heat transfer can occur. As a result, we see a tighter grouping of both the low and high TBC values into the bottom-left and top-right corners, respectively. While we see good correlation in figure 5(e), the TBC values for Al_2O_3 interfaces are outliers. To further regress our TBC values, we split the cube of the substrate sound velocity into one part substrate sound velocity and two parts substrate Debye temperature. It is here where we find the most correlation between the material properties considered and the calculated TBC values with the fewest outliers. We find that the inverse of the product of 2D atomic mass, substrate sound velocity, substrate Debye temperature squared, substrate mass density, and 2D flexural phonon bandwidth—that is, $1/(m_{2D}v_{sub}\Theta_{sub}^2\rho_{sub}BW_{ZA})$ —is strongly correlated to the TBC of 2D–3D interfaces with crystalline substrates. Therefore, substrates that are softer and have low sound velocity, low Debye temperature, and low mass density are best for TBC. On the other hand, a 2D layer with lighter atomic mass and a small phonon bandwidth also results in higher TBC.

At the same time, the choice of substrate has influence on the 2D-material's electronic properties, as recently demonstrated by interchanging the rough amorphous surface of SiO_2 with smoother and lattice-matched hBN as a substrate material for graphene-based devices, which improved the conductance of electrons in graphene [73]. Our results show that hBN consistently has the highest TBC of all the 2D materials on each substrate, so it may be a useful ther-

mal intermediary for graphene, with which it forms a good thermal interface [44]. Soft and amorphous substrates boost TBC even further, but are known to have low thermal conductivity [74, 75]. Even though heat from the 2D layer will transfer readily across the interface into an amorphous material, the low thermal conductivity medium might impede the flow of thermal energy away from the interface once transferred across it. This task is better suited to a crystalline material with higher thermal conductivity. Therefore, we speculate that the use of a thin (few nm) layer of amorphous material atop a high thermal conductivity substrate at the interface with a 2D material might improve the TBC enough to aid in overall heat removal in 2D-based devices. For crystalline substrates that have native oxides this approach could be fairly simple, but for materials without a native oxide alternative avenues must be employed. Thin polymer films also exhibit large vDOS at low-frequencies [76] and may be used to boost TBC, as long as the interfacial resistance between the polymer film and the crystalline substrate is small. Polymer substrates have also been shown to boost electronic mobility [77] and reduce hysteresis [78] in MoS₂ devices.

5. Conclusion

We have investigated the thermal boundary conductance between six common 2D-materials and seven substrates. Our results show that the TBC can vary by nearly two orders of magnitude, from $\approx 0.6 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ to $\approx 40 \text{ MW} \cdot \text{m}^{-2} \cdot \text{K}^{-1}$ by changing the substrate material. We find that amorphous materials boost the TBC of 2D–3D interfaces due to their large low-frequency disorder-induced Boson peak in the vDOS, and that the location of the Boson peak can impact the TBC. We applied a Debye model for the vDOS of the crystalline substrates to help map the material properties comprising the 2D–3D interfaces with their respective TBCs. We show that the inverse of the product of 2D atomic mass, substrate sound velocity, substrate Debye temperature squared, substrate mass density, and 2D flexural phonon bandwidth—that is, $1/(m_{2D} v_{sub} \Theta_{2D}^2 \rho_{sub} B W_{ZA})$ —is strongly correlated to the TBC of 2D–3D interfaces with crystalline substrates. Therefore, choosing substrates with simultaneously low density and speed of sound will improve TBC. However, substrate properties that we find to improve the 2D–3D TBC are also known to reduce its bulk thermal conductivity away from the interface, pointing to a trade-off between the bulk and the interface which will require careful simultaneous co-optimization in the future.

Appendix

A.1. First-principles DFPT simulations

The phonon dispersions used here for graphene, MoS₂ and WSe₂ were performed in our previous

works [16, 26]. All calculations are performed with the first-principles software Quantum Espresso [50]. For single-layer hexagonal boron nitride (h-BN) we used a Kresse–Joubert projected augmented wave (KJ-PAW) potential for ion-electron interactions. The electron wave function basis set is expanded up to a cutoff energy of 40 Ry on a shifted $25 \times 25 \times 1$ Monkhorst–Pack (MP) grid, and a Perdew–Burke–Ernzerhof (PBE) type of the generalized gradient approximation (GGA) [79] is used for the exchange-correlation energy. Dynamical matrices are produced for a reduced $8 \times 8 \times 1$ MP grid. The convergence thresholds for the self-consistent total energy and phonon calculations are 10^{-14} Ry and 10^{-12} Ry, respectively. For MoSe₂ we used a Martins–Troullier potential for ion-electron interactions and a PBE-GGA functional for the energy exchange correlation. The electron wave function basis set is expanded up to a cutoff energy of 140 Ry on a shifted $6 \times 6 \times 4$ MP grid. Dynamical matrices are also produced for a $6 \times 6 \times 4$ MP grid. The convergence thresholds for the self-consistent total energy and phonon calculations are 10^{-14} Ry and 10^{-12} Ry, respectively. For WS₂ we used a Vanderbilt potential for ion-electron interactions with a PBE-GGA functional. Electron wave functions are expanded up to 100 Ry on a shifted $27 \times 27 \times 1$ MP grid with a convergence threshold of 10^{-14} Ry. Dynamical matrices are generated for a reduced $8 \times 8 \times 1$ MP grid with a convergence threshold of 10^{-14} Ry. The lattice constants for BN, MoSe₂ and WS₂ are $a = 3.19 \text{ \AA}$, $a = 3.288 \text{ \AA}$ ($h = 3.44 \text{ \AA}$), and $a = 3.19 \text{ \AA}$ ($h = 3.15 \text{ \AA}$), respectively, where h is the chalcogen–chalcogen distance for the TMDs. To minimize interactions between repeating images of the 2D layers, a vacuum spacing of at least $15 \text{ \AA}$ in the vertical direction perpendicular to the 2D layer plane is employed which is sufficiently large to eliminate interplanar interactions [13].

The DFPT simulations for GaN, AlN, 6H-SiC and diamond proceed as follows. For the phonon dispersion of GaN we used a Vanderbilt ultrasoft potential for the ion-electron interactions where the electron wavefunction basis is expanded up to a cutoff energy of 50 Ry on a $7 \times 7 \times 7$ Monkhorst–Pack (MP) grid. The equilibrium lattice constants of the 4 atom hcp unit cell after structural optimization were $a = 3.207 \text{ \AA}$ and $c = 5.227 \text{ \AA}$. The convergence threshold of the self-consistent total energy and phonon calculations was 10^{-14} Ry. Similarly for AlN we used a Vanderbilt ultrasoft potential for the ion-electron interactions with a cutoff energy of 50 Ry for the electron wavefunction basis on a $7 \times 7 \times 5$ MP grid. The equilibrium lattice constants were $a = 3.11 \text{ \AA}$ and $c = 5.01 \text{ \AA}$. A Perdew–Wang (PW91) type of the generalized gradient approximation (GGA) [80] was used for the exchange-correlation functional for both GaN and AlN. For SiC we used a Goedecker–Hartwigsen–Hutter–Teter pseudopotential [81] for Si and a Von Barth–Car pseudopotential for C to treat

ion-electron interactions. Electron wavefunctions are expanded up to 50 Ry on an unshifted $6 \times 6 \times 4$ MP grid with a convergence threshold of 10^{-10} Ry. The lattice constants for the 12-atom hcp unit cell of SiC were $a = 3.081$ Å and $c = 15.12$ Å. Dynamical matrices were generated on a reduced $4 \times 4 \times 2$ MP grid with a convergence threshold of 10^{-14} Ry. For calculating the phonon dispersion of (fcc) diamond we again used a Von Barth–Car pseudopotential to treat ion–electron interactions. The electron wavefunction basis was expanded up to 24 Ry on a shifted $7 \times 7 \times 7$ MP grid with a convergence threshold of 10^{-12} Ry. Dynamical matrices were generated for a $7 \times 7 \times 7$ MP grid with a convergence threshold of 10^{-12} Ry. For both SiC and diamond we used a Perdew–Zunger type of the local density approximation (LDA) [82] to the exchange correlation functional. The pseudopotentials used in this study were chosen based on previously published DFPT simulations that well-reproduced the phonon dispersion alongside the most recent available potentials in the Quantum Espresso repositories at the time of simulation.

A.2. Debye model approximation for substrate density of states

In this section, we derive analytical expressions for the TBC based on the Debye approximation of the phonon dispersion in the substrate. First, we repeat the key ingredients of the TBC model. The thermal boundary conductance (TBC) is given by the integral of the product of the 2D-material's specific heat $C_{2D}(\omega, T)$, 2D density of states (DOS) $D_{2D}(\omega)$, and substrate scattering rate $\Gamma_{sub}(\omega)$

$$G_{sub}(T) = \int d\omega C_{2D}(\omega, T) D_{2D}(\omega) \Gamma_{sub}(\omega), \quad (A.1)$$

where the substrate scattering rate takes the following form

$$\Gamma_{sub}(\omega) = \frac{\pi K_a^2}{2 \omega^2} \frac{D_{sub}(\omega)}{m_{sub} m_{2D}}. \quad (A.2)$$

Here K_a is the van der Waals spring coupling constant in (N/m), $D_{sub}(\omega)$ is the substrate DOS, and m_{sub}/m_{2D} are the masses of the substrate and 2D-layer, respectively. If we assume the phonon dispersion in the substrate is linear with a speed of sound v_s , then the DOS $D_{sub}(\omega)$ can be well approximated by the Debye model as

$$D_{sub}(\omega) = 3 \frac{\omega^2}{2\pi^2 v_s^3} \quad (A.3)$$

where v_s is the sound velocity in the substrate. Combining this DOS with equation (A.2), we cancel out the ω^2 term and a power of π to arrive at

$$\begin{aligned} \Gamma_{sub} &= \left[\frac{\pi K_a^2}{2 \omega^2} \frac{1}{m_{sub} m_{2D}} \right] \left[3 \frac{\omega^2}{2\pi^2 v_s^3} \right] \\ &= \frac{3}{4\pi} \frac{K_a^2}{m_{sub} m_{2D} v_s^3}. \end{aligned} \quad (A.4)$$

We can now combine this $\Gamma_{sub}(\omega)$ with equation (A.1), factor out terms that are not ω -dependent, and perform the integral over the phonon spectrum. After integrating the specific heat over the frequency range, we get the following

$$G_{sub}(T) = \frac{3}{4\pi} \frac{K_a^2}{m_{sub} m_{2D} v_s^3} C_{2D}^v(T), \quad (A.5)$$

where $C_{2D}^v(T)$ is the heat capacity of the 2D material.

It is typical to normalize the D_{sub} such that the total density of modes equals the atomic density

$$\int_0^{\omega_D} 3 \frac{\omega^2}{2\pi^2 v_s^3} d\omega = \frac{3N}{V}. \quad (A.6)$$

Solving the definite integral for the Debye frequency ω_D we get

$$\omega_D = \left(\frac{6\pi^2 v_s^3 N}{V} \right)^{1/3}. \quad (A.7)$$

Introducing this DOS into equation (A.2) and then replacing the resulting Γ_{sub} in equation (A.5) we get

$$G_{sub}(T) = \frac{3\pi}{2} \frac{K_a^2}{m_{sub} m_{2D}} \frac{1}{\omega_D^3} C_{2D}^v(T). \quad (A.8)$$

However, recall equation (A.7); in the denominator if we look at the product of $m_{sub} \omega_D^3$ we recognize that

$$m_{sub} \omega_D^3 = \frac{6\pi^2 v_s^3 m_{sub} N}{V} = 6\pi^2 v_s^3 \rho_{sub} \quad (A.9)$$

with $\rho_{sub} = m_{sub} N/V$. Then equation (A.8) can be simplified further to

$$G_{sub}(T) = \frac{K_a^2}{4\pi m_{2D} v_s^3 \rho_{sub}} C_{2D}^v(T). \quad (A.10)$$

From this derivation, we conclude that the TBC should have an inverse relationship to the substrate density, speed of sound, and Debye temperature so long as the Debye model captures the phonon dispersion in the substrate.

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ORCID iDs

Zlatan Aksamija <https://orcid.org/0000-0001-9085-9641>

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