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Tuning Thermal Boundary Conductance of 2D-Substrate Interfaces by Electrostatic Forces

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Abstract— **Despite their potential for miniaturization, electronic devices made of 2D materials face thermal management challenges due to their reduced dimensionality, which can limit their efficiency and lifespan. Low thermal boundary conductance (TBC) is one major limiting factor in realizing efficient heat transfer to the substrate. Due to the roughness at the interface, the adhesion of 2D materials to their substrates tend to be weak, resulting in low TBC. Therefore, to improve heat flow from the 2D material, we need to discover novel ways of increasing TBC. In this study, we have used a numerical model combined with first-principles DFPT simulations to investigate a possible method to increase TBC using an electrostatic field due to gate voltage. Our study shows that electrostatic pressure can be used to effectively enhance TBC for an interface formed by a 2D material and a rough substrate. We find that electrostatic pressure can improve TBC by more than 300 % when an electric field of 3 V/nm is applied. This is due to an improvement in the vdW spring coupling constant, which shows a more than two-fold increase when a substrate roughness of 1.6 nm and correlation length of 10.8 nm, 2D-material's bending stiffness of 1.5 eV, and adhesion energy of 0.1 J/m^2 were used. We show that TBC is enhanced more when the substrate has a large roughness and small correlation length, and the 2D material has a large bending stiffness. This is because a stiff 2D sheet resist bending when voltage/pressure is applied, thus causing it to press more on the roughness peaks, resulting in a tremendous increase in the coupling constants at the peaks in the atomically rough surface of the substrate. However, a flexible 2D material can easily bend to conform to the topography of the rough substrate when voltage/pressure is applied, which makes the coupling constants across the interface more uniform. Here we show that TBC is enhanced more when adhesion is weak because a weak vdW bond is easily compressed by external pressure. Therefore, our study provides valuable information that can be applied in designing electronic devices with efficient heat management by using gate voltage, substrate roughness combined with the mechanical properties.**

I. INTRODUCTION

Due to their unique properties 2D materials have a wide range of potential application. Their unique properties make them attractive for application in nanoelectronics [1] and nanophotonics [2]. Their small thickness enables the miniaturization of electronic devices. However, the small size increases the density of components in integrated circuits, causing significant heating problems which prevent us from fully utilizing the advantages of 2D materials. Efficient heat dissipation from electronic device is therefore, vital for the continued miniaturization and integration of electronic devices. Low thermal boundary conductance (G) of the 2D-substrate interfaces is a key bottleneck to the realization of efficient thermal management in devices based on 2D materials [3]. Roughness at the interface determines how well a 2D material can conform to the substrate and influences the 2D-substrate van der Waals (vdW) bond, affecting phonon coupling [4], [5]. This influences G since the stiffness of the vdW bond and the area of real contact are major factors that influence heat flow across interfaces [6], [7].

Heat is transferred across the interface by vertical flexural phonons through van der Waals (vdW) coupling between the 2D material and substrate. Thus, a change in the stiffness of the interface bond is expected to influence G. For instance, using hydrostatic pressure, various research groups have demonstrated that external pressure can be used to modulate G [8], [9], [10], [8]. Applying pressure can increase the weak vdW bond to values similar to that of strongly bonded, clean interfaces, hence improving G [11], [12]. It has been shown that hydrostatic pressure of up to 1 GPa can improve G of h-BN/SiO_2 and graphene/SiO_2 interfaces by 2-3 times [8]. Like hydrostatic pressure, electrostatic field due to gate voltage (V_g) can produce a pressure (P_{elec}) that can modulate phononic heat flow across interfaces [13], [11], [10], [14], [15]. However, despite the reported impact of pressure on G, there are significant gaps in the current understanding of the underlying physics of how P_{elec} influences G of interfaces formed by 2D materials and rough substrates. For instance, the contribution of mechanical properties of 2D materials and the substrate roughness to the impact of P_{elec} on G is lacking. A comprehensive investigation of the interplay between the mechanical properties of 2D materials, substrate roughness, and P_{elec} is necessary to provide a holistic understanding of the heat-flow mechanism across 2D-substrate interfaces.

Therefore, to address this issue, we have employed a numerical modelling together with first-principles DFPT simulations to study the impact of P_{elec} on G of interfaces formed by 2D materials and rough substrate. Our study shows that P_{elec} can improve G by more than 300 % when an electric field of 3e+9 V/m is applied. We find that G is enhanced more when the substrate has a large Δ_{rms} and small L_{cor}, and the 2D material has a large bending stiffness (D_{bend}) since a stiff 2D sheet resist bending when P_{elec} is applied thus causing it to press more on the roughness peaks. This causes a tremendous increase in K_s at the roughness peaks that leads to enhanced phonon coupling at the interface. However, a

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flexible 2D material can easily bend to conform to the topography of the rough substrate when P_{elec} is applied which makes K_s values across the interface more uniform. Thus, an interface formed by a stiff 2D material, shows many-large K_s values than that formed by one that is flexible when P_{elec} is applied. Besides, we show that G is enhanced more when Γ_0 is weak because a weak vdW bond is easily compressed by P_{elec} resulting to a large increase in K_s. Therefore, our findings contribute to a better understanding of heat flow across 2D-substrate interfaces which will help in designing electronic devices with more efficient heat management.

II. METHODS

In this study, we have designed a numerical model that utilizes phonon-dispersion data computed from first-principles Density Funcitonal Theory (DFT) and Density Functional Perturbation Theory (DFPT) simulations, as implemented in the numerical code VASP. We have used graphene and SiO_2 substrate as our sample material. We model the adhesion of the 2D sheet on the atomically rough substrate as described in our earlier work [16]. Then, add P_{elec} due to applied V_g, as illustrated in Fig. 1. Applying V_g produce an electric field (F) that induce a layer of charge in the 2D material. This result in an electrostatic force (F_{elec}) between the 2D material and a gate which pull the 2D sheet toward the substrate.

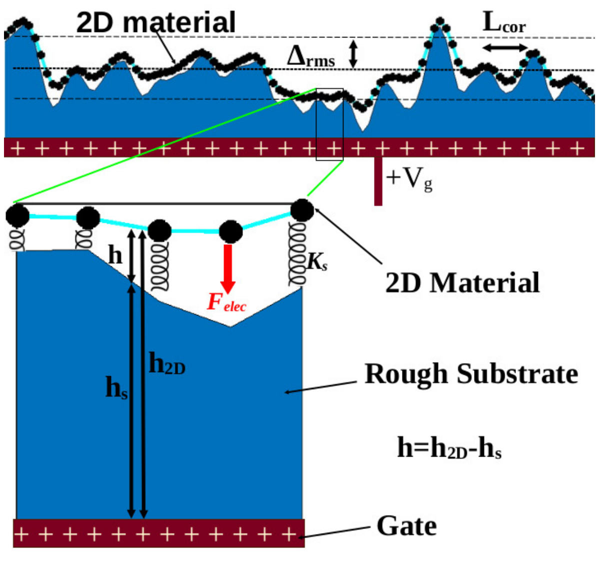

Fig. 1. A schematic showing a single layer of 2D material on a rough substrate and a gate. The 2D material cannot bend enough to conform fully to the rough substrate. We have regions that are in contact and others that are delaminated. This leads to variation in the 2D-substrate distance (h) across the interface.

Here the electrostatic force is expressed as $F_{elec}(h,V) = \frac{\epsilon A}{2}(\frac{V_g}{h_s+h})^2$ then the electrostatic pressure is given by $P_{elec}(h,V) = \frac{\epsilon}{2}(\frac{V_g}{h_s+h})^2$, where A is area, ϵ is the dielectric constant of SiO_2 taken here as 3.7, V_g is the applied gate voltage, h_s is the thickness of the dielectric material, and h is the 2D-substrate distance illustrated in Fig. 1. To get the total energy of the system, we calculate the sum of the vdW energy, bending energy, and the flexural energy as described in Ref. [16], and the energy due to electrostatic field expressed as $U_{elec} = F_{elec} \times (h_s + h)$. We then initialize the 2D-substrate distance to the equilibrium value for a flat substrate h_0 taken here as 0.6 nm [17] and adhesion energy (Γ_0) of 0.1 J/m^2 [17] then minimize the total energy by changing h using a quasi-Newton algorithm. This gives us the relaxed h values at every point in the interface. Using the optimized values of h we calculate K_s as described in our previous work [16]. Finally, we use a Landauer formalism that uses K_s values to find G at each interface point as described in Ref. [18], [16]. We determine the induced electron concentration by solving the quadratic relation $en^2 - n\epsilon F - en_i^2 = 0$, where e is the electron charge, n is the electron concentration, n_i is the intrinsic electron concentration, and F is the electrostatic field due to the applied V_g. Then the hole concentration, p, is calculated as $p = n_i^2/n$. Our model allows us to study the impact of P_{elec} on G and the contribution of roughness and the mechanical properties of the 2D material.

III. RESULTS AND DISCUSSION

By applying a gate voltage, we induce a net charge concentration on the 2D material that causes an attractive force (F_{elec}) that pull the 2D material towards the substrate. Unlike setups where hydrostatic pressure is used, here F_{elec} can cause the 2D sheet to bend and conform to the substrate roughness without destroying roughness peaks. As illustrated in Fig. 2(a), P_{elec}, increases quadratically with increasing V_g while the square of induced net charge carrier density, $(n-p)^2$, varies linearly with V_g. Substrate roughness causes variations in h with some values being far larger than h_0. However, applying P_{elec} significantly reduces h values as shown in Fig. 2(b). As a result, a significant increase in the K_s is observed, as shown in Fig. 2(c). We note that K_s values at roughness peaks, which are already significant, are enhanced more when we apply P_{elec}. This leads to large improvement in G at these regions due to better phonon coupling, as shown in Fig. 2(d). However, due to the variation in h we observe disparities in K_s between different regions which leads to corresponding variations in G since it is proportional to K_s [18], [16].

Our analysis of K_s when an electric field of 3e+9 V/m is applied reveals that K_s increases linearly with the $(n-p)^2$ and P_{elec} as shown in Fig. 3(a). We observe 244 %, 70 %, and 48 % increase in K_s for Δ_{rms} of 1.6 nm, 0.9 nm, and 0 nm respectively and L_{cor} of 10.8 nm. Likewise Fig. 3(b) shows that G is also linear with $(n-p)^2$ and P_{elec} which is in agreement with previous studies [13]. We observe an increase in G of 333 %, 92 %, and 80 % for interface with Δ_{rms} of 1.6 nm, 0.9 nm, and 0 nm respectively and L_{cor} of 10.8 nm. The increase in G is because external pressure enhance phonon transmission

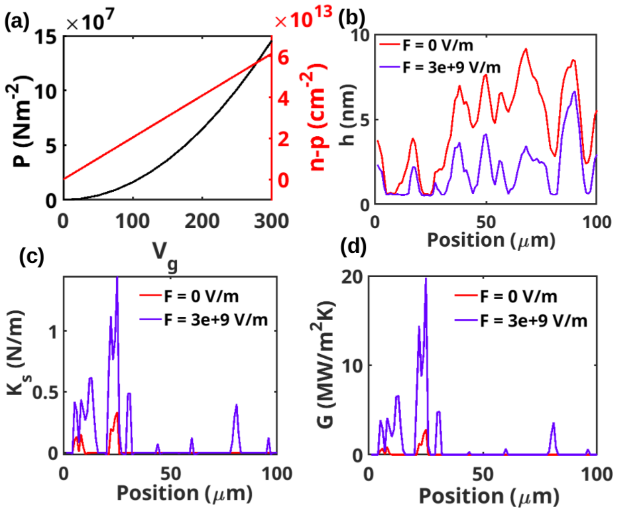

Fig. 2. (a) P_{elec} and $(n-p)$ as a function of V_g. (b) h, (c) K_s, and (d) G as a function of position for a substrate with Δ_{rms} of 2.5 nm and L_{cor} of 8.8 nm.

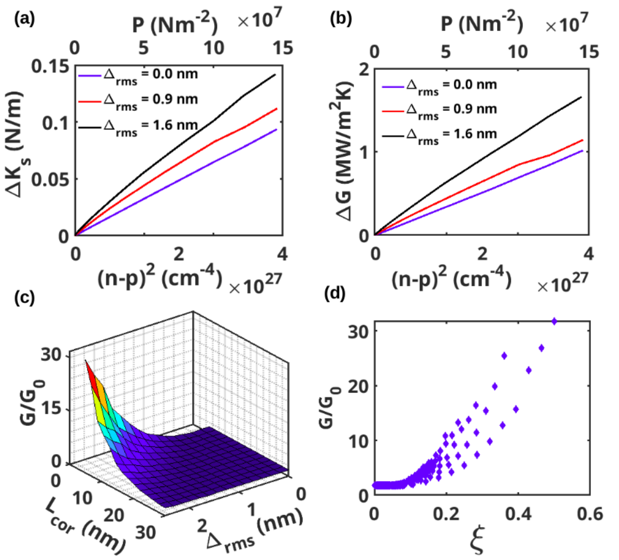

Fig. 3. Analysis of K_s and G at 300 K (a) the difference between K_s at an electric field of 3e+9 V/m and at 0 V/m, ΔK_s and (b) the difference between G at an electric field of 3e+9 V/m and G_0 at 0 V/m, ΔG as a function of $(n-p)^2$ and P_{elec} for interface with different Δ_{rms} values and L_{cor} of 10.8 nm. (c) The ratio of G and G_0 as a function of Δ_{rms} and L_{cor}. (d) The ratio of G and G_0 as a function of surface slope (ξ).

across the interface. According to a previous study, this enhancement saturate when the vdW bond at the interface attain a stiffness similar to the stiffness of the bonds in the bulk of the individual materials [19]. However, P_{elec} is not large enough to achieve such a strong bond at the interface. As observed in Fig. 3(c), G increases more when Δ_{rms} is large and L_{cor} is small, which implies that G is improved more when surface slope (ξ), given by $\xi = \Delta_{rms}/L_{cor}$, is large as shown in Fig. 3(d). However, for small ξ values, G remains fairly constant before increasing almost linearly with increasing ξ.

To better understand the impact of substrate roughness on G, we analyze the effect of varying Γ_0 since substrate roughness is expected to influence the adhesion between the 2D material and the substrate. We find that G is enhanced more when Γ_0 is weak and the ξ is large as shown in Fig. 4(a). This is due to the fact that it is easier to compress the weak vdW bonds and significantly reduce h values increasing K_s which leads to an enhancement in G.

As shown in 4(b), we observe that G increases more when the bending stiffness of the 2D material (D_{bend}) is large. This can be explained by considering how the 2D sheet interacts with a rough substrate; the large K_s values at the peaks of the roughness features dictate the heat flow across the interface, as seen in 2(c) and (d) and in our previous study [16]. If D_{bend} is small the 2D materials can easily bend to follow the topography of the rough-substrate surface which leads to a more uniform K_s values. However, if the 2D material has a large D_{bend}, applying P_{elec} pushes the 2D material towards the substrate so that the substrate distance at roughness peaks becomes very small; hence, we get very large K_s values at these regions. As illustrated in Fig. 4(c), we find that interfaces formed by stiff 2D material show many-large K_s values than that formed by a flexible material with small D_{bend}). Therefore, G is enhanced more when D_{bend} is large.

IV. CONCLUSIONS

In this study, we have demonstrated that electrostatic pressure (P_{elec}) due to a gate voltage (V_g) is a viable approach to improve thermal boundary conductance (G) and, hence, thermal management in electronic devices made of 2D materials. We show that applying P_{elec} on graphene/SiO_2 interface can increase K_s by more than two-folds and cause G to increase by more than 300 % when an electric field of 3e+9 V/m is applied. Our study reveals that G is enhanced more when the substrate has a large Δ_{rms} and small L_{cor}, and the 2D material has a large bending stiffness (D_{bend}). This is because the vdW spring coupling constant (K_s) at roughness peaks is large due to the small 2D-substrate distance (h) at these regions. Therefore, at the roughness peaks we have better phonon coupling that enhance the overall heat flow across the interface. A stiff 2D sheet resist bending when P_{elec} is applied thus causing it to press more on the roughness peaks. This causes a tremendous increase in K_s at the roughness peaks that leads to even better phonon coupling. However, a flexible 2D material can easily bend to conform to the topography of the rough substrate when P_{elec} is applied which makes K_s values across the interface more uniform. Thus, an interface formed by a stiff 2D material, shows many-large K_s values than that formed by one that is flexible when P_{elec} is applied. Due to the roughness at the interface, the average adhesion of 2D materials to their substrates (Γ_0) tend to be weak. Here we show that G is enhanced more when Γ_0 is

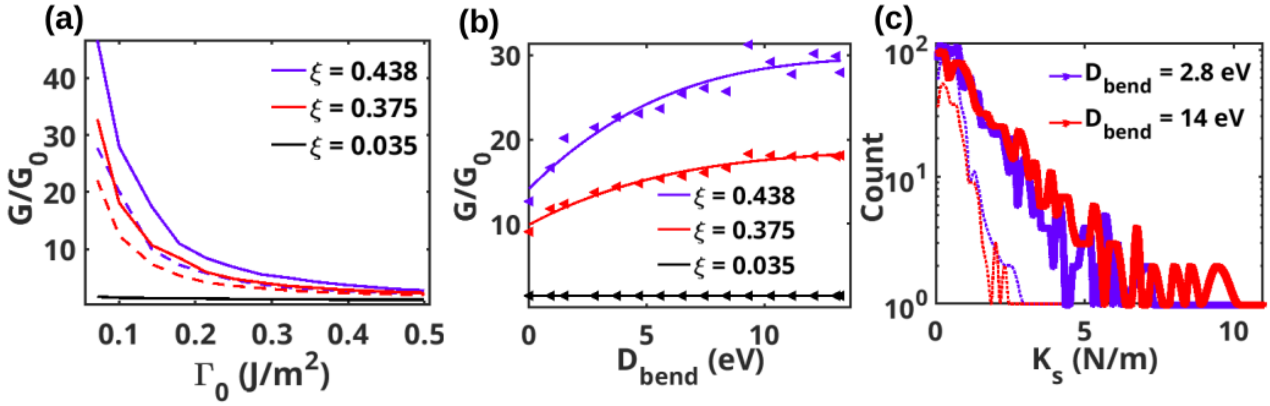

Fig. 4. (a) The ratio of G (at an electric field of 3×10^9 V/m) and G_0 (no electric field) as a function of Γ_0 for D_{bend} of 1.5 eV (broken lines) and 13.24 eV (full lines) showing that G is enhanced more when Γ_0 is weak and ξ is large. (b) The ratio of G and G_0 as a function of D_{bend} for different values of ξ and Γ_0 of 0.1 J/m^2 where the symbols show the data points and the line is a fit. We observe that G is enhanced more when D_{bend} and ξ are large. (c) The K_s values when an electric field of 3e+9 V/m is applied (full lines) and when there is no electric field (dotted lines) showing that in a stiffer 2D sheet, application of electric field leads to many-large K_s values than when the 2D sheet is flexible.

weak because a weak vdW bond is easily compressed by P_{elec} resulting to a large increase in K_s. Thus, our study demonstrates how gate voltage, substrate roughness, and mechanical properties of 2D materials can be leveraged to enhance thermal performance in electronic devices made of 2D materials.

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