Theoretical analysis of thermal boundary conductance of MoS$_2$-SiO$_2$ and WS$_2$-SiO$_2$ interface

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Abstract

Understanding the physical processes involved in interfacial heat transfer is critical for the interpretation of thermometric measurements and the optimization of heat dissipation in nanoelectronic devices that are based on transition metal dichalcogenide (TMD) semiconductors. We model the phononic and electronic contributions to the thermal boundary conductance (TBC) variability for the MoS$_2$-SiO$_2$ and WS$_2$-SiO$_2$ interface. A phenomenological theory to model diffuse phonon transport at disordered interfaces is introduced and yields $G = 13.5$ and $12.4$ MW K$^{-1}$ m$^{-2}$ at 300 K for the MoS$_2$-SiO$_2$ and WS$_2$-SiO$_2$ interface, respectively. We compare its predictions to those of the coherent phonon model and find that the former fits the MoS$_2$-SiO$_2$ data from experiments and simulations significantly better. Our analysis suggests that heat dissipation at the TMD-SiO$_2$ interface is dominated by phonons scattered diffusely by the rough interface although the electronic TBC contribution can be significant even at low electron densities ($n \leq 10^{12}$ cm$^{-2}$) and may explain some of the variation in the experimental TBC data from the literature. The physical insights from our study can be useful for the development of thermally aware designs in TMD-based nanoelectronics.

Keywords: transition metal dichalcogenides, phonon, thermal boundary conductance, 2D materials

(Some figures may appear in colour only in the online journal)

1. Introduction

Atomically thin two-dimensional (2D) transition metal dichalcogenide (TMD) semiconductors such as MoS$_2$ and WS$_2$ hold great potential for the development of next-generation electronic devices [1, 2]. At the nanoscale, high power densities in these devices require efficient thermal management crucial for optimal device performance, with the thermal boundary conductance (TBC) of the 2D crystal-substrate interface playing a key role in the dissipation of excess Joule heat [3, 4]. Therefore, clearer insights into the role of the different physical mechanisms underlying the TBC of the TMD-substrate interface may lead to superior thermally aware TMD-based nanoelectronic device designs.

One widely studied mechanism is the van der Waals coupling between the phonons of the 2D crystal and its substrate which is believed to be the dominant component in the overall TBC [4]. The phononic TBC ($G_{ph}$) has been estimated using molecular dynamics (MD) simulations [5, 6], elasticity theory [7–9] and density functional theory-based models [10]. Another mechanism of heat dissipation is through the inelastic scattering of electrons in the 2D crystal by dipoles in the dielectric substrate, a phenomenon known widely as ‘remote phonon (RP) scattering’ or ‘surface optical (SO) phonon scattering’ [11–14]. This inelastic scattering mechanism, which plays an important role in limiting the electron mobility in TMDs [15, 16], also underlies the electronic TBC ($G_{el}$) and depends strongly on the electron or hole...
density. Although it is predicted to be insubstantial for heat dissipation in graphene [17, 18], the contribution from RP scattering may be significant and comparable to the phononic TBC for MoS2 and WS2 [19].

Nonetheless, even though the phononic TBC has been studied using various theoretical methods, it is useful to have a model that relates $G_{ph}$ to the fundamental elastic properties of the 2D crystal and its substrate while accounting for quantum statistics at low temperatures. One such theory that is based on coherent flexural phonons [7] and developed in [8] yields relatively accurate values for the graphene-SiO2 interface [8, 9] and also predicts a modest room-temperature phononic TBC of $\sim 3.1$ MW K$^{-1}$ m$^{-2}$ for the MoS2-SiO2 interface, in good agreement with earlier published data [20] but considerably smaller than later more accurately measured TBC values [21, 22] in the range of 10–15 MW K$^{-1}$ m$^{-2}$. This substantial discrepancy necessitates a relook of the heat dissipation mechanisms at the TMD-substrate substrate.

In our paper, we model the relative contribution of the different physical mechanisms to the overall TBC of the MoS2-SiO2 and WS2-SiO2 interface. Our objective is to understand how these mechanisms (e.g. coherent versus diffuse phonon transport and the electron density dependence of $G_d$) lead to the variability in their TBC. We revisit the phononic TBC problem and introduce a phenomenological diffuse phonon theory that is more suitable for atomically disordered interfaces and allows us to model the effect of disorder on the TBC. To understand its difference to the coherent theory, we compare the TBC predictions from the coherent and diffuse phonon models with data from experiments and MD simulations. We also compare the phononic contribution (coherent or diffuse) to the electronic contribution in the TBC. Finally, we use the combined electronic and phononic BC results ($G = G_{el} + G_{ph}$) to analyze reported experimental TBC data [20–23] for the MoS2-SiO2 interface and to discuss the possible physics underlying the variability of the TBC data. We also use the combined electronic and phononic BC results to predict the variability of the TBC for the WS2-SiO2 interface. Although the theoretical methods in this paper are used specifically to analyze the TBC variability of the single-layer MoS2-SiO2 and WS2-SiO2 interface, they can also be extended to other 2D crystals (e.g. graphene and other TMDs) and substrates (e.g. Al2O3).

2. Theoretical models

We discuss our theoretical models of the phononic and electronic TBC processes, as depicted in the schematic of the TMD-substrate interface in figure 1, in the following subsections. Roughly speaking, we attribute the phononic TBC ($G_{ph}$) to the linear mechanical coupling between the flexural phonons of the TMD sheet and the bulk elastic waves (i.e. acoustic phonons) of the substrate, and the electronic TBC ($G_{el}$) to the remote scattering of the TMD electrons by the polar optical phonons of the dielectric substrate, also otherwise known as ‘RP scattering’ [11, 19]. We assume that the two TBC components are independent (i.e. the electron–phonon interaction has no effect on the mechanical coupling between the TMD and the substrate and vice versa) and can be added in parallel.

2.1. Heat dissipation from coherent and diffuse flexural phonon scattering

We adopt a linear elasticity theory-based approach to model the phononic TBC from [8]. Although the model is applied to the TMD-SiO2 interface here, it is sufficiently general to be applied to any linear elastic isotropic substrate. The model, which assumes heat dissipation by coherent 2D flexural phonons at a perfectly smooth interface [7], is used to derive the following expression for the coherent phononic TBC

$$G_{coh} = \frac{1}{2\pi} \int_0^\infty d\omega \frac{\hbar \omega}{\omega - i\gamma} \Xi_{coh}(\omega),$$

(1)

where

$$\Xi_{coh}(\omega) = \frac{1}{(2\pi)^2} \int_{q < q_{max}} d^2q 4K^2 \text{Im} D_{coh}(q, \omega) \text{Re} D_{coh}(q, \omega)$$

(2)

is the coherent areal transmission function, $q_{max}$ is the maximum wave vector which we can set as $q_{max} = 2\pi/\sqrt{\Lambda} \approx 2 \times 10^{10}$ m$^{-1}$ (A is the unit cell area of the TMD), and

$$N(\omega, T) = \exp \left( \frac{\hbar \omega}{k_B T} - 1 \right)$$

is the Bose–Einstein occupation factor at frequency $\omega$ and temperature $T$. The transmission function in equation (2) depends on the spring constant at the TMD-substrate interface $K$, the retarded Green’s function for the flexural motion of the TMD monolayer $D_{2D}(q, \omega)$, and the retarded Green’s function for the free surface displacement of the isotropic solid substrate $D_{coh}(q, \omega)$. The value of $K$ for the MoS2-SiO2 interface is taken from [8] while its value for the WS2-SiO2 interface is calculated using density functional theory like in [8].

The expressions for $D_{2D}(q, \omega)$ and $D_{coh}(q, \omega)$ are [8]

$$D_{2D}(q, \omega) = \lim_{\eta \to 0} [\rho \omega^2 + i\rho \gamma(\omega) - \kappa q^4 + i\eta]^{-1},$$

(3)
where $\rho$ and $\kappa$ denote the areal mass density and the bending stiffness of the uncoupled 2D crystal, respectively, and

$$D_{ab}(q, \omega) = \frac{i}{\rho_{ab}c_T^2} \frac{p_L(q, \omega)}{S(q, \omega)} \left( \frac{\omega}{c_T} \right)^2 \Theta(\omega_D - \omega),$$

(4)

where

$$S(q, \omega) = \left[ \left( \frac{\omega}{c_L} \right)^2 - 2q^2 \right]^2 + 4q^2p_Tp_L,$$  \hspace{1cm} (5a)

$$p_L(q, \omega) = \lim_{\eta \to 0^+} \left[ \left( \frac{\omega}{c_L} \right)^2 - q^2 + i\eta \right]^{1/2},$$  \hspace{1cm} (5b)

$$p_T(q, \omega) = \lim_{\eta \to 0^+} \left[ \left( \frac{\omega}{c_T} \right)^2 - q^2 + i\eta \right]^{1/2},$$  \hspace{1cm} (5c)

and $c_L$, $c_T$ and $\rho_{ab}$ are the longitudinal and transverse speed of sound, and the voluminal mass density of the substrate, respectively. In equation (4), $\Theta(\ldots)$ represents the Heaviside function, and $\omega_D = (6\pi^2N_{sub})^{1/3}c_L = 9.39 \times 10^{13}$ rad s$^{-1}$ (or 61.8 meV) and $N_{sub} = 6.62 \times 10^{28}$ m$^{-3}$ are respectively the longitudinal Debye frequency and number density [24] of the substrate (amorphous SiO$_2$). As in [8], $\gamma(\omega)$ in equation (3) is the frequency-dependent damping coefficient representing the intrinsic damping of the flexural motion, given as $\gamma(\omega) = \frac{\omega_T}{\alpha}c_T$ where $\alpha$ is the ratio of the phonon lifetime to its period at room temperature and $T_{RT}$ is the room temperature (300 K). It has been shown [8] that it is necessary to include the intrinsic damping of the flexural motion in equation (3) to account for the interfacial heat dissipation correctly.

Although the predictions of the coherent flexural phonon model are relatively accurate for graphene [8, 9], its predictions for the MoS$_2$-SiO$_2$ interface are substantially smaller than experimental data [21, 22], highlighting a possible shortcoming of the model. The theory in equation (2) assumes a perfectly smooth interface [7] which limits energy transfer to processes that conserve parallel momentum, a restriction that may not apply to the more disordered TMD-substrate interface which can have surface roughness, trapped charged impurities and surface adsorbates [25]. For a highly disordered interface, this assumption can be relaxed and we may assume that each incoming phonon is scattered by the disordered interface without constraints to all available outgoing phonon modes with equal probability like in the diffuse mismatch model (DMM) [26]. Using this assumption, we apply the DMM approximation to modify equation (1) by writing the diffuse transmission function between the 2D crystal and the substrate as [27]

$$\Xi_{diff}(\omega) = \frac{M_{2D}(\omega)M_{sub}(\omega)}{M_{2D}(\omega) + M_{sub}(\omega)},$$

(6)

where $M_{2D}(\omega)$ and $M_{sub}(\omega)$ are the number of available transmitting modes per unit area at frequency $\omega$ in the 2D crystal and substrate, respectively. We stress that equation (6) describes a phenomenological model derived from purely kinetic considerations of detailed balance [26, 27] and ignores the details of the interactions at the interface. The corresponding TBC is

$$G_{ph}^{diff} = \frac{1}{2\pi} \int_0^\infty d\omega \frac{dN_2D(\omega, T)}{dT} \Xi_{diff}(\omega).$$

(7)

To determine $\Xi_{diff}(\omega)$ in equation (6), we need to count the number of modes in the 2D crystal and the substrate that contribute to cross-plane phonon transport at each frequency $\omega$. This problem is particularly tricky for a single-layer 2D crystal since it has no extended volume in the cross-plane direction as shown in figure 2(a). Instead, we estimate $M_{2D}(\omega)$ in our model phenomenologically by assuming that the interface between the 2D crystal and a substrate consisting of a semi-infinite number of the same 2D material is acoustically transparent, e.g. a single-layer MoS$_2$ (1L-MoS$_2$) on a bulk MoS$_2$ (lim$_{N \to \infty}$NL-MoS$_2$) substrate, such that the transmission function between the two materials is equal to the number of transmitting modes in the single-layer 2D crystal. To do this, we make the replacement $K \to K_{2D}$ and $D_{sub}(q, \omega) \to D_{2D,\infty}(q, \omega)$ in equation (2), where

$$D_{2D,\infty}(q, \omega) = \Theta(z)D_z(q, \omega) + \Theta(-z)D_{-z}(q, \omega),$$

(8)

is the surface response function of a semi-infinite 2D layered substrate [28], with

$$D_z(q, \omega) = \frac{2 \sqrt{(q, \omega)^2 - 4z(q, \omega)K_{2D}}}{z(q, \omega) + \sqrt{(q, \omega)^2 - 4z(q, \omega)K_{2D}}}.$$
for \( \mathbf{z}(q, \omega) = \lim_{\omega \to 0} d_{2D}(q, \omega)^{-1} = \rho q^2 - kq^4 \), and \( K_{2D} \) is the interlayer spring constant in the substrate. Hence, we obtain the expression analogous to equation (2), i.e.

\[
M_{2D}(\omega) = \frac{1}{(2\pi)^3} \int_{|q| < q_{\text{max}}} d^3q \left( \frac{4K_{2D}^2 \text{Im} D_{2D,\infty}(q, \omega) \text{Im} D_{2D}(q, \omega)}{|1 - K_{2D} D_{2D,\infty}(q, \omega) + D_{2D}(q, \omega)|^2} \right),
\]

which can be evaluated numerically. The set up of the calculation for \( M_{2D}(\omega) \) for a single-layer TMD (1L-TMD) is shown in figure 2(b). In the case of SiO\(_2\) as shown in figure 2(c), we estimate the number of modes per unit area in an isotropic elastic substrate \( M_{\text{sub}}(\omega) \) as

\[
M_{\text{sub}}(\omega) = \frac{\omega^2}{2\pi} \left( \frac{1}{c_L^2} + \frac{2}{c_T^2} \right) \Theta(\omega_D - \omega).
\]

Since \( M_{2D}(\omega) \ll M_{\text{sub}}(\omega) \) for \( \omega < \omega_D \), we have \( \Xi_{\text{diff}}(\omega) \approx M_{2D}(\omega) \Theta(\omega_D - \omega) \) and equation (7) becomes

\[
G_{\text{ph}}^{\text{diff}} = \frac{1}{2\pi} \int_0^{\omega_D} d\omega \frac{dN(\omega, T)}{dT} M_{2D}(\omega)
\]

which depends on the substrate through its longitudinal Debye frequency \( \omega_D \) and the 2D crystal though \( M_{2D}(\omega) \). The expression in equation (11) suggests that in the diffuse limit, the phononic TBC for a particular 2D crystal can be maximized only through \( \omega_D \) which depends on \( c_L \). Hence, the phononic TBC would be high for hard insulators such as diamond and sapphire for which \( c_L = 17500 \) and \( 10890 \text{ m s}^{-1} \), respectively [26]. We use the simulation parameters from table 1 for our \( G_{\text{ph}} \) calculations.

2.2. Heat dissipation by RP scattering

To model the electronic TBC \( G_{\text{el}} \), we use the theory developed in [19]. The expression for \( G_{\text{el}} \) is written as a 2D integral that can be evaluated numerically, i.e.

\[
G_{\text{el}} = \sum_{\gamma = \text{SO1,SO2}} \int_0^{q_{\text{max}}} dq \int_0^{\infty} d\mu \left( \frac{e^2 h^2 \omega^2}{4k_B T \cosh^2 \left( \frac{\omega}{2k_BT} \right)} \right)
\]

\[
\times \frac{1}{N(\omega, T) + 1} \frac{1}{N(\omega, T)} \left( \frac{1}{\epsilon_{\gamma,hi}} - \frac{1}{\epsilon_{\gamma,lo}} \right)
\]

where \( q_{\text{max}} = 2 \times 10^{10} \text{ m}^{-1} \) is the cutoff wave vector like in equation (2), \( \gamma \) is the frequency of the \( \gamma \) phonon, \( d \) is the TMD-substrate gap size, \( \mu \) is the chemical potential, and

\[
F_{\gamma} = \frac{e^2 h^2 \omega^2}{4k_B T} \left( N(\omega, T) + 1 \right) \left( \frac{1}{\epsilon_{\gamma,hi}} - \frac{1}{\epsilon_{\gamma,lo}} \right)
\]

In equation (12), \( P(q, \omega; \mu, T) \) is the electron screening function [19] while the screening function \( \varepsilon(q) \) is given by [31]

\[
\varepsilon(q)^{-1} = 1 + \frac{e^2}{2\epsilon_0 q} \text{Re} P(q, 0; \mu, T) \left[ 1 - \frac{\varepsilon(q)^{-1}}{\varepsilon(q) + \epsilon_0} \exp(-2qd) \right],
\]

where \( e \) and \( \epsilon_0 \) are the electron charge and the permittivity of vacuum, respectively. In equation (13), the expressions for \( \varepsilon_{\text{SO1,hi}}, \varepsilon_{\text{SO1,lo}}, \varepsilon_{\text{SO2,hi}}, \varepsilon_{\text{SO2,lo}} \) and \( \varepsilon_{\text{SO1,hi}}, \varepsilon_{\text{SO1,lo}}, \varepsilon_{\text{SO2,hi}}, \varepsilon_{\text{SO2,lo}} \) are given by [31].

We use the simulation parameters from table 2 for our \( G_{\text{el}} \) calculations. The longitudinal optical (LO) phonon frequencies \( \omega_{\text{LO1}} \) and \( \omega_{\text{LO2}} \) are determined from the zeros of \( \varepsilon_{\text{sub}}(\omega) = \varepsilon_{\text{sub}}^{-1} + (\varepsilon_{\text{sub}}^{-1} - \varepsilon_{\text{LO1}}^{-1}) \frac{\omega^2}{\omega^2 - \omega_{\text{LO1}}^2} + (\varepsilon_{\text{sub}}^{-1} - \varepsilon_{\text{LO2}}^{-1}) \frac{\omega^2}{\omega^2 - \omega_{\text{LO2}}^2} \), while the SO phonon frequencies \( \omega_{\text{SO1}} \) and \( \omega_{\text{SO2}} \) are determined from solving \( \varepsilon_{\text{sub}}(\omega) + \epsilon_0 = 0 \) [13].

3. Numerical results and discussion

3.1. Comparison of coherent and diffuse phononic TBC

It is intuitively expected that disorder in the interface leads to higher thermal resistance because of increased phonon scattering [34]. However, the presence of interfacial disorder can enlarge the scattering phase space and possibly improve interfacial thermal transport by allowing energy transfer to proceed through scattering pathways that do not conserve parallel momentum [35]. This difference in scattering is manifested in the transmission spectra \( \Xi_{\text{coh}}(\omega) \) and \( \Xi_{\text{diff}}(\omega) \) from equations (2) and (6) in figure 3(a) which shows a marked difference in the transmission spectra for the MoS\(_2\)-SiO\(_2\) interface, especially at low frequencies (\( \omega < 12 \text{ meV} \)) where \( \Xi_{\text{diff}}(\omega) \) is substantially higher than \( \Xi_{\text{coh}}(\omega) \). The sharp drop in the transmission spectra observed at 61.8 meV is due to the frequency cutoff at the longitudinal Debye frequency of the substrate.

To understand the implications of the difference in the transmission spectra between the ordered and disordered TMD-substrate interface, we compare \( G_{\text{ph}}^{\text{coh}} \) and \( G_{\text{ph}}^{\text{diff}} \) of equations (1) and (7) for \( T = 100 \) to 600 K, using the parameters in table 1, for the WS\(_2\)-SiO\(_2\) and MoS\(_2\)-SiO\(_2\) interface in figure 3(b). For the MoS\(_2\)-SiO\(_2\) interface at \( T = 300 \) K, we obtain \( G_{\text{ph}}^{\text{diff}} = 13.5 \text{ MW K}^{-1} \text{ m}^{-2} \), which is significantly larger than \( G_{\text{ph}}^{\text{coh}} = 3.1 \text{ MW K}^{-1} \text{ m}^{-2} \) and more comparable to the value of \( G = 14.0 \pm 3.7 \text{ MW K}^{-1} \text{ m}^{-2} \) at 311 K in [21], \( G = 20.3-33.5 \text{ MW K}^{-1} \text{ m}^{-2} \) at 295 K in [23], \( G = 21.0 \text{ MW K}^{-1} \text{ m}^{-2} \) at 300 K in [36] and \( G = 18.6 \text{ MW K}^{-1} \text{ m}^{-2} \) at 300 K in [37]. This suggests that the MoS\(_2\)-SiO\(_2\) interface is disordered, possibly connected to the absence of long-range order in a-SiO\(_2\) and chemical inhomogeneity of its surface [25], and that its thermal transport properties are better described by the diffuse phonon model. The larger \( G_{\text{ph}}^{\text{diff}} \) for the MoS\(_2\)-SiO\(_2\) interface is also closer to classical MD simulation results (\( G = 15.6 \text{ MW K}^{-1} \text{ m}^{-2} \) in [23], \( G = 12.2-23.5 \text{ MW K}^{-1} \text{ m}^{-2} \) in [38] and \( G = 25.6 \pm 3.3 \text{ MW K}^{-1} \text{ m}^{-2} \) in [6]). For the WS\(_2\)-SiO\(_2\) interface at 300 K, we also find that \( G_{\text{ph}}^{\text{diff}} = 12.4 \text{ MW K}^{-1} \text{ m}^{-2} \) is significantly larger than \( G_{\text{ph}}^{\text{coh}} = 3.0 \text{ MW K}^{-1} \text{ m}^{-2} \).
Figure 3. (a) Coherent and diffuse phonon transmission spectra for the MoS2-SiO2 interface at 300 K from equations (2) and (6), respectively. (b) Temperature dependence of the coherent (dotted lines) and the diffuse (solid lines) phononic TBC ($G_{\text{coh}}^\text{ph}$ versus $G_{\text{diff}}^\text{ph}$) for the WS2-SiO2 (blue lines) and MoS2-SiO2 (red lines) interface from $T$ = 100–600 K. At 300 K, we have $G_{\text{coh}}^\text{ph} = 3.1$ MW K$^{-1}$ m$^{-2}$ and $G_{\text{diff}}^\text{ph} = 13.5$ MW K$^{-1}$ m$^{-2}$ ($G_{\text{coh}}^\text{ph} = 3.0$ MW K$^{-1}$ m$^{-2}$ and $G_{\text{diff}}^\text{ph} = 12.4$ MW K$^{-1}$ m$^{-2}$) for the MoS2-SiO2 (WS2-SiO2) interface.

Table 1. Parameters in our numerical simulations of equations (1) and (7). $K$, which depends on the type of 2D crystal, is the spring constant per unit area for the OH-terminated SiO2 interface and is calculated using DFT [8]. $\kappa$ and $\rho$ are respectively the intrinsic bending rigidity and mass density per unit area of the 2D crystal used in equation (3). $K_{2D}$ is the spring constant per unit area for the bulk version of the 2D crystal [29].

| TMD   | WS2 | MoS2 |
|-------|-----|------|
| $K$ (10$^{19}$ N m$^{-2}$) | 6.12 | 4.94 |
| $\kappa$ (eV) | 11.25 [30] | 9.61 [30] |
| $\rho$ (10$^{-7}$ kg m$^{-2}$) | 47.9 | 31.3 |
| $\alpha$ | 100 [8] |  |
| $\rho_{\text{sub}}$ (kg m$^{-3}$) | 2200 [7] |  |
| $c_L$ (m s$^{-1}$) | 5953 [7] |  |
| $c_T$ (m s$^{-1}$) | 3743 [7] |  |
| $\omega_2$ (meV) | 61.8 |  |
| $K_{2D}$ (10$^{19}$ N m$^{-3}$) | 9.55 [29] | 8.90 [29] |

Table 2. Remote phonon scattering simulation parameters for WS2 and MoS2 [32]. The effective electron masses $m_e$ are expressed in terms of the free electron mass $m_0$ and taken from [32]. The variables $m_e$, $g_s$, and $g_v$ are used in $P(q, \omega; \mu, T)$. The parameters $\epsilon_0$, $\epsilon_{\text{sub}}$, $\omega_{\text{TO1}}$ and $\omega_{\text{TO2}}$ for SiO2 are taken from [33].

| Substrate | WS2 | MoS2 |
|-----------|-----|------|
| $m_e/m_0$ | 0.31 | 0.51 |
| $d$ (Å) | 3.0 [19] |  |
| $g_s$ | 2 |  |
| $g_v$ | 2 |  |
| $\epsilon_{\text{sub}}/\epsilon_0$ | 2.50 |  |
| $\epsilon_0/\epsilon_0$ | 3.05 |  |
| $\omega_{\text{TO1}}$ (meV) | 55.60 |  |
| $\omega_{\text{TO2}}$ (meV) | 138.10 |  |
| $\omega_{\text{TO1}}$ (meV) | 60.99 |  |
| $\omega_{\text{TO2}}$ (meV) | 148.97 |  |

3.2. Interpretation of experimental data for MoS2-SiO2 interface

We combine the results for $G_{\text{coh}}^\text{ph}$, $G_{\text{coh}}^\text{el}$ and $G_{\text{diff}}^\text{ph}$ to analyze the experimental TBC data ($G_{\text{expt}}$) for the MoS2-SiO2 interface from Yalon et al [21]. As mentioned earlier, we assume that the phononic and electronic TBC components are independent and can be added in parallel, and that the electronic TBC is unaffected by the disorder at the TMD-substrate interface. From Figure 4, we plot $G_{\text{coh}}^\text{ph} + G_{\text{coh}}^\text{el} + G_{\text{diff}}^\text{ph}$, which captures the essential physics of heat dissipation at the MoS2-SiO2 interface.

experimentally derived values for the MoS2-SiO2 interface suggests that the real TBC value for the WS2-SiO2 interface is probably closer to the predicted $G_{\text{diff}}^\text{ph} = 12.4$ MW K$^{-1}$ m$^{-2}$. We also note that the $G_{\text{coh}}^\text{ph}$ and $G_{\text{diff}}^\text{ph}$ values for the WS2-SiO2 and MoS2-SiO2 interface are comparable because of their similar elasticity parameters ($\rho$, $\kappa$ and $K_{2D}$).
Figure 4. Temperature dependence of (a) $G_{\text{coh}}^\text{ph}$ (thick dashed line labeled ‘Coherent ph.’) and $G_{\text{coh}}^\text{ph} + G_{\text{el}}$ (fine purple dashed lines labeled ‘Coherent ph. + RP’), and (b) $G_{\text{diff}}^\text{ph}$ (thick solid line labeled ‘Diffuse ph.’) and $G_{\text{diff}}^\text{ph} + G_{\text{el}}$ (fine green solid lines labeled ‘Diffuse ph. + RP’) for the MoS$_2$-SiO$_2$ interface at different values of the electron density between $n = 10^{11}$ to $10^{12}$ cm$^{-2}$ in steps of $\Delta n = 10^{11}$ cm$^{-2}$. As $n$ increases, so do $G_{\text{coh}}^\text{ph} + G_{\text{el}}$ and $G_{\text{diff}}^\text{ph} + G_{\text{el}}$ as indicated by the black arrow. The corresponding experimental TBC data from Yalon et al. are indicated by red diamond symbols with error bars. The $G_{\text{coh}}^\text{ph} + G_{\text{el}}$ ($n = 3 \times 10^{11}$ cm$^{-2}$) and $G_{\text{diff}}^\text{ph} + G_{\text{el}}$ ($n = 0$ cm$^{-2}$) curves that best fit the experimental data are indicated by the short blue arrows.

MD simulation results [6, 23, 38] which have no electronic contribution.

The substantial spread in the $G_{\text{diff}}^\text{ph} + G_{\text{el}}$ predictions in the $n = 0$–$10^{12}$ cm$^{-2}$ range may also explain the variation in the experimental TBC values for the MoS$_2$-SiO$_2$ interface. It has been reported that the intrinsic doping of single-layer MoS$_2$ can reach up to $\sim 10^{13}$ cm$^{-2}$ on SiO$_2$ substrates [39, 40], although not all the electrons are delocalized [41]. In samples grown by chemical vapor deposition, a maximum electron density of $n \approx 10^{13}$ cm$^{-2}$ can be reached using an applied gate voltage [42]. Nonetheless, we limit our analysis in the following discussion to a conservative $n \leq 10^{12}$ cm$^{-2}$ range as there has been no systematic attempt to measure how the TBC varies with $n$. As $n$ increases from 0 to $10^{12}$ cm$^{-2}$, the theoretical $G_{\text{diff}}^\text{ph} + G_{\text{el}}$ TBC at 300 K increases from 13.5 to 32.1 MW K$^{-1}$ m$^{-2}$, producing a spread comparable to the TBC variation ($G = 20.3$–$33.5$ MW K$^{-1}$ m$^{-2}$) obtained at 295 K in [23]. This suggests that the spread in TBC values may be caused by the variability in the electron density due to intrinsic doping in MoS$_2$ [43] which is often $n$-doped as a result of sulfur atom vacancies [41]. It also implies that it should be possible to increase the TBC of the MoS$_2$-SiO$_2$ interface by doping the MoS$_2$ through the introduction of dopants [44, 45] or the use of a metal gate electrode (as in a field-effect transistor) to modulate the electron density. In addition to the change in the TBC as $n$ increases, another striking feature is the change in the temperature dependence of the TBC.

3.3. Phononic and electronic TBC for WS$_2$-SiO$_2$ interface

We also present the simulated electronic and phononic TBC for the WS$_2$-SiO$_2$ interface in figure 5 in which we plot $G_{\text{coh}}^\text{ph}$, $G_{\text{coh}}^\text{ph} + G_{\text{el}}$ (coherent phononic and electronic) and $G_{\text{diff}}^\text{ph} + G_{\text{el}}$ (diffuse phononic and electronic) at different values of the electron density from $n = 10^{11}$ to $10^{12}$ cm$^{-2}$ in steps of $\Delta n = 10^{11}$ cm$^{-2}$ as a function of temperature. Unlike figure 4, no comparison with experimental data is made because no such data is available for the WS$_2$-SiO$_2$ interface. Nevertheless, the data in figure 5 can be useful for comparison with future experimental TBC measurements of the WS$_2$-SiO$_2$ interface and understanding its TBC variability. We observe similar trends in figure 5 to the data in figure 4. At 300 K, as $n$ increases from 0 to $10^{12}$ cm$^{-2}$, $G_{\text{coh}}^\text{ph} + G_{\text{el}}$ increases from 3.0 to 18.6 MW K$^{-1}$ m$^{-2}$ while $G_{\text{diff}}^\text{ph} + G_{\text{el}}$ increases from 12.4 to 28.1 MW K$^{-1}$ m$^{-2}$. We note that the $G_{\text{coh}}^\text{ph}$, $G_{\text{diff}}^\text{ph}$ and $G_{\text{el}}$ data for the WS$_2$-SiO$_2$ interface are comparable but smaller than their corresponding values for the MoS$_2$-SiO$_2$ interface in figure 4. The large change in the $G_{\text{coh}}^\text{ph} + G_{\text{el}}$ and $G_{\text{diff}}^\text{ph} + G_{\text{el}}$ data with respect to $n$ means that a change in electron density, from either intrinsic doping or an applied gate voltage, can also lead to a significant detectable change in the measured TBC as with the MoS$_2$-SiO$_2$ interface.

4. Summary and conclusion

In this work, we analyze the theoretical phononic and electronic TBC of the WS$_2$-SiO$_2$ and MoS$_2$-SiO$_2$ interface. To describe the phononic TBC contribution for the disordered TMD-substrate interface, we introduce a diffuse phonon model. We compare the coherent and diffuse phonon models for the WS$_2$-SiO$_2$ and MoS$_2$-SiO$_2$ interface, and find that the diffuse phonon model yields significantly higher TBC values that fit the TBC data for the MoS$_2$-SiO$_2$ interface from MD simulations and thermometric experiments better, implying that the TMD-substrate interfaces are disordered. Our analysis of the experimental TBC data from [21] for the MoS$_2$-SiO$_2$ interface indicates that the overall TBC is
dominated by diffuse phonon transport although the electronic contribution from RP scattering can be significant even at low electron densities and becomes comparable to the phononic contribution at higher electron densities. The simulated phononic and electronic TBC data for the WS$_2$-SiO$_2$ interface also indicate that the electronic contribution to its TBC is also significant. Our results show that the spread in experimental TBC values of the MoS$_2$-SiO$_2$ interface can possibly be explained by the variability in intrinsic doping which affects the electronic TBC.

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