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# Power Dissipation of WSe<sub>2</sub> Field-Effect Transistors Probed by Low-Frequency Raman Thermometry

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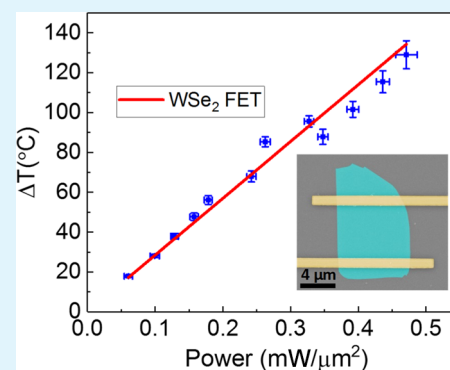
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## Supporting Information

**ABSTRACT:** The ongoing shrinkage in the size of two-dimensional (2D) electronic circuitry results in high power densities during device operation, which could cause a significant temperature rise within 2D channels. One challenge in Raman thermometry of 2D materials is that the commonly used high-frequency modes do not precisely represent the temperature rise in some 2D materials because of peak broadening and intensity weakening at elevated temperatures. In this work, we show that a low-frequency E<sub>2g</sub> shear mode can be used to accurately extract temperature and measure thermal boundary conductance (TBC) in back-gated tungsten diselenide (WSe<sub>2</sub>) field-effect transistors, whereas the high-frequency peaks (E<sub>2g</sub><sup>1</sup> and A<sub>1g</sub>) fail to provide reliable thermal information. Our calculations indicate that the broadening of high-frequency Raman-active modes is primarily driven by anharmonic decay into pairs of longitudinal acoustic phonons, resulting in a weak coupling with out-of-plane flexural acoustic phonons that are responsible for the heat transfer to the substrate. We found that the TBC at the interface of WSe<sub>2</sub> and Si/SiO<sub>2</sub> substrate is ~16 MW/m<sup>2</sup> K, depends on the number of WSe<sub>2</sub> layers, and peaks for 3–4 layer stacks. Furthermore, the TBC to the substrate is the highest from the layers closest to it, with each additional layer adding thermal resistance. We conclude that the location where heat dissipated in a multilayer stack is as important to device reliability as the total TBC.

**KEYWORDS:** WSe<sub>2</sub>, power dissipation, Raman thermometry, thermal boundary conductance, 2D materials, thermal transport, Boltzmann transport



## INTRODUCTION

The demand for further miniaturization of nanoelectronics<sup>1–7</sup> with high-density vertical integration provides an excellent opportunity for two-dimensional (2D) materials to rival conventional silicon technology.<sup>8,9</sup> Successful implementation of these thin materials within the architecture of future devices could enable the fabrication of field-effect transistors (FETs) with sub-nanometer channel thickness and minimized short-channel effects.<sup>10–13</sup> However, weak energy transport through the van der Waals (vdW) interface of 2D materials with their three-dimensional (3D) underlying substrates could impose a serious challenge for the practical application of these materials because of thermal management issues.<sup>14–16</sup> To shed light on this problem, we performed Raman thermometry experiments and first-principles calculations to characterize the thermal boundary conductance (TBC) at the interface of tungsten diselenide (WSe<sub>2</sub>) and a Si/SiO<sub>2</sub> substrate, within an FET platform, at different applied powers. WSe<sub>2</sub> is selected because of its semiconducting behavior with a high charge carrier mobility (e.g., extrinsic back-gate electron mobility of 142 cm<sup>2</sup> V<sup>–1</sup> s<sup>–1</sup> for the monolayer).<sup>17,18</sup> However, its low in-plane

thermal conductivity (3.935 W m<sup>–1</sup> K<sup>–1</sup> for 1 μm WSe<sub>2</sub> sample size<sup>19</sup>), coupled with the small cross-sectional area, makes the role of TBC more critical for heat dissipation during device operation. Furthermore, the lowest thermal conductivity of any solid was measured in the cross-plane direction of disordered WSe<sub>2</sub>.<sup>20</sup> In this study, we employed a low-frequency shear-mode peak of WSe<sub>2</sub> Raman spectra to measure the TBC of back-gated WSe<sub>2</sub> transistors.

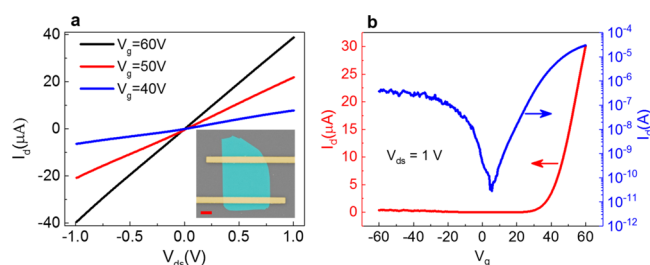
## RESULTS AND DISCUSSION

WSe<sub>2</sub> flakes were exfoliated from bulk powder (Alfa Aesar Company) by standard mechanical exfoliation onto the silicon substrate with a 300 nm oxide layer. Source–drain metal electrodes were patterned by e-beam lithography, followed by the deposition of 5 nm silver (Ag) and 35 nm gold (Au). The inset in Figure 1a shows the colored scanning electron microscopy (SEM) image of a typical fabricated device with

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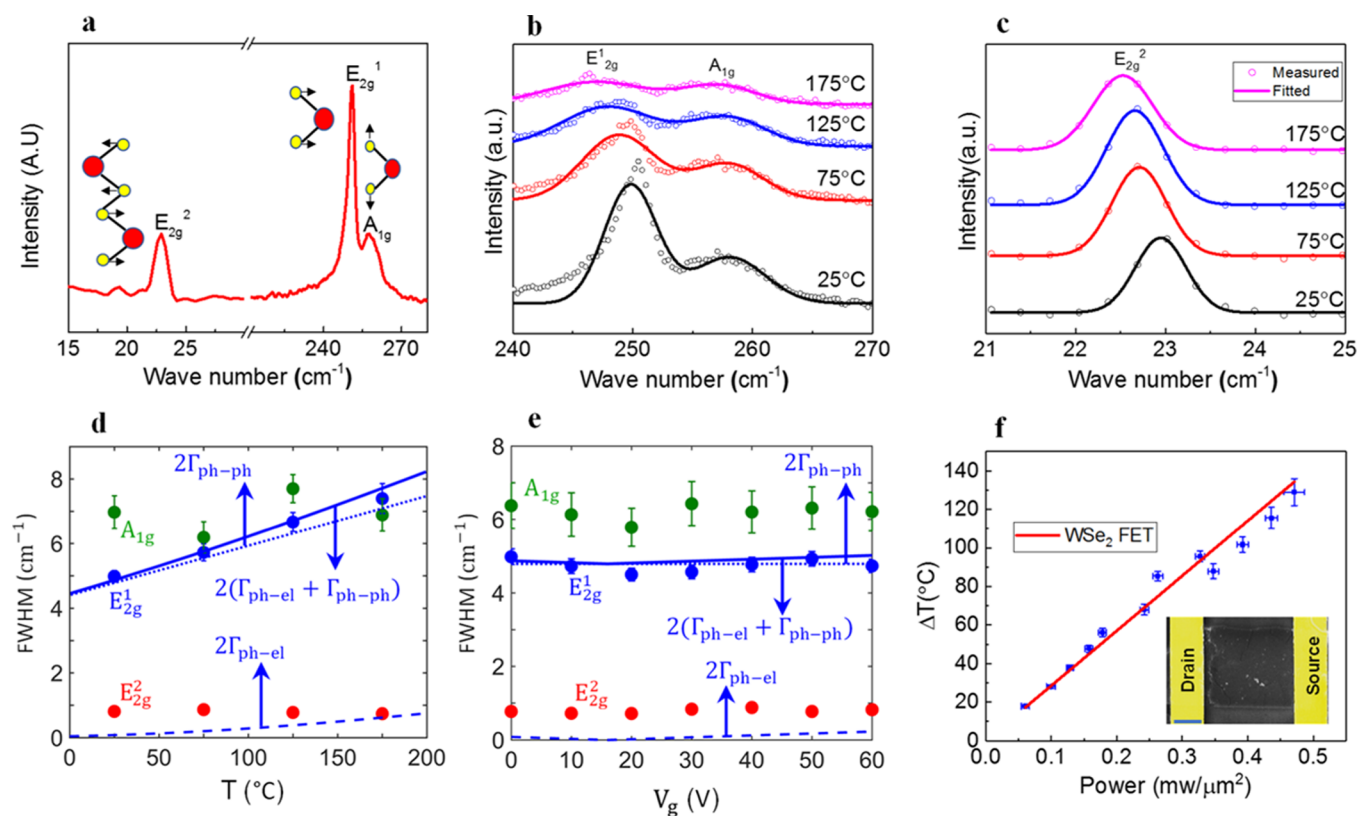
**Figure 1.** (a) Electrical transport characteristic of WSe<sub>2</sub> FET. The inset shows the colorized SEM image of the back-gated WSe<sub>2</sub> FET (scale bar is 2  $\mu\text{m}$ ). (b) Electrical transfer characteristic of WSe<sub>2</sub> FET in the vacuum environment.

a length and a width of  $\sim 6.5$  and  $\sim 8.5$   $\mu\text{m}$ , respectively. The  $I_d$ – $V_{ds}$  measurements in the vacuum environment at three different applied gate biases (Figure 1a) show linear behavior, implying an ohmic contact between Ag electrodes and WSe<sub>2</sub>, which is consistent with a previous study.<sup>18</sup>

Figure 1b shows the electrical transfer characteristics ( $I_d$ – $V_g$ ) of the device measured by applying 1 V source–drain bias and sweeping the gate voltage from  $-60$  to  $+60$  V in vacuum. The measurements indicate an ambipolar behavior with an extrinsic back-gate electron mobility of  $\sim 65$   $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$  and a small hole current at high negative gate voltages (2 orders of magnitude lower than that of the electrons).<sup>18</sup> We have also investigated the effect of current annealing on the mobility of our devices by applying a large (e.g., 10 V) drain–source bias.

After this process, the mobility of some of the devices has improved by up to a factor of 2 (Figure S1). The range of back-gated extrinsic electron mobility values for six different fabricated devices varies from  $\sim 20$  to  $\sim 70$   $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$  with  $I_{\text{ON}}/I_{\text{OFF}}$  ratio of higher than  $10^6$ . Figure S2 shows the atomic force microscopy (AFM) image of the WSe<sub>2</sub> flake for the device shown in Figure 1a. The height profile shows a thickness of  $\sim 7$  nm for this flake.

Because Raman thermometry measurements are taken in the air environment, we tested the  $I_d$ – $V_g$  experiments of WSe<sub>2</sub> FET under ambient conditions. However, the results (Figure S3, black) show that the measured current reduces by about 2 orders of magnitude when the device is exposed to air. This could be due to the absorption of moisture from the air, which increases the induced charge traps.<sup>21,22</sup> To passivate the WSe<sub>2</sub> channel, we deposited a 20 nm AlO<sub>x</sub> layer directly on top of the flakes using atomic layer deposition (ALD). Figure S4 shows AFM images of the AlO<sub>x</sub> layer deposited at 150 and 200  $^\circ\text{C}$  on top of the WSe<sub>2</sub> flakes. The height profiles from the surface of these flakes show nonuniform deposition of the AlO<sub>x</sub> layer with many pinholes, which caused degradation in the device performance under the ambient condition. Increasing the temperature of the growth from 150 to 200  $^\circ\text{C}$  did not improve the quality of the deposition layer but resulted in island-by-island deposition of this layer on top of the WSe<sub>2</sub> flake. We found that the deposition of 1 nm SiO<sub>x</sub> layer as a seeding layer by e-beam evaporation followed by the deposition of 20 nm AlO<sub>x</sub> layer in the ALD system at 150



**Figure 2.** (a) Raman point spectra from the WSe<sub>2</sub> flake. (b)  $E_{2g}^1$  and  $A_{1g}$  Raman peaks of WSe<sub>2</sub> at different temperatures. (c)  $E_{2g}^2$  Raman peak of WSe<sub>2</sub> at different temperatures. (d) Measured (symbols) and calculated (lines) fwhm vs temperature at zero gate voltage. (e) Experimentally measured fwhm vs gate voltage for  $A_{1g}$ ,  $E_{2g}^1$ , and  $E_{2g}^2$  modes at room temperature (symbols) and calculated fwhm due to phonon–electron and phonon–phonon interactions (lines). (f) Temperature rise vs applied powers for the channel of the WSe<sub>2</sub> FET. The inset shows the false-colored SEM micrograph of the device after the breakdown test (scale bar is 1  $\mu\text{m}$ ).

°C is needed to form a uniform layer of  $\text{AlO}_x$ . The AFM image (Figure S5) from the surface of the  $\text{WSe}_2$  flake after the passivation of the  $\text{AlO}_x$  layer by this method shows an average roughness of  $\sim 1.5$  nm. To establish electrical connections to the device, the  $\text{AlO}_x$  layer was etched away from the gold pad areas by using a chlorine etch system (see the Methods section for more details).

Transfer characteristic measurements were performed under the ambient condition to test the stability and performance of devices. Results of the  $I_d$ – $V_g$  measurement are shown in Figure S3 (green) at  $V_{ds} = 1$  V. We did not observe a noticeable change in the mobility of the passivated device measured under ambient conditions compared with the non-passivated device tested in a vacuum environment (Figure 1b). However, the maximum current of the device at  $V_g = 60$  V was decreased by a factor of  $\sim 2$  compared to the maximum current seen in the vacuum environment, but the device operation remained steady for the entire duration of the subsequent tests. We note that the  $\text{AlO}_x$  passivation results in 14-fold improvement in mobility compared with the bare devices in air.

Next, confocal Raman spectroscopy at low laser powers ( $< 7.5 \mu\text{W } \mu\text{m}^{-2}$ , laser spot diameter  $\approx 1.3 \mu\text{m}$ ) was utilized to obtain the Raman spectra of the  $\text{WSe}_2$  flakes under the thermal equilibrium condition. Figure 2a exhibits the characteristic Raman peaks of the  $\text{WSe}_2$  flake acquired from the fabricated  $\text{WSe}_2$  FET. The  $E_{2g}^1$ ,  $A_{1g}$ , and  $E_{2g}^2$  peaks correspond to the in-plane, out-of-plane, and interlayer vibration of atoms, respectively.<sup>23</sup> It is worth noting that Horiba's ULF Bragg filter was used to detect the  $E_{2g}^2$  low-frequency Raman peak. For Raman thermometry measurements, the Raman peak shift of  $\text{WSe}_2$  FET was first calibrated versus temperature. Figure 2b,c shows the temperature dependence of  $E_{2g}^1$ ,  $A_{1g}$ , and  $E_{2g}^2$  Raman-active peaks, respectively. We fitted each individual Raman peak with a Gaussian curve to obtain the full width at half-maximum (fwhm). The extracted fwhm values of  $E_{2g}^1$ ,  $A_{1g}$ , and  $E_{2g}^2$  peaks at several temperatures are shown by blue, green, and red markers, respectively, in Figure 2d. The two high-frequency Raman-active peaks,  $E_{2g}^1$  and  $A_{1g}$ , show large fwhm compared to the  $E_{2g}^2$  peak at all temperatures. The broadening of  $E_{2g}^1$  increases significantly with temperature and, when coupled with reduced intensities at high temperatures, could result in substantial errors in the thermometry measurements. We explain this trend by simulating all of the possible anharmonic decay channels of the Raman-active high-frequency modes. We calculate the phonon dispersion using density functional perturbation theory (DFPT), described in the Methods section and elaborated in section S6, and find the energy of the Raman-active mode to be  $\hbar\omega_{\text{op}} = 30$  meV, based on Figure S6a. The decay of a high-frequency Raman-active mode into a pair of lower frequency modes requires simultaneous conservation of energy and momentum,<sup>24</sup> as discussed in detail in section S7. Our simulations show that of all the possible decay paths shown in Figure S7a, the simultaneous energy and momentum conservation conditions strongly favor the decay into two longitudinal acoustic (LA) phonons, each having one-half the energy  $\hbar\omega_{\text{LA}} = \hbar\omega_{\text{op}}/2 = 15$  meV, as the most frequent decay channel, shown in the inset of Figure S7a. The  $\text{OP} \rightarrow \text{LA} + \text{LA}$  decay is typically called the Klemens channel<sup>25</sup> (see the schematic in Figure S7b), and it leads to a fwhm proportional to  $\Gamma_0[2N_0(\omega_{\text{LA}}T) + 1]$ , where  $N_0$  is the Bose–Einstein distribution function. The resulting temperature trend of fwhm due to the anharmonic phonon–phonon interaction ( $\text{fwhm}_{\text{ph-ph}} = 2\Gamma_{\text{ph-ph}}$ ), driven by  $N_0$ , is

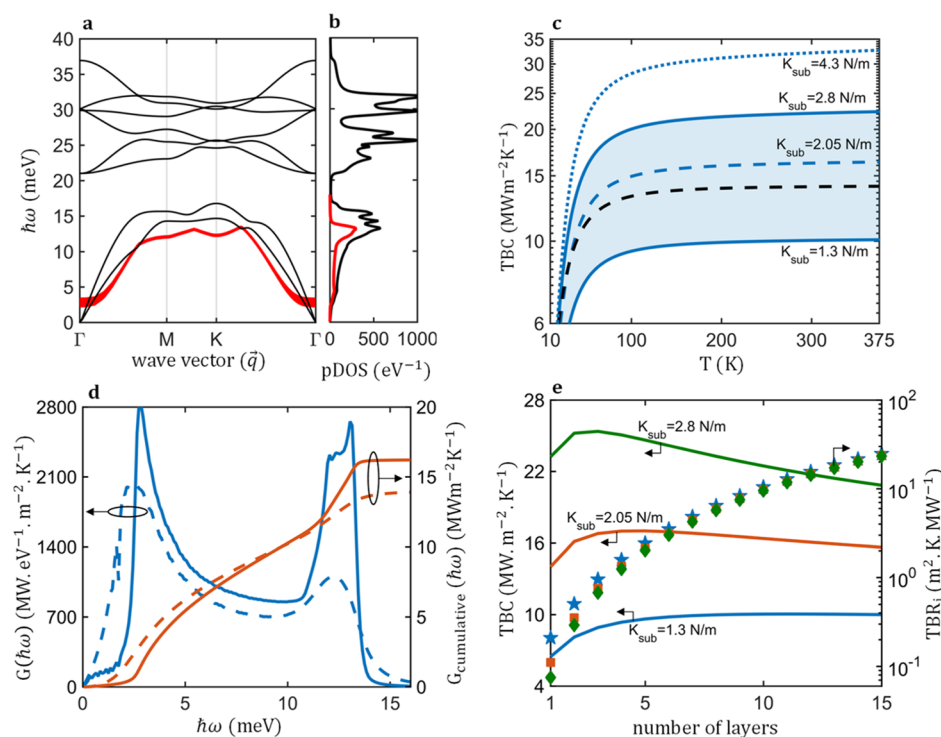
shown by the blue dotted line in Figure 2d. A  $\Gamma_0 = 0.68$  fits our experimental data for  $E_{2g}^1$  quite well. The blue solid line in Figure 2d represents the total line width obtained by summing the contribution from phonon–phonon ( $2\Gamma_{\text{ph-ph}}$  shown by the dotted blue line) and phonon–electron interactions ( $2\Gamma_{\text{ph-el}}$  shown by the dashed blue line). In contrast, because of symmetry rules,  $A_{1g}$  modes decay into one optical and one acoustic phonon, which is popularly known as the Ridley decay channel.<sup>25,26</sup> However, there exists a strong interaction between out-of-plane modes and the substrate,<sup>27</sup> which is further enhanced by encapsulation.<sup>16</sup> As a result, the width of the  $A_{1g}$  peak displays a weaker temperature dependence (see section S7 for further details).

The coupling between the low-frequency out-of-plane flexural acoustic (ZA) phonons, which are the main carrier of heat into the substrate, and high-frequency optical phonon modes is weak because their interaction is largely prohibited by reflection symmetry,<sup>28</sup> which requires an even number of flexural modes to interact. This leads to strong nonequilibrium among all phonon branches, which renders the extraction of lattice temperature from the shift in Raman frequency of optical modes,  $E_{2g}^1$  and  $A_{1g}$ , unreliable.<sup>29</sup> On the other hand, the low-frequency  $E_{2g}^2$  mode shows a sharp peak (smaller fwhm) even at high temperatures, as shown in Figure 2c. Thus, in this study, we used the frequency shift of the  $E_{2g}^2$  mode for Raman thermometry to extract TBC values. Moreover, we found that the line widths of the  $E_{2g}^1$ ,  $A_{1g}$ , and  $E_{2g}^2$  modes exhibit only a weak dependence on gate voltage (Figure 2e), suggesting that phonon–phonon interactions are the dominant contributor to Raman line widths.<sup>30</sup> We derive expressions for the line widths due to phonon–electron coupling and plot the resulting contribution to fwhm ( $=2\Gamma_{\text{ph-el}}$ ) as dashed blue lines in Figure 2d,e, showing that this contribution is relatively small for the Raman-active modes. The electron–phonon coupling rates are calculated and further discussed in the Supporting Information (section S8). The temperature dependence of  $\text{fwhm}_{\text{ph-el}}$  for different gate voltages is shown in Figure S8a,b. Besides broadening, we also observe that the difference between the frequencies of  $A_{1g}$  and  $E_{2g}^1$  peaks increases with temperature. Similar shifts in frequencies for Raman-active modes have been observed in few-layer  $\text{MoS}_2$ ,<sup>26</sup> where the  $E_{2g}^1$  mode exhibits a stronger shift than that of  $A_{1g}$ ; it has been attributed to the temperature contribution from thermal expansion and phonon anharmonicity. Previous studies<sup>31,32</sup> report this difference in frequencies to be also dependent on the number of layers (NL), and this thickness-dependent  $\omega_{A_{1g}} - \omega_{E_{2g}^1}$  has been attributed to the interlayer coupling in TMDs.

To measure the temperature rise of the  $\text{WSe}_2$  channel and silicon substrate under device operation, the Raman peak shifts were first calibrated based on the applied gate voltage of 60 V (Figure S9). The temperature rise was then calculated from the acquired Raman spectra at different applied powers (at 60 V gate voltage) and calibration curves (Figure S10a,b). Figures 2f and S11 exhibit the temperature rise versus applied powers for the  $\text{WSe}_2$  channel (based on the  $E_{2g}^2$ ) and silicon, respectively. For instance, the temperature rises of  $\sim 110$  and  $10$  °C were found for the  $\text{WSe}_2$  channel and silicon substrate, respectively, at an applied power density of  $0.4 \text{ mW}/\mu\text{m}^2$ .

To determine the TBC between the  $\text{WSe}_2$  channel and the substrate, we considered an equivalent thermal circuit from the  $\text{WSe}_2$  flake to the silicon substrate (Figure S12). All of the





**Figure 3.** (a) Full phonon dispersion of an 11-layer WSe<sub>2</sub> with 11 gapped flexural modes highlighted in red. (b) Calculated pDOS of WSe<sub>2</sub> (black line), where the pDOS of the gapped flexural branches is highlighted in red. (c) TBC as a function of temperature from 10 to 375 K for encapsulated WSe<sub>2</sub> on SiO<sub>2</sub>. The blue lines represent values for an 11-layer stack, whereas the dashed black line represents a single layer with  $K_{\text{sub}} = 2.05 \text{ N m}^{-1}$ . (d) Spectral TBC (blue) and cumulative TBC (red) as functions of phonon energy  $\hbar\omega$ . (e) TBC (left axis only) as a function of the number of layers (1 to 15) in a 15-layer stack (symbols). Colors of the symbols correspond to the colors of the solid lines for values of  $K_{\text{sub}}$ .

fabricated devices for Raman thermometry measurements had the length of  $>4 \mu\text{m}$ ; thus, the in-plane heat conduction can be neglected because of large device dimensions compared with the lateral thermal healing length of the device.<sup>11,33,34</sup> As a result, the total thermal resistance between the hot and cold zones can be considered as a sum of the thermal resistance between the WSe<sub>2</sub> flake and the SiO<sub>2</sub> substrate ( $R_{\text{int}}$ ), the thermal resistance of SiO<sub>2</sub> ( $R_{\text{SiO}_2}$ ), and the thermal resistance of silicon ( $R_{\text{Si}}$ ). The resistance between Si and SiO<sub>2</sub> is neglected because of their low thermal boundary resistance (TBR) ( $R_{\text{oxs}} = 10^{-8} \text{ m}^2 \text{ K/W}$  near room temperature).<sup>34–36</sup> The total thermal resistance of the device ( $R_{\text{th}} = \Delta T/P$ ) was extracted from the slope of Figure 2f. Also, the thermal resistance of the silicon substrate can be calculated based on either the slope of Figure S11 or the known values for the thermal conductivity of silicon.<sup>34</sup> We found that TBC values for six tested devices with the same thicknesses ( $\sim 7 \text{ nm}$ ) vary in the range of 10–32  $\text{MW/m}^2 \text{ K}$  (Table S1, section S13) using  $\text{TBC} = 1/(A \cdot R_{\text{int}})$ , where  $A$  is the area of the device and  $R_{\text{int}}$  is the thermal resistance between the WSe<sub>2</sub> channel and the SiO<sub>2</sub>/Si substrate. Moreover, we investigated the electrical breakdown characteristic of WSe<sub>2</sub> devices. Figure S13 shows a typical  $I_d$ – $V_{\text{ds}}$  result before and after the breakdown test. The maximum current of  $49 \mu\text{A}/\mu\text{m}$  was recorded at  $V_{\text{ds}} = 80 \text{ V}$  right before the breakdown point. This provides a power density of  $1.184 \text{ mW}/\mu\text{m}^2$  for most of the tested devices, implying an average temperature of  $\sim 370^\circ\text{C}$  for the WSe<sub>2</sub> channel at the breakdown point. The SEM (inset of Figure 2f) and AFM (inset of Figure S13) images of the WSe<sub>2</sub> FET after

the breakdown test reveal that the failure happened near the drain electrode.

To model the transfer of phonons from the encapsulated few-layer WSe<sub>2</sub> stack to its underlying SiO<sub>2</sub> substrate, we used a combination of density functional theory (DFT) and phonon Boltzmann transport equation. We first calculated the full phonon dispersion of monolayer (1H phase) WSe<sub>2</sub> from first principles. We then modified the dispersion of the flexural ZA branch to include gapping because of the coupling to the substrate and branching due to interlayer coupling, as described in the Methods section. In vdW monolayers, the ZA phonon branch is the main contributor to thermal heat transfer into the substrate.<sup>27</sup> In multilayers, each of the  $N$  layers in the vdW stack contributes one distinct ZA branch and thus adds to the total TBC. We modified our previously developed 1-layer 2D–3D TBC<sup>16</sup> for  $N$ -layer vdW stacks, following the formulation by Ong on multilayer TBR.<sup>37</sup> The multilayer TBC model, including the effect of encapsulation by the  $\text{AlO}_x$  layer, is further described in the Supporting Information (section S15).

Taking the  $N$ -layer WSe<sub>2</sub> film to be  $\sim 7 \text{ nm}$  thick (based on AFM measurements in Figure S2) and the combined thickness of the trilayer (Se–W–Se) and the vdW gap to be  $6.45 \text{ \AA}$ ,<sup>38</sup> we estimated the NL to be 11. Figure 3a shows the phonon dispersion of an 11-layer WSe<sub>2</sub> stack, where we can see the collection of 11 flexural modes highlighted in red. Similarly, Figure 3b shows the phonon density of states (pDOS) of the 11-layer WSe<sub>2</sub>, where the pDOS of the ZA branches is highlighted in red. Figure 3c shows the calculated TBC as a function of temperature for an encapsulated 11-layer WSe<sub>2</sub> stack on SiO<sub>2</sub> for various values of vdW coupling constants

$K_{\text{sub}}$ . The blue shaded region spans a range of  $K_{\text{sub}} = [1.3, 2.8]$  N/m with a median value of 2.05 N/m (blue dashed line) corresponding to TBC values of  $[10, 22]$  MW/m<sup>2</sup> K and 16.15 MW/m<sup>2</sup> K, respectively. Increasing the coupling to  $K_{\text{sub}} = 4.3$  N/m recovers the highest measured value of 32 MW/m<sup>2</sup> K. The black dashed line corresponds to the TBC of a 1-layer WSe<sub>2</sub> on SiO<sub>2</sub>, where we see a smaller TBC as a result of only one layer contributing to the overall TBC, whereas each of the layers in the 11-layer sample contributes to the total TBC.

Figure 3d shows the room-temperature spectral TBC (blue line) and the cumulative TBC (red line) as functions of phonon energy. The solid lines refer to an 11-layer stack, whereas the dashed lines represent a 1-layer stack. In both cases, we see two distinct peaks in the spectral TBC, a larger one in the range 2.5–3 meV (22–25 cm<sup>−1</sup>), just above the gap caused by the substrate interaction and overlapping with the E<sub>2g</sub><sup>2</sup> Raman mode, and a smaller peak at around 13 meV from the zone-edge van Hove singularity of the ZA branch. In moving from 1 to 11 layers, the spectral TBC sees an upward shift in energy and increased height of the first peak because of interlayer coupling and additional ZA branches in the dispersion. As a result, we see an increase in the total TBC from 1 to 11 layers. To further probe the thickness dependence of the TBC of the encapsulated few-layer WSe<sub>2</sub> on SiO<sub>2</sub>, in Figure 3e we plot the TBC (left axis) as a function of NL in the vdW stack for several values of substrate coupling constants. For weak substrate coupling and small values of  $K_{\text{sub}}$  (i.e.,  $K_{\text{sub}} = 1.3$  N/m), the TBC increases monotonically with the number of the layers, the same trend as previously calculated for bare (unencapsulated) few-layer graphene<sup>37</sup> and measured in few-layer MoS<sub>2</sub>, both on SiO<sub>2</sub>.<sup>39</sup>

In contrast, as  $K_{\text{sub}}$  increases in the encapsulated multilayer WSe<sub>2</sub>, we observe a peak in the thickness dependence of TBC in Figure 3e. For  $K_{\text{sub}} = 2.05$  and 2.8 N/m, the TBC peaks at 4 layers and 3 layers, respectively, and then slowly declines with additional layers. This trend is different from the previous work and caused by encapsulation, which improves TBC in monolayers<sup>40</sup> by increasing damping and enhancing the transmission of low-frequency flexural phonons to the substrate. In a multilayer, the layer closest to the substrate contributes most to the TBC but benefits less from encapsulation when there are many layers added on top of it. To elucidate this point, we plot individual contributions from each layer in a 15-layer WSe<sub>2</sub> on the right-hand side  $y$ -axis of Figure 3e. The per-layer TBR ( $\text{TBR}_j = \text{TBC}_j^{-1}$ ) is indicated by symbols (star, square, and diamond), whereas the  $x$ -axis stands for the  $j$ th layer in a 15-layer stack. The color of the symbols corresponds to the colors of the lines for the three values of  $K_{\text{sub}}$ . Most of the TBC comes from the first three layers closest to the substrate where the TBR is the lowest ( $\text{TBR}_{1-3} < 1 \text{ m}^2 \text{ K/MW}$ , depending on  $K_{\text{sub}}$ ), whereas the TBR increases steadily for layers away from the substrate, indicating an additional interlayer TBR. After 8 of the 15 layers, the TBR <sub>$j$</sub>  of each layer converges to the same value because the resistance between consecutive layers is dominated by the interlayer coupling  $K_z$  rather than  $K_{\text{sub}}$ . Hence, stronger substrate adhesion favors few-layer WSe<sub>2</sub> because most of the heat transfer is contributed by the first few layers closest to the substrate, whereas in the case of weak substrate coupling, a multilayer stack or bulk WSe<sub>2</sub> would result in largest total TBC. In all cases, the location where heat is dissipated in the vertical stack will impact its removal via substrate. Because of the large Thomas–Fermi screening length ( $\lambda_{\text{TF}} \approx 7 \text{ nm}$ ) and

interlayer resistance in TMDs, the location of Joule heating in the multilayer stack varies with the gate voltage.<sup>41</sup> The thickness of our WSe<sub>2</sub> flakes is 7 nm, comparable to  $\lambda_{\text{TF}}$ , so we do not expect large variations of dissipation in the vertical direction in our measurements. The impact on heat removal would be more pronounced in samples thicker than  $\lambda_{\text{TF}}$  and at larger carrier concentrations.

## CONCLUSIONS

In summary, we investigated the power dissipation of WSe<sub>2</sub> FETs under electrical operation using the low-frequency Raman mode. We found that the typical E<sub>2g</sub><sup>1</sup> and A<sub>1g</sub> peaks do not provide accurate temperature information because of peak broadening and intensity weakening at elevated temperatures. Our experimental and DFT results revealed that the low-frequency E<sub>2g</sub><sup>2</sup> shear mode, which overlaps with the ZA modes, can be used to measure the temperature rise of the WSe<sub>2</sub> channel and determine TBC values. We also showed that the TBC at the interface of WSe<sub>2</sub> and Si/SiO<sub>2</sub> varies with the number of layers. The conductance is highest from the layers closest to the substrate, whereas those away from it suffer from additional interlayer resistance. We conclude that the location where heat is dissipated matters as much as the overall TBC.

## METHODS

**AlO<sub>x</sub> Deposition.** The Savannah-ALD system was used for this process. The WSe<sub>2</sub> samples were transferred to the ALD chamber directly after exfoliation and annealing at 200 °C in the presence of the argon gas. The system pumped down immediately to remove the unwanted gases from the deposition chamber. Then, 20 nm Al<sub>2</sub>O<sub>3</sub> layer was deposited on top of the flakes by using water as an oxygen source and trimethylaluminum (TMA) as an aluminum source. During the process, TMA was purged into the chamber by a pulse time of 0.015 s, and after 8 s waiting time, the water was also purged into the chamber by a pulse time of 0.018 s. During the process, 5 standard cubic centimeters per minute (sccm) nitrogen gas was purged into the chamber as a carrier and venting gas.

**AlO<sub>x</sub> Etch Process.** To establish an electrical connection to devices after the deposition of the Al<sub>2</sub>O<sub>3</sub> layer, we etched the layer from the gold-pad areas. The Plasma-Therm ICP chlorine etch system was used for this process. Boron trichloride (BCl<sub>3</sub>; 5 sccm) gas was used to etch the Al<sub>2</sub>O<sub>3</sub> layer, and the power of the system was set at 100 W. Argon gas (15 sccm) was also flown during the entire process, and it took about 6 min to etch the 20 nm Al<sub>2</sub>O<sub>3</sub> layer.

**Calculation of Phonon Dispersion and TBC.** We calculated the full phonon dispersion from DFPT as implemented within the Quantum ESPRESSO<sup>42</sup> package. The dispersion of the flexural branches that contribute to the TBC in the  $N$ -layer stack is gapped by coupling to the substrate and split into  $N$  subbranches by interlayer coupling. The modification of the flexural ZA phonon branch dispersion due to these interactions can be written as

$$\tilde{\omega}_{j,\text{ZA}}(\vec{q}) = \sqrt{\omega_{\text{ZA}}^2(\vec{q}) + \omega_0^2 + \frac{2K_z}{M_{\text{WSe}_2}} \sin^2\left[\frac{(j-1)\pi}{2NL}\right]}, \text{ where } \omega_{\text{ZA}}(\vec{q}) \text{ is}$$

the as-calculated DFPT phonon dispersion of the ZA branch of WSe<sub>2</sub>,  $\omega_0$  is the gapping of the first monolayer in the stack  $\omega_0 = \sqrt{K_{\text{sub}}/M_{\text{Se}}}$ ,  $K_{\text{sub}}$  is the vdW spring coupling constant between the bottommost WSe<sub>2</sub> layer and the substrate,  $K_z$  is the interlayer spring coupling constant between adjacent layers of WSe<sub>2</sub>,<sup>23</sup> taken as  $K_z = 8.24 \text{ N m}^{-1}$ ,  $M_{\text{Se}}$  is the atomic mass of Se,  $j$  represents the  $j$ th layer in the  $N$ -layer vdW stack spanning  $[1, NL]$ , and  $NL$  is the total  $NL$ . The dispersion of the gapped ZA branches is next input into our multilayer TBC model to obtain phonon transport from an encapsulated few-layer WSe<sub>2</sub> flake into its underlying SiO<sub>2</sub> substrate. Further details of the phonon dispersion calculation and the multilayer TBC model are described in the Supporting Information (sections S6 and S15,

respectively). Figure S6 shows the full phonon dispersion and pDOS for the monolayer WSe<sub>2</sub> as calculated from the first principles.

## ■ ASSOCIATED CONTENT

### Supporting Information

The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.8b04724.

Mobility improvement of the devices after current annealing; AFM characterizations of WSe<sub>2</sub> flakes; electrostatic breakdown test; detailed information about first-principles calculation of phonon dispersion of the monolayer WSe<sub>2</sub>; derivation of phonon–electron rates; and details about phonon–phonon coupling, optical decay channel, and our multilayer TBC model (PDF)

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### Author Contributions

A.S.K. and A.B. conceived the idea. A.S.K. led the fabrication and experiments. Z.A. developed the BTE model and led the first-principles calculations. A.B. developed the fabrication process, experimental setup, and thermal analyses. A.B. and Z.H. fabricated all of the devices. A.B., P.Y., and Z.H. performed Raman thermometry measurements and thermal analysis. C.J.F. performed numerical calculations, and A.K.M. derived phonon interaction rates. All authors contributed to the write-up of the article.

### Notes

The authors declare no competing financial interest.

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