Ambipolar Thickness-Dependent Thermoelectric Measurements of WSe₂

Victoria Chen, Hye Ryoung Lee, Çağıl Koroğlu, Connor J. McClellan, Alwin Daus, and Eric Pop*

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ABSTRACT: Thermoelectric materials can harvest electrical energy from temperature gradients, and could play a role as power supplies for sensors and other devices. Here, we characterize fundamental in-plane electrical and thermoelectric properties of layered WSe₂ over a range of thicknesses, from 10 to 96 nm, between 300 and 400 K. The devices are electrostatically gated with an ion gel, enabling us to probe both electron and hole regimes over a large range of carrier densities. We extract the highest n- and p-type Seebeck coefficients for thin-film WSe₂, −500 and 950 μV/K respectively, reported to date at room temperature. We also emphasize the importance of low substrate thermal conductivity on such lateral thermoelectric measurements, improving this platform for future studies on other nanomaterials.

KEYWORDS: Seebeck coefficient, power factor, thin-film semiconductors, 2D materials

Over half of the energy generated by humanity is ultimately dissipated without being utilized, and the majority of that rejected energy is lost in the form of waste heat. In this context, thermoelectric energy harvesting could play an important role, by converting spatial temperature gradients into an electrical voltage. Most existing commercial thermoelectric devices are primarily based on Bi₂Te₃ and its related compounds; however, many of these materials are relatively inefficient and expensive.

The efficiency of a thermoelectric material is quantified by its figure of merit, $zT$, which is defined as $zT = S^2\sigma T/\kappa$, where $S$ is the Seebeck coefficient, $\sigma$ is the electrical conductivity, $T$ is the absolute temperature, and $\kappa$ is the thermal conductivity. High $zT$ can be achieved by maximizing the power factor $PF = S^2\sigma$ and minimizing $\kappa$. This can be accomplished by tuning the material’s (electron or hole) carrier concentration, but because the constituent parameters of $zT$ are interconnected, this can be a difficult process. For example, $S$ is often inversely proportional to the material’s carrier density while $\sigma$ is directly proportional. Therefore, increasing the carrier density can cause varying effects on $zT$, depending on the regime.

Another method to achieve more efficient thermoelectrics is to consider novel material systems. For example, low-dimensional materials with quantum confinement effects are uniquely suited to have efficient thermoelectric properties. Due to sharp features in their density of states, certain semiconducting two-dimensional (2D) materials could achieve large Seebeck coefficients, boosting their PF. Furthermore, the dominant presence of interfaces and mass differences between constituent atoms in some 2D materials can lead to relatively low $\kappa$. Although some theory and experiments have been done on 2D material thermoelectric properties, additional in-depth studies are required before these can be considered a viable commercial technology.

In this work, we measure in-plane, thickness-dependent thermoelectric properties of WSe₂ probing both n- and p-type regimes. Among 2D materials, WSe₂ is relatively unique, as it is one of the few options that is readily ambipolar, either through gating or doping, which has also made it promising for nanoscale electronic applications. Using ion gel gating, we can tune the Fermi level, evaluating $S$ and $\sigma$ over a range of carrier densities, mapping the PF through its peak value, which is difficult to accomplish with traditional back-gating through a thick oxide. We extract peak room-temperature $S_p \approx 950 \pm 70$ $\mu$V/K for p-type and $S_n \approx -500 \pm 40$ $\mu$V/K for n-type, some of the highest reported values for WSe₂ to date at room temperature. This establishes WSe₂ as a promising candidate for applications such as temperature sensing, which benefit from higher induced voltages in a given temperature gradient. Such large thermopower may also play a role in WSe₂ devices such as transistors or diodes, contributing to significant heating or cooling (and therefore a direct effect on
their performance and reliability) depending on current flow direction.\textsuperscript{19–21} For instance, due to the large Seebeck coefficient of WSe\textsubscript{2}, one contact of a transistor may experience non-negligible thermoelectric heating while the other experiences cooling.\textsuperscript{19}

In order to accurately measure the Seebeck coefficient of our WSe\textsubscript{2} films, it is important to sustain a constant, known temperature difference across the material. To accomplish this, we use a measurement structure whereby the WSe\textsubscript{2} samples are placed on a glass substrate, which has a low, uniform \( \kappa \) unlike the commonly used SiO\textsubscript{2}/Si substrates.\textsuperscript{22} The uniform \( \kappa \) of the substrate limits vertical heat flow, resulting in a longer and more gradual lateral temperature gradient, which enables a greater temperature difference to be sustained across the WSe\textsubscript{2} sample. This effect is quantified in Supporting Information section S1, with finite-element method simulations showing an \( \sim10\times \) greater temperature difference (\( \Delta T \)) across the sample when using a glass substrate. The larger \( \Delta T \), in turn, leads to a larger measurable Seebeck voltage, increasing the signal-to-noise ratio and reducing measurement error. In addition, the lower \( \kappa \) of the glass substrate reduces the lateral temperature drops within the thermometer lines, which, especially coupled with wide thermometer lines, can be a substantial source of error in the extracted Seebeck coefficient. Crucially, non-negligible thermometer line widths relative to the thermometer spacing, if not accounted for, can lead to a sizable underestimation of the true Seebeck coefficient, as we show in Supporting Information section S2. The Seebeck coefficients reported in this work have been corrected by the underestimation factors listed in Table S1, which were computed using finite-element method simulations.

The WSe\textsubscript{2} samples are exfoliated from a bulk crystal using the “tape method” directly onto the glass substrates, and subsequently contacted with 40 nm of Pd deposited by electron-beam evaporation. An additional Pd line running parallel to WSe\textsubscript{2} contacts serves as the heater, as shown in Figure 1. While not in direct contact with the WSe\textsubscript{2} channel, this heater line is closely positioned and Joule-heated to create the lateral temperature gradient. All metal lines are \( \sim400\ \mu \text{m} \) long, significantly longer than the width of the WSe\textsubscript{2} sample, to ensure a one-dimensional temperature gradient along its length rather than any temperature gradients along the width, which would confound the measurement.\textsuperscript{23} The Pd lines in contact with the WSe\textsubscript{2} serve as the source and drain for electrical transport measurements, as well as the resistive thermometers. These metal lines (except for the channel area) are then capped with 100 nm evaporated SiO\textsubscript{2} for electrical isolation. The ionic liquid EMIM-TFSI (1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)imide), is mixed with PS–PEO–PS triblock copolymer and dissolved in acetonitrile before being deposited on the WSe\textsubscript{2} channel to form an ion gel for more structural stability. The sample is then cured in air at 80 °C for 10 min. The ion gel contacts the WSe\textsubscript{2} channel as well as the gate pad, such that an applied voltage on the gate pad leads to an electric field over the channel.

We first characterize the electrical properties of the WSe\textsubscript{2} devices. Representative transfer (\( I_D \) vs \( V_{GS} \)) and output (\( I_D \) vs \( V_{DS} \)) characteristics are shown for a 96 nm thick sample in Figure 2a and 2b, respectively. By using an ion gel for gating, we are able to sweep over a large range of carrier densities with a relatively small gate voltage range. This device shows nearly “perfect” ambipolar behavior [i.e., it conducts both with electrons (for positive \( V_{GS} \)) and holes (for negative \( V_{GS} \)] and does so with nearly the same maximum current for both electron and hole branches in Figure 2a. The maximum-to-minimum current ratio is \( I_{\text{max}}/I_{\text{min}} \approx 10^8 \), where \( I_{\text{min}} \) is limited by gate leakage and \( I_{\text{max}} \) reaches \( \sim3\ \mu \text{A}/\mu \text{m} \) at \( V_{DS}=100 \text{ mV} \).

Using these test structures and measurements, we investigate six different WSe\textsubscript{2} sample thicknesses, between 10 and 96 nm. Figure 3a displays the sheet conductance at each gate bias, \( G_{sh} = (I_D/V_{DS})(L/W) \), where \( W \) and \( L \) are the width and length of the sample, respectively, for the range of WSe\textsubscript{2} sample thicknesses investigated. The intrinsic voltage \( V_{DS} = V_{DS} - I_DR_{sc} \) subtracts out the series resistance \( R_{sc} \) caused by the long and thin Pd lines, which becomes a significant component.

![Figure 1](https://example.com/f1.png)

**Figure 1.** (a) Schematic of the electro-thermal measurement platform, on a glass substrate (not to scale). (b) Zoomed-in optical image of a WSe\textsubscript{2} sample with Pd contacts and heater line capped with 100 nm SiO\textsubscript{2} for electrical isolation. The dashed yellow line marks the window opened into the SiO\textsubscript{2} exposing the WSe\textsubscript{2} sample to the ion gel for electrostatic gating. The two thermometer lines in contact with the WSe\textsubscript{2} sample also serve as the source and drain for further electrical characterization. \( L \) is the sample length between thermometers, and \( W \) is the average WSe\textsubscript{2} sample width.
that the measured Seebeck voltage is not due to inadvertent electrical coupling effects, we confirm that it varies quadratically with the applied heater current, as expected for a temperature gradient induced by Joule heating. Additionally, reversing the polarity of the applied heater current does not affect the measured Seebeck voltage. These verifications are discussed in Supporting Information section S5.

In Figure 3b, we plot the Seebeck coefficients for both electrons ($S_n < 0$) and holes ($S_p > 0$) over the range of our gate bias voltages. These values are measured with our electro-thermal platform, and subsequently adjusted based on finite-element method simulations to account for the nonzero heater widths in the experiment design. As expected for a semiconductor, the absolute value of the Seebeck coefficient increases with decreasing carrier density, reaching peak values of 950 $\mu$V/K for $p$-type (in the 96 nm thick sample) and $-500$ $\mu$V/K for $n$-type (in the 12 nm thick sample), at room temperature. These are the highest experimentally reported Seebeck coefficient values for thin-film WSe$_2$, exceeding those of bulk Bi$_2$Te$_3$, a commonly used commercial thermoelectric material. Care must be taken in the Seebeck voltage measurement, as the input impedance of the measurement tool must be much greater than the device resistance for an accurate reading. Therefore, no points are measured for the Seebeck coefficient when the devices are in their off-states, as indicated on Figure 3b.

While the electrical conductivity of the WSe$_2$ channel increases with increasing carrier density, the magnitude of the Seebeck coefficient follows the opposite trend. Thus, in order to gain a deeper understanding of the material’s overall thermoelectric performance, we consider the PF, defined as $S^2\sigma$. As the electrical conductivity increases and there are additional charge carriers in the channel (either $n$- or $p$-type), the PF will first increase to a maximum point, and then subsequently decrease as the Seebeck coefficient drops when the Fermi level is pushed deeper into the conduction or valence band. The PF is plotted vs electrical conductivity in Figure 4 for each sample thickness. We find a maximum PF of 1200

Figure 4. Plot of our calculated power factor (PF) vs electrical conductivity ($\sigma$) for WSe$_2$ samples of varying thicknesses. All reported Seebeck values are relative to the contact metal, Pd. To verify
Figure 5. Sheet conductance ($G_{sh}$) vs sample thickness ($t_s$) for (a) holes and (b) electrons at several $V_{GS}$ values. Power factor (PF) vs $t_s$ for (c) holes and (d) electrons at several $V_{GS}$ values, showing a general decrease of PF for increasing sample thicknesses.

μW m$^{-1}$ K$^{-2}$ for p-type conduction and 400 μWm$^{-1}$K$^{-2}$ for n-type, both in the 10 nm thick WSe$_2$ sample. These PF estimates use the entire WSe$_2$ sample thickness ($t_s$) to calculate the electrical conductivity, $\sigma = G_{sh}/t_s$; however, the entirety of the sample is not conducting charge equally—rather, current flow is confined to a channel within the top few layers gated under the ion gel. Therefore, if we considered only the thickness of the conducting channel in these calculations, the PF values would be substantially higher, comparable to the highest WSe$_2$ PFs reported to date. Supporting Information section S6 includes a PF analysis with an estimated conducting channel thickness, and Supporting Information section S8 presents a survey of WSe$_2$ Seebeck coefficients and PFs reported in existing literature.

Additionally, we consider the effect of the WSe$_2$ sample thickness on the measured electrical and Seebeck data. While the variation in $G_{sh}$ may be in part due to device-to-device variation, there is a general trend of decreasing PF with increasing thickness for a given gate voltage. The maximum PF values we extract are for the 10 nm sample, and these roughly decrease for the thicker samples. Figure 5a and b plot $G_{sh}$ vs sample thickness for holes and electrons, respectively. As expected, higher $V_{GS}$ leads to higher carrier densities and higher $G_{sh}$. Figure 5c and d display the PF vs sample thickness for electrons and holes, respectively, where we can see that the PF roughly decreases with increasing thickness. This can be partially explained by the electrical conductivity component of the PF—since $\sigma$ is normalized by thickness, the thicker samples have lower average $\sigma$ if the thickness of the conducting channel is limited to the surface layers of the sample. (Additional discussion of sample-to-sample variation is given in Supporting Information section S7.)

While quantum confinement in low-dimensional materials has long been recognized to boost the Seebeck coefficient and PF by introducing sharp features in the electronic density of states, the WSe$_2$ samples used in this work ($\geq 10$ nm thickness) are not thin enough to benefit from confinement along the cross-plane direction. However, we note that in the on-state, at the high electric fields enabled by the high-gate capacitance ion gel, the charge carriers that are responsible for conduction are in fact confined in a near-triangular field-induced potential well at the surface of WSe$_2$. The width of this potential well, and the thickness of the 2D electron gas formed therein, are on the order of nanometers. It has been experimentally shown in other materials that this field-induced quantum confinement can lead to an enhancement in Seebeck coefficient.

Therefore, thicker WSe$_2$ samples do not have inherently lower Seebeck coefficients than thinner samples in the thickness regime probed here, if they are electrostatically surface-gated to tune the carrier density. We expect that this, coupled with good material quality, is the reason that values of $S$ we extract here are higher than those reported for monolayer or few-layer WSe$_2$, and that the PFs we calculate are likewise competitive (see Supporting Information Table S5). Like these previous studies, we are also unable to determine the $\kappa$ of our substrate-supported devices, which are ill-suited for such a measurement. However, the in-plane $\kappa$ of WSe$_2$ in our thickness range is expected to be between 2 and 40 Wm$^{-1}$K$^{-1}$ near room temperature, with the larger values corresponding to thicker samples with fewer defects. More accurate $\kappa$ investigations for samples in the thickness range considered here (~10 to 96 nm) are possible on larger suspended samples and should be the focus of future work.

In conclusion, we investigated fundamental thermoelectric properties for layered WSe$_2$ sweeping through a range of carrier densities for both electrons and holes and measuring the highest reported Seebeck coefficients for this material. Our on-chip measurement platform demonstrates the advantages of utilizing a low thermal conductivity substrate to preserve a greater temperature difference along the WSe$_2$ channel, and the use of an ion gel for electrostatic gating allowed for a high degree of control over Fermi level tuning. These results, as well as the ability of WSe$_2$ to support both n- and p-type conduction, make it a promising candidate for thermoelectric applications such as temperature sensing. In addition, the large Seebeck coefficients uncovered here also mean that significant
heating or cooling may occur at WSe₂ device (e.g., transistor) contacts, depending on current flow direction.

■ ASSOCIATED CONTENT

Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acs.nanolett.2c03468.

Finite-element method simulations quantifying the substrate thermal conductivity impact on the resultant temperature gradient; finite-element method simulations to correct for Seebeck coefficient underestimation due to non-negligible thermometer widths; temperature coefficient of resistance (TCR) calibrations; discussion of additional measured data; band gap (E<sub>g</sub>) analysis; comparison of data with literature; temperature-dependent measurements. (PDF)

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Author Contributions
V.C. and E.P. conceived the idea and wrote the manuscript with inputs from all authors. V.C. and H.R.L. designed the experiments, and fabricated and measured the samples. C.K. performed the finite-element method modeling. C.J.M. and A.D. provided insights on the measurement setup and data analysis.

Notes
The authors declare no competing financial interest.

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(33) We note that impurity states localized in energy could also lead to an enhanced Seebeck coefficient; however, these defects would be accompanied by kinks in the transfer characteristics, which we do not see in our measurements.


Supporting Information

Ambipolar Thickness-Dependent Thermoelectric Measurements of WSe$_2$

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Section S1. Substrate Impact on the Temperature Difference across a Flake

To highlight the importance of the choice of substrate in the thermal design of the test structure, we have performed thermal finite-element method simulations for a cross-section of the test structure (Figure S1). For the same heater temperature, we compare the temperature differences, $\Delta T$, between the two thermometer lines for the glass substrate used in this work [Figure S1(a)] and a substrate with 300 nm-thick SiO$_2$ on Si [Figure S1(b)] similar to those used in other studies.$^1,2$ The high thermal conductivity Si substrate ($\kappa_{\text{Si}} \approx 140$ W/m/K for undoped Si near room temperature) acts like a thermal ground, causing the temperature at the top of the sample to drop close to ambient temperature ($T_0$) within a short distance (comparable to the SiO$_2$ thickness) from the heater line. This causes the temperatures on both thermometer lines, as well as their difference, to be much less than the heater temperature.

Figure S1. Temperature rise ($\Delta T$) normalized by the peak heater temperature rise ($\Delta T_H$) in the vicinity of the heater and thermometer lines for typical heater-thermometer line spacings, with (a) the glass substrate used in this work, and (b) a typical substrate with 300 nm-thick SiO$_2$ on Si ($\kappa_{\text{SiO}_2} = 1.4$ W/m/K) on Si. The temperature rise is normalized to the peak temperature rise in the heater line. The bottom of the substrate is assumed to be at ambient temperature, $T_0$. The thermometer temperature differences ($\Delta T_1 - \Delta T_2$) in (a) and (b) are 14% and 1.5% of $\Delta T_H$, respectively. The thicknesses of the metal lines have been exaggerated for this visualization (300 nm instead of the 40 nm thickness used in the experiments and simulation).
This problem can be largely avoided by using a uniform substrate such as glass, for which $\Delta T$ between the thermometer lines is close to $10\times$ that with the SiO$_2$/Si substrate. The low thermal conductivity of glass ($k_{\text{glass}} \approx 0.8 \text{ W/m/K}$) is also beneficial for preventing a large temperature drop within thermometer lines, as they are much higher in thermal conductivity than the substrate. This is important for an accurate measurement of the Seebeck coefficient: see section S2 on measurement error these temperature drops contribute.

**Section S2. Effect of Thermometer Widths on Seebeck Coefficient Measurements**

Extracting the Seebeck coefficient as the ratio of the measured thermometer potential difference to the measured temperature difference implicitly assumes that the separation of the thermometer lines is much greater than the thermometer widths (2.4 μm). However, due to the limited sizes of our exfoliated WSe$_2$ samples, the distances between the closest edges of the thermometer lines are 2.5 to 5.9 μm (see Table S3), short enough that the thermometer widths are not negligible. In this section, we quantify this error and explain how to properly estimate the Seebeck coefficients based on our measurements.

![Figure S2.](image)

Figure S2. Electrothermal simulation results for the sample with 2.5 μm channel and 18 nm thick WSe$_2$ film, showing (a) the temperature rise over ambient, and (b) the electric potential along the device cross section. The Pd line on the left is the heater and the two Pd lines on the right are the thermometers. The thicknesses of the Pd lines and WSe$_2$ are exaggerated for better visualization. A Seebeck coefficient of 200 μV/K was assumed. The thermal properties assumed are the nominal values listed in Table S2.

Figure S2 shows electrothermal simulation results for the device with the shortest channel (2.5 μm, on the 18 nm thick WSe$_2$ film), for which the measurement error is consequently highest. $\Delta T$ and $\Delta V$ refer to the temperature and potential differences between the centers of the thermometer lines, respectively, while $\Delta T'$ and $\Delta V'$ are the temperature and potential differences between the inner edges of the thermometers (2.5 μm apart for this particular device), as defined in Figure S2. The thermometer temperatures are measured via the electrical resistance, which is inversely proportional to the metal conductivity averaged within the thermometer cross section, and thus approximately proportional to the average metal resistivity. Since the resistivity changes approximately linearly with temperature rise, the electrically measured temperature corresponds to the average temperature within the thermometer. Assuming the temperature gradient within each thermometer is roughly uniform, this average is close to the temperature at the center of the thermometer. Consequently, $\Delta T$ is approximately equal to the experimentally measured temperature difference $\Delta T_{\text{avg}}$ between the thermometers, and $S = \frac{\Delta V}{\Delta T_{\text{avg}}} \approx \Delta V/\Delta T$ thus represents the measured Seebeck coefficient.

Under the thermometer lines, the effective Seebeck coefficient of the Pd/WSe$_2$ stack is
\[ S' = \frac{G_{\text{WSe}_2} S + G_m S_m}{G_{\text{WSe}_2} + G_m} = \frac{G_{\text{WSe}_2}}{G_{\text{WSe}_2} + G_m} S \ll S, \]

where \( S \) and \( S_m \) are the \( \text{WSe}_2 \) and metal Seebeck coefficients, and \( G_{\text{WSe}_2} \) and \( G_m \) are the \( \text{WSe}_2 \) and metal sheet conductances, respectively. Here, we have taken \( S_m = 0 \) since we measure the Seebeck coefficient \( S \) relative to that of the Pd thermometers, and also used the fact that \( G_m \gg G_{\text{WSe}_2} \).

The measured Seebeck voltage is

\[ \Delta V = \Delta V' + S' (\Delta T - \Delta T') \]
\[ S = \frac{\Delta V - S' (\Delta T - \Delta T')}{\Delta T'} \equiv \frac{\Delta V}{\Delta T'} \]

where we have used \( S' \ll S \). Letting \( \delta T = \Delta T - \Delta T' \), which is the sum of the temperature drops from the center of each thermometer to its inner edge,

\[ S \cong \frac{\Delta V}{\Delta T - \delta T} \equiv \frac{\Delta V}{\Delta T} \frac{1}{1 - \delta T/\Delta T} \equiv \frac{S_{\text{meas}}}{1 - \delta T/\Delta T} \]  \( \text{(S1)} \)

Equation S1 shows that the true Seebeck coefficient is greater than the measured Seebeck coefficient \( (S_{\text{meas}} = \Delta V/\Delta T_{\text{avg}}) \) by a factor of \( 1/(1 - \delta T/\Delta T) \), which depends on the temperature drops across the thermometer lines. To calculate these corrections, we have performed electrothermal finite-element method simulations for all six devices, and the results are listed in Table S1, where the uncertainties in the factor \( \delta T/\Delta T \) are due to the uncertainties in various thermal properties, the ranges of which are included in Table S2. The Seebeck coefficients we report in the manuscript are the values corrected by the factors and uncertainties listed in Table S1, after the raw Seebeck coefficients are measured as \( \Delta V/\Delta T_{\text{avg}} \). Device 3 with the shortest channel (2.5 \( \mu m \)) requires the largest correction, as expected. Thermometer line widths must be taken into account when designing such experiments and extracting results, and for our set of thermometer widths and separations, these errors range from 17 to 30%.

**Table S1.** Factors by which raw Seebeck coefficients extracted as \( \Delta V/\Delta T_{\text{avg}} \) underestimate the true Seebeck coefficient. The details of the device geometries, such as the flake thicknesses, are listed in Table S3.

<table>
<thead>
<tr>
<th>Device number</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \delta T/\Delta T ) (%)</td>
<td>17 ± 4</td>
<td>21 ± 5</td>
<td>30 ± 7</td>
<td>20 ± 5</td>
<td>26 ± 6</td>
<td>22 ± 4</td>
</tr>
</tbody>
</table>

**Table S2.** Material and interface thermal properties used in simulations. Ranges of parameters considered for uncertainty estimation are indicated in parentheses. Due to lack of data the thermal boundary resistance ranges for \( \text{WSe}_2 \) were chosen to include those measured for other interfaces of \( \text{WSe}_2 \), as well as interfaces of other transition metal dichalcogenides such as MoS\(_2\). The thermal conductivity range considered for Pd is lower than bulk values to account for the reduced thermal conductivities of thin metal films.

<table>
<thead>
<tr>
<th>Material</th>
<th>Thermal conductivity (Wm(^{-1})K(^{-1}))</th>
<th>Material interface</th>
<th>Thermal boundary resistance (Km(^2)/GW)</th>
</tr>
</thead>
<tbody>
<tr>
<td>WSe(_2)(^3,4)</td>
<td>25 (15 – 40)</td>
<td>WSe(_2) – Pd(^5,8)</td>
<td>62.5 (31 – 125)</td>
</tr>
<tr>
<td>Pd</td>
<td>50 (30 – 70)</td>
<td>WSe(_2) – glass(^5,8)</td>
<td>62.5 (31 – 125)</td>
</tr>
<tr>
<td>glass(^9)</td>
<td>0.8</td>
<td></td>
<td></td>
</tr>
<tr>
<td>SiO(_2)(^10)</td>
<td>1.4</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Finally, rather than calculating $\Delta V/\Delta T_{\text{avg}}$ directly in simulations, we may instead calculate $\delta T/\Delta T$ from simulated temperatures only (i.e. with no regard to simulated potential differences, without having to model the thermoelectric effects) to approximate the Seebeck coefficient underestimation factor, in accordance with Eq. S1. We find that doing so yields estimates that are within 1.5% of the underestimation factors given in Table S3, which confirms the validity of Eq. S1 and the approximations that lead to it. This also confirms that Seebeck coefficient underestimation arises from the non-negligible temperature drops along the widths of the thermometer lines, which in turn are caused by the fact that these widths are not negligible compared to the channel length.

**Section S3. Temperature Coefficient of Resistance (TCR) Calibration**

To quantify the temperature gradient induced by the metal heater line along the sample, we measure the four-point resistance of each Pd thermometer line at elevated temperatures by uniformly heating the substrate on a heated stage. The TCR is calculated as:

$$\alpha = \frac{R - R_0}{R_0(T - T_0)}$$

where $\alpha$ is the TCR, $R$ is the measured resistance, $R_0$ is the resistance of the line at ambient temperature, $T$ is the temperature, and $T_0$ is the ambient temperature.

Figure S3 shows plots of $R/R_0$ vs. $\Delta T = T - T_0$ for the two metal thermometer lines on each device, with dashed lines representing the linear fit from which the TCR is extracted. Although the TCR for thin Pd is well characterized in the literature, small nonidealities in the deposition and fabrication processes may cause the value to deviate slightly, and so it is still important to calibrate each line individually. The values are compiled in Table S3, and all fall within the expected range.\textsuperscript{11}

![Figure S3. TCR calibrations for each of the two Pd thermometer lines on six devices. The four-point resistance is measured for each line at room temperature and elevated temperatures, and the ratio of the measured resistance ($R$) to the room temperature resistance ($R_0$) is plotted against the temperature increase. The dashed lines represent linear fits of the data, and the slopes of these lines are the TCRs. The TCRs are listed in Table S3 below.](image-url)
Table S3. Compiled device information

<table>
<thead>
<tr>
<th>Device</th>
<th>Thickness (nm)</th>
<th>Width (µm)</th>
<th>Length (µm)</th>
<th>TCR\textsubscript{line1} (K\textsuperscript{-1})</th>
<th>TCR\textsubscript{line2} (K\textsuperscript{-1})</th>
<th>R\textsubscript{line1} (Ω)</th>
<th>R\textsubscript{line2} (Ω)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>10</td>
<td>3.7</td>
<td>5.9</td>
<td>1.74×10\textsuperscript{-3}</td>
<td>1.77×10\textsuperscript{-3}</td>
<td>1104</td>
<td>1157</td>
</tr>
<tr>
<td>2</td>
<td>12</td>
<td>5.6</td>
<td>4.2</td>
<td>1.87×10\textsuperscript{-3}</td>
<td>1.86×10\textsuperscript{-3}</td>
<td>961</td>
<td>997</td>
</tr>
<tr>
<td>3</td>
<td>18</td>
<td>3.8</td>
<td>2.5</td>
<td>1.80×10\textsuperscript{-3}</td>
<td>1.74×10\textsuperscript{-3}</td>
<td>1202</td>
<td>1389</td>
</tr>
<tr>
<td>4</td>
<td>23</td>
<td>5.6</td>
<td>5.4</td>
<td>1.49×10\textsuperscript{-3}</td>
<td>1.50×10\textsuperscript{-3}</td>
<td>1440</td>
<td>1622</td>
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<tr>
<td>5</td>
<td>30</td>
<td>3.9</td>
<td>3.5</td>
<td>1.59×10\textsuperscript{-3}</td>
<td>1.62×10\textsuperscript{-3}</td>
<td>1420</td>
<td>1593</td>
</tr>
<tr>
<td>6</td>
<td>96</td>
<td>13.6</td>
<td>5</td>
<td>1.94×10\textsuperscript{-3}</td>
<td>1.90×10\textsuperscript{-3}</td>
<td>938</td>
<td>1030</td>
</tr>
</tbody>
</table>

In Table S3, we tabulate the physical dimensions of each exfoliated WSe\textsubscript{2} sample, measured using atomic force microscopy (AFM). Although all Pd lines are nominally the same thickness of evaporated material, small differences in the processing conditions or imperfections in patterning may lead to differences in their TCR values and resistances. Therefore, we individually calibrate TCR values for each line separately and compile them in Table S3. There is some variation, however the measured TCR values are within a reasonable range for thin, evaporated Pd.\textsuperscript{11}

Table S4. Absolute temperature rises over WSe\textsubscript{2} samples

<table>
<thead>
<tr>
<th>Device</th>
<th>ΔT\textsubscript{line1} (K)</th>
<th>ΔT\textsubscript{line2} (K)</th>
<th>ΔT\textsubscript{across} (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>14.7</td>
<td>10.6</td>
<td>4.1</td>
</tr>
<tr>
<td>2</td>
<td>12.9</td>
<td>10.1</td>
<td>2.8</td>
</tr>
<tr>
<td>3</td>
<td>19.9</td>
<td>14.8</td>
<td>5.1</td>
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<tr>
<td>4</td>
<td>22.7</td>
<td>16.6</td>
<td>6.1</td>
</tr>
<tr>
<td>5</td>
<td>21.0</td>
<td>16.8</td>
<td>4.2</td>
</tr>
<tr>
<td>6</td>
<td>13.1</td>
<td>9.9</td>
<td>3.2</td>
</tr>
</tbody>
</table>

In Table S4, we list the absolute temperature increases measured at the two Pd thermometer lines (ΔT\textsubscript{line1} and ΔT\textsubscript{line1}) when the heater current is applied, as well as the temperature difference between them (ΔT\textsubscript{across}). We note that the total temperature differences across the WSe\textsubscript{2} samples (ΔT\textsubscript{across}) are large enough to measure an appreciable induced thermovoltage, yet small enough such that we are able to neglect the Thomson effect from our analysis.

Section S4. Sheet Conductance Calculation Details

Due to the relatively long and thin Pd lines used as the source and drain contacts to the WSe\textsubscript{2} samples, there is a series resistance added to the WSe\textsubscript{2} channel resistance that becomes especially significant when the channel is in the on-state. Therefore, to more accurately estimate the sheet conductance (G\textsubscript{sh}) of the samples, we subtract the Pd line resistances as shown in the diagram of Figure S4 below. We apply a drain-to-source voltage (V\textsubscript{DS}), calculate the actual voltage drop across the WSe\textsubscript{2} (V\textsubscript{DS'}), then use this V\textsubscript{DS'} value to calculate G\textsubscript{sh}. Here,
\[ V'_{DS} = V_{DS} - I_D \frac{R_{\text{line1}} + R_{\text{line2}}}{2}, \text{ and therefore } G_{sh} = \frac{I_D L}{V'_{DS} W}. \]

The four-point resistance values for each Pd line at room temperature are included in Table S1.

**Figure S4.** A simplified circuit diagram showing the current path through half of one Pd line, through the WSe\(_2\) channel, and out through the other half Pd line.

**Section S5. Measured Thermovoltage vs. Heater Current**

When measuring low voltage values, we validate that these are from the thermovoltage, and not from spurious signals. In Figure S5, we plot the measured voltage between the source and drain contacts (i.e. the two parallel thermometer lines) against the heater current. Because Joule heating is directly proportional to \(I^2R\) and the Seebeck voltage is directly proportional to the temperature gradient, the measured voltage should be approximately quadratic with respect to the heater current. This relationship is confirmed in Figure S5.

Additionally, we further verify that the measured voltage is not due to leakage from the heater line by reversing the direction of the heater current and noting that the measured thermovoltage value does not change, in magnitude or sign.

**Figure S5.** The voltage measured between the source and drain contacts of a 96 nm thick WSe\(_2\) sample biased at a gate-source voltage \(V_{GS} = -1\) V, plotted as a function of the applied heater current.
Section S6. Power Factor (PF) Calculation Considering Conducting Channel Thickness

In a 2D, multilayered transistor, the layers contributing to conduction are a subset of the total number of layers, and the specifics depend on the channel thickness as well as the applied gate-source voltage.\textsuperscript{12} In this analysis, we make a rough estimate that the majority of the current flows through the top two layers of the samples, and thus, we recalculate the PFs using this “effective” channel thickness ($t_{\text{eff}}$), where $t_{\text{eff}} = 1.3$ nm.\textsuperscript{13,14} As shown in Figure S6, the adjusted PF values for each device follow the same trend, increasing with electrical conductivity to a maximum and turning over as the Seebeck coefficient decreases. However, the PFs are higher when only $t_{\text{eff}}$ is considered, peaking at $\sim 9300$ $\mu\text{W/m/K}^2$ for holes and $\sim 3100$ $\mu\text{W/m/K}^2$ for electrons in the 10 nm sample. Compared to the values in Table S5 (further below), this represents one of the highest PF values for a $p$-type layered 2D material.

![Figure S6. Power factor (PF) plotted against electrical conductivity ($\sigma$), where $\sigma$ is calculated using an “effective” channel thickness, $t_{\text{eff}} = 1.3$ nm.](image)

Section S7. Estimating Electrical Band Gap Variations Between Devices

For a high enough gate capacitance, the difference between the $n$- and $p$-branch threshold voltages at low $V_{\text{DS}}$ in our ambipolar WSe$_2$ can be used to roughly approximate the material band gap.\textsuperscript{15} We perform this extraction from the transfer characteristics of our devices and plot the sheet conductances ($G_{\text{sh}}$) against $E_G$ in Figure S7(a) and (b) for holes and electrons, respectively. Devices with higher extracted $E_G$ values tend to have lower $G_{\text{sh}}$ for a given $V_{\text{GS}}$, and consequently, the PF values are also lower, as shown in Figure S7(c) and (d). While the Seebeck coefficient depends on the shape of the band structure, which contributes to the effective mass of the carriers, it is not directly dependent on $E_G$ itself. Although the range of extracted $E_G$ values is larger than expected for the sample thicknesses here (due to device-to-device variation), the overall trend is consistent with wider band gap samples having a lower PF.
Figure S7. Plots of $G_{sh}$ for (a) holes and (b) electrons as well as PF for (c) holes and (d) electrons against the estimated $E_G$, extracted from the transfer characteristics of the flakes. There is a general trend of $G_{sh}$ decreasing with increasing extracted $E_G$, and therefore a corresponding decrease in PF.

Section S8. Comparison of Our Results with Existing Literature

The second row of Table S5 below contains values from our work, and subsequent rows contain a compilation of other thermoelectric studies of semiconducting 2D materials. The maximum Seebeck coefficient and power factor (PF) values reported in these works are listed, along with the material type, thicknesses measured, and further pertinent details. In the last column, we list the method used to gate the material and modulate its carrier density.

From this table, we note that Seebeck coefficients that are orders of magnitude higher than the median reported value are at very low carrier densities, such that the power factor would likely be too low for use in efficient thermoelectric energy harvesting. Additionally, for 2D materials specifically, it is important to distinguish between the total material thickness vs. the thickness that is conducting charge—these values will differ for multilayer systems and affect the estimation of electrical conductivity. Our reported Seebeck coefficient is one of the highest obtained for a $p$-type 2D material, and our PF values are competitive with other works when only the thickness of the conducting channel ($t_{eff}$) is considered.
Table S5. Comparing these results to literature values

| Ref.                      | Material | Thicknesses Studied | $|S|$ [$\mu$V/K] | PF  [$\mu$W/m/K$^2$] | Gating          |
|---------------------------|----------|---------------------|----------------|-----------------------|-----------------|
| This work                 | WSe$_2$  | 10-96 nm            | 950 (p-type, 96 nm) | 1200 (p-type, 10 nm) | EMIM-TFSI       |
|                           |          |                     | 500 (n-type, 12 nm) | 400 (n-type, 10 nm)  |                 |
| M. Kayyalha et al.$^1$    | MoS$_2$  | 1-23 layers         | 500 (n-type, 4 layers) | 5000 (n-type, 2 layers) | 300 nm SiO$_2$/Si |
| M. Yoshida et al.$^{13}$  | WSe$_2$  | 3 layers*           | ~300 (p-type, 3 layers) | 3200 (n-type) | DEME-TFSI       |
|                           |          |                     | ~280 (n-type, 3 layers) | 3700 (p-type) |                 |
| Hippalgaonkar et al.$^2$  | MoS$_2$  | 1, 2, 3 layers      | ~520 (n-type, 2 layers) | 8500 (n-type, 2 layers) | 275 nm SiO$_2$/Si |
| S. Timpa et al.$^{16}$    | WSe$_2$  | ~3-4 nm (4-6 layers)| 180 (n-type, 6 layers) | 2400 (p-type, 4 layers) | 40-60 nm hBN/Au/280 nm SiO$_2$/Si |
| J. Pu et al.$^{17}$       | MoS$_2$, WSe$_2$ | monolayer | 380 (p-WSe$_2$) | 300 (p-WSe$_2$) | EMIM-TFSI |
|                           |          |                     | 250 (n-WSe$_2$) | 100 (n-WSe$_2$) |                 |
|                           |          |                     | 160 (n-MoS$_2$) | 200 (n-MoS$_2$) |                 |
| M. Buscema et al.$^{18}$  | MoS$_2$  | monolayer           | $1 \times 10^5$ (n-type**) | Not reported | 285 nm SiO$_2$/Si, laser heating |
| J. Wu et al.$^{19}$       | MoS$_2$  | monolayer           | $3 \times 10^4$ (n-type*** | Not reported | 285 nm SiO$_2$/Si |
| Y. Saito et al.$^{20}$    | BP       | 40 nm               | 510 (p-type****) | 460 (p-type) | DEME-TFSI |
| S. Choi et al.$^{21}$     | BP       | 10 and 30 nm        | 400 (p-type) | Report “sheet” PF | 300 nm SiO$_2$/Si |
| J. Fleurial et al.$^{22}$ | Bi$_2$Te$_3$ | Bulk (3 mm) | 250 (n-type) | Not reported | Annealed to vary carrier concentrations |
|                           |          |                     | 230 (p-type) |                 |                 |
| M. Saleemi et al.$^{23}$  | Bi$_2$Te$_3$ | Bulk, nanostructured | 120 (n-type) | 2800 (n-type) | None |

*to calculate $\sigma$, only the thickness of the conducting channel, 1.3 nm, is considered

**Seebeck value is measured for a very low carrier density

***at 280 K and at very low carrier density

****at 210 K
Section S9. Temperature-Dependent Electrical and Seebeck Measurements

For the thinnest two WSe$_2$ samples in this work, we carry out temperature-dependent electrical and Seebeck coefficient measurements, which are summarized in Figure S8 below. Transfer characteristics as well as Seebeck coefficients are plotted for 296 K (both in the beginning and again after cooling back down), 350 K, and 400 K. The temperature range we measure over is restricted due to limitations of the ion gel used. At higher temperatures, the ion gel becomes more conductive and leakage currents increase$^{24}$; in turn, any current through the ion gel causes measurements to become unreliable. These measurements confirm that the WSe$_2$ sample characteristics are consistent and stable between room temperature and 400 K. Additionally, these values do not include the corrections from the finite-element method simulations, which were not performed for higher temperatures.

**Figure S8.** Temperature-dependent transfer characteristics for (a) 10 nm and (b) 12 nm thick WSe$_2$ samples, and Seebeck coefficients for the same (c) 10 nm and (d) 12 nm thick samples. Use of the ionic liquid limits our temperature range between approximately room temperature and 400 K to ensure stable measurements.

**Supporting References:**


