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Unraveling Thermal Transport Properties of MoTe₂ Thin Films Using the Optothermal Raman Technique

Carlos Rodriguez-Fernandez,* Arttu Nieminen, Faisal Ahmed, Jesse Pietila, Harri Lipsanen, and Humeyra Caglayan*

ABSTRACT: Understanding phonon transport and thermal conductivity of layered materials is not only critical for thermal management and thermoelectric energy conversion but also essential for developing future optoelectronic devices. Optothermal Raman characterization has been a key method to identify the properties of layered materials, especially transition-metal dichalcogenides. This work investigates the thermal properties of suspended and supported MoTe₂ thin films using the optothermal Raman technique. We also report the investigation of the interfacial thermal conductance between the MoTe₂ crystal and the silicon substrate. To extract the thermal conductivity of the samples, temperature- and power-dependent measurements of the in-plane E₁g and out-of-plane A₁g optical phonon modes were performed. The results show remarkably low in-plane thermal conductivities at room temperature, at around 5.16 ± 0.24 W/m·K and 3.72 ± 0.26 W/m·K for the E₁g and the A₁g modes, respectively, for the 17 nm thick sample. These results provide valuable input for the design of electronic and thermal MoTe₂-based devices where thermal management is vital.

KEYWORDS: optothermal characterization, MoTe₂, Raman spectroscopy, TMDs, thermal conductivity, and interfacial thermal conductance.

INTRODUCTION

Transition-metal dichalcogenides (TMDs) have been extensively studied as potential materials for the next generation of optoelectronic devices because of their unique and tunable physical and chemical properties at nanometric scale, including thickness-dependent band gaps, strain engineering, high carrier mobility, broadband optical absorption, high optical response, etc.¹⁻⁷ These layered materials exhibit strong covalent bonds in-plane and weak out-of-plane van der Waals forces that enable the preparation of uniform atomic layers by mechanical or chemical exfoliation. Moreover, these materials have good mechanical strength and nanoscale dimensionality, resulting in huge potential for electronic applications, energy, storage, biomedicine, catalysis, etc.⁸⁻¹¹ Besides these applications, mono- and few-layer TMD materials have been considered forthcoming candidates for thermoelectric (TE) and energy-efficient device applications.¹²,¹³ The conversion efficiency to transform waste heat into electrical power of a TE material is defined by the dimensionless figure of merit ZT which is directly proportional to the square of the Seebeck coefficient and inversely proportional to the thermal conductivity.¹⁴ The dichalcogenides of group VI, 2H-MX₂ (M = Mo, W; X = S, Se, Te) are promising materials for these applications, owing to their phonon properties, relatively low thermal conductivity, and high Seebeck coefficient.¹⁵⁻¹⁷

Among the widely studied TMDs, the compound MoTe₂ has been in the spotlight for its polymorphic nature at room temperature with an extremely small energy difference (40 meV) between the semiconducting 2H phase (α-phase) and the metastable semi-metallic 1T’ phase (β-phase).¹⁸⁻²⁰ In addition, MoTe₂ is the only material among group VI that can be grown in both α- (2H) and β-(1T’) phases, and their phase change has been reported by strain,²¹ joule heating,²² ionic liquid gating,²³ electrostatic doping,²⁴ and laser irradiation.²⁵ This makes MoTe₂ an excellent candidate for the fabrication of two-dimensional (2D) hetero-phase homojunction and opens new possibilities for designing low-cost applications in utilizing the phase-engineering of MoTe₂. Nevertheless, as reported earlier, the characteristics of MoTe₂ are greatly influenced by external perturbations such as thermal stress compared with

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other counterpart materials²⁵ The influence of thermal stress is directly proportional to the heat capacity and thermal conductivity of a certain material. This combined with the readily phase change of MoTe₂ by localized heating may generate Te migration during the operation of MoTe₂-based memory devices²⁶ or heat-induced structural phase transition of the 2D MoTe₂ film under Joule heating²⁷ Understanding phonon properties would enable the extraction of the thermal coefficient and thermal conductivity, which is of fundamental importance for such device applications. The thermal transport properties of MoTe₂ have only been investigated theoretically via computational methods²⁸,²⁹ which revealed MoTe₂ as a promising candidate for thermoelectric applications. Although these studies provide a basic understanding of thermal transport in monolayer 2H-MoTe₂ for future applications, the thermal transport properties have not been experimentally analyzed.

The noncontact optothermal Raman technique has been considered one of the most robust methods for measuring the thermal properties and thermal conductivity of a wide range of 2D materials. In this technique, the sample is exposed to a focused laser beam, and the thermal response is measured using a temperature-sensitive detector. The temperature change induced by the laser is directly proportional to the laser power and the thermal properties of the sample. The technique is not limited to atomically thin layers and has also been extended to relatively thick layers with adequate laser power intensity to cause the local heating of the sample. In this work, we present the temperature- and power-absorbed- dependent analysis of thin MoTe₂ films that permit the extraction and calculation of the interfacial thermal conductance with the substrate and the thermal conductivity of the material. The obtained room-temperature thermal conductivity for MoTe₂ flake is remarkably low compared with other TMD materials characterized by the optothermal Raman method. These results provide valuable information about the understanding of the heat conduction of MoTe₂ and should be considered in the fabrication of polymorphic engineering of MoTe₂ for thermoelectric and optoelectronic applications.

RESULTS AND DISCUSSION

Prior to the detailed optothermal analysis, multilayered MoTe₂ flakes were mechanically exfoliated from commercially purchased MoTe₂ crystal using Scotch tape and transferred onto clean Si substrates with prefabricated holes. The Si substrate presents a higher thermal conductivity (around 150 W/m·K)³¹ that can act as a heat sink. This enables the direct comparison of the temperature and power coefficients between the suspended and the supported characterized samples. The schematic of the optothermal Raman technique employed in the characterization of MoTe₂ flakes suspended over circular holes on the Si substrate can be observed in Figure 1a. This technique requires a two-step procedure. First, the temperature...
dependence of the position of the optical phonon modes of the MoTe₂ flakes is studied to extract the first-order temperature coefficient ($\chi_T$), which acts as a Raman thermometer. After that, the laser-power-dependent Raman spectra of the selected phonon modes are recorded to determine the amount of absorbed power on either the supported or the suspended MoTe₂ at room temperature. The 100× and 50× objectives with different numerical apertures were employed to perform the power-dependent measurements. Having the local temperature raised by the laser power, the heat diffusion equations can be solved. The cross section of the MoTe₂ on the Si substrate is also displayed in Figure 1a, where holes with a diameter of 10 μm were fabricated by maskless lithography (see the Methods Section). The microscope image in Figure 1b presents a thin MoTe₂ flake that covers holes on the Si substrate. The multilayer MoTe₂ samples with a thickness in the range of 15–40 nm are preferred in our experiments due to the fact that multilayer samples are environmentally robust, exhibit band gap close to Silicon, and can be transformed from semiconducting to the metallic phase with laser. Importantly, we use two samples with similar thicknesses, noted as sample 1 (17 nm thick) and sample 2 (37 nm thick), in order to monitor the reproducibility of the employed method for the determination of thermal conductivity. The height profile measured by atomic force microscope (AFM) across the edge of the flakes is displayed in Figure 1d for both samples. To corroborate the quality of the transferred sample over the holes, the samples were characterized by micro-Raman spectroscopy at room temperature and low power intensity, as presented in Figure 1e.

As the point group of semiconducting bulk 2H-MoTe₂ belongs to $D_{3d}$ symmetry, the unit cell is composed of six atoms, resulting in 18 phonon modes (15 of them are optical modes). Due to the center of inversion, bulk 2H-MoTe₂ can be split into gerade (marked with subindex g) and ungerade (subindex u) modes. The gerade modes present a change in the polarizability but do not have a dipole moment, and they are Raman-active modes. Ungerade modes have a dipole moment and, therefore, these modes are infrared active. Since bulk 2H-MoTe₂ is a center-symmetric crystal, the Raman modes cannot appear in an infrared spectrum and vice versa, they are exclusive. Thus, the irreducible representation of the lattice vibration at Γ point of the Brillouin zone (zone center) can be expressed as $\Gamma = A_{1g} + 2A_{2g} + 2B_{1g} + 2B_{2g} + 2E_{1g} + 2E_{2g}$ (E modes are double degenerated), where the Raman-active modes are $A_{1g}$, $B_{1g}$, and $B_{2g}$, while the bulk $B_{2g}$ modes are optically inactive. In the selected region of the Raman spectrum (17 nm flake) of Figure 1e can be identified the characteristic optical phonon modes $A_{1g}$, $E_{2g}$ (A modes are double degenerated), where the Raman-active modes are $A_{1g}$, $B_{1g}$, and $B_{2g}$, while the bulk $B_{2g}$ modes are optically inactive. In the selected region of the Raman spectrum (17 nm flake) of Figure 1e can be identified the characteristic optical phonon modes $A_{1g}$, $E_{2g}$ (E modes are double degenerated), where the Raman-active modes are $A_{1g}$, $B_{1g}$, and $B_{2g}$, while the bulk $B_{2g}$ modes are optically inactive. In the selected region of the Raman spectrum (17 nm flake) of Figure 1e can be identified the characteristic optical phonon modes $A_{1g}$, $E_{2g}$ (E modes are double degenerated), where the Raman-active modes are $A_{1g}$, $B_{1g}$, and $B_{2g}$, while the bulk $B_{2g}$ modes are optically inactive.

Figure 2 shows the calibration of temperature coefficients for the supported and suspended parts of the $E_{2g}$ and the $A_{1g}$ modes while heating the substrate (temperature ranges from room temperature to 500 K). All of the measurements were collected after the sample’s temperature was stable. The temperature increase leads to softening of the active Raman modes, and the $E_{2g}$ and $A_{1g}$ modes follow a systematic redshift with increasing temperature (see Figure 2a and b for suspended MoTe₂ sample 1). This evolution is given approximately by the linear equation $\Delta \omega = \chi_T \Delta T$ (Figure 2c).
and d), where $\chi_T$ is the first-order temperature coefficient for MoTe$_2$ optical modes and $\Delta T$ is the temperature change. Note that $E_{2g}^1$, the in-plane phonon mode, is more affected by the strain and thermal expansion of the lattice with respect to the $A_{1g}$. Furthermore, the fitting $\chi_T$ parameters for suspended MoTe$_2$ are slightly different from the supported part because of the influence of the thermal expansion coefficient mismatch between the sample and the substrate.

To extract the linear relation between the sample temperature and the absorbed laser power, we investigated the effect of laser-induced heat on the selected modes for MoTe$_2$ suspended and over Si substrate at various laser powers. All spectra were collected using 50× (numerical aperture, NA = 0.5) and 100× (NA = 0.85) objectives. To avoid sample heating and the appearance of nonlinear effects, the Raman characterization was done at lower power intensities within the linear dependence range (see Figure S1, Supporting Information). For example, in Figure 3, we can observe the power-dependence evolution of the mentioned modes for supported (Figure 3a) and suspended (Figure 3b) MoTe$_2$ sample 1 using 100× objective. The data has been extracted from the Lorentzian fit of the phonon modes as a function of the laser power. The power-dependence measurements for the MoTe$_2$ sample 2 and Raman shift evolution using a 50× objective can be found in Figures S4 and S5. In general, we have observed that in all of the cases, the Raman peaks downshift as the laser power increases, indicating a local heating of the sample. The redshift of $E_{2g}^1$ and the $A_{1g}$ modes is more evident for the suspended samples because the incident laser beam is the main contributor to the heat change of the sample.

The linear Raman shift rates can be expressed by the equation $\Delta \omega = \chi_T \Delta P$, where $\chi_T$ is the first-order power coefficient and $\Delta P$ is the change in the laser power.
By combining equations for $\chi_T$ and $\chi_P$, we can obtain the change in temperature as a function of change in the absorbed laser power, defined as the thermal resistance $R_m$

$$R_m = \frac{\Delta T}{\alpha \Delta P} = \frac{\chi_T}{\chi_P}$$

(1)

where $\alpha$ is the absorption. We assume an absorption coefficient of 4.69 $\times$ 10^7 1/m for MoTe$_2$, which gives $\alpha$ values of 0.55 and 0.82 for 17 and 37 nm MoTe$_2$ samples, respectively.

Thermal resistance given by eq 1 correlates the value of the temperature increase with increasing laser power. To link this value to the thermal conductivity of the material, we need to calculate the theoretically predicted temperature profile in the case of an external heat source applied to the material, which in our case is the illuminating laser beam. The heat is diffused in the plane of the MoTe$_2$, and the efficiency of the diffusion, and thus the overall temperature increase, is directly related to the thermal conductivity. Also, in the supported parts of the MoTe$_2$, the heat is transferred to the Si substrate, which acts as a heat sink. Neglecting the heat convection to air and assuming continuous heating by the laser beam, the steady-state temperature profile is determined by the heat diffusion equations

$$\frac{k_1}{r} \frac{d}{dr} \left( r \frac{dT}{dr} \right) = -q_v(r), \text{ (suspended)}$$

(2)

$$\frac{k_2}{r} \frac{d}{dr} \left( r \frac{dT}{dr} \right) - G \frac{T(r) - T_0}{t} = -q_v(r), \text{ (supported)}$$

(3)

where $k_1$ is the thermal conductivity of the suspended (supported) MoTe$_2$, $G$ is the interfacial thermal conductance between the MoTe$_2$ and the substrate, $t$ is the thickness of the MoTe$_2$ flake, $T_0$ is the ambient temperature, and $q_v$ is the volumetric optical heating from the Gaussian laser beam

$$q_v(r) = \frac{\alpha P}{\pi r_0^2 G} \exp\left( -\frac{r^2}{r_0^2} \right)$$

(4)

where $r_0$ is the laser beam radius. The laser beam radius was experimentally determined by performing line scan Raman spectroscopy across an Au layer-coated cleavage edge (Figure 4a,b) using 100x and 50x objectives. Then, the Si peak intensity was extracted as a function of the moving position (dI/dx), which can be fitted by a Gaussian function $\exp(-x^2/r_0^2)$ for both: 100x (Figure 4c) and 50x (Figure 4d). The experimental values of $r_0$ were 0.23 and 0.35 $\mu$m, which slightly differs from the estimated through numerical aperture 0.19 and 0.33 $\mu$m for 100x (NA = 0.85) and 50x (NA = 0.5), respectively. The temperature profile depends on whether the MoTe$_2$ layer is either suspended or supported, leading to different boundary conditions. In all cases, the applied boundary conditions are $dT/dx|_{x=0} = 0$, and $T(r \rightarrow \infty) = T(a)$, ensuring that the temperature stays finite at the origin and that the temperature is the ambient temperature far away from the laser-illuminated area.

We will first start our analysis on supported MoTe$_2$ for which eq 3 applies everywhere. This analysis will help us obtain $k_2$ and $G$, which are also needed to calculate the thermal conductivity for the suspended MoTe$_2$, $k_1$. By applying the aforementioned boundary conditions, we can solve the temperature profile by using the variation of parameters technique

$$T(\gamma) = T_s + \frac{\alpha P}{\pi r_0 G}(K_0(\gamma) \int_0^r I_0(\gamma) \exp(-\gamma^2/r_0^2) d\gamma + I_0(\gamma) \int_r^{\infty} K_0(\gamma) \exp(-\gamma^2/r_0^2) d\gamma)$$

(5)

where we have defined an auxiliary parameter $\gamma(r) = r_0 \sqrt{G/k_2t}$ and $\gamma_0 = \gamma(r_0)$. The area of the illuminated laser beam determines the experimentally observed temperature $T_m$. This is obtained as an average temperature over the 2D space weighted by the Gaussian beam profile

$$T_m = \frac{\int_0^{\infty} T(\gamma) q_v(\gamma) r d\gamma}{\int_0^{\infty} q_v(\gamma) r d\gamma}$$

(6)

and thus the observed temperature change is $\Delta T = T_m - T_0$ and by dividing this with absorbed laser power $\alpha P$ we have the analytical value for the thermal resistance $R_m$. Thus, by using eq 6 with I, we have linked the experimentally obtained temperature increase with the analytical temperature profile determined by the thermal conductivity $k_2$ and interfacial thermal conductance $G$, which are the two unknown values. Thus, two different experiments are needed to determine them uniquely.

To determine $k_2$ and $G$, we follow the method presented by Cai et al., where the power-dependent Raman measurements are done for two different objectives, 100x and 50x in our case, which give two different thermal resistances $R_m$. By taking the ratio of these two values, we get a formula solely dependent on the ratio of the two parameters, $k_2/G$, which can then be solved numerically. Then, we applied the solved ratio to the thermal resistance of one of the objectives to extract both $k_2$ and $G$. 

[Table 1. Temperature and Power Coefficients, Thermal Conductivities, and Interfacial Thermal Conductances of MoTe$_2$]

<table>
<thead>
<tr>
<th>MoTe$_2$ mode</th>
<th>$\chi_T$ [cm$^{-1}$/K]</th>
<th>$\chi_P$ [cm$^{-1}$/mW]</th>
<th>$\chi_P$ [cm$^{-1}$/mW]</th>
<th>$G$ [MW/m$^2$K]</th>
<th>$k$ [W/m-K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>sample 1 A$^g$</td>
<td>$-0.0085 \pm 0.0006$</td>
<td>$-34.6 \pm 0.7$</td>
<td>$-21.4 \pm 0.7$</td>
<td>3.72 $\pm 0.26$</td>
<td>5.16 $\pm 0.24$</td>
</tr>
<tr>
<td>suspended E$^g$</td>
<td>$-0.0165 \pm 0.0005$</td>
<td>$-57.8 \pm 1.6$</td>
<td>$-23.2 \pm 1.3$</td>
<td>1.16 $\pm 0.2$</td>
<td>5.07 $\pm 0.3$</td>
</tr>
<tr>
<td>sample 1 A$^g$</td>
<td>$-0.0075 \pm 0.0004$</td>
<td>$-0.71 \pm 0.03$</td>
<td>$-0.32 \pm 0.02$</td>
<td>16.2 $^{\pm}$</td>
<td>23.9 $^{\pm}$</td>
</tr>
<tr>
<td>supported E$^g$</td>
<td>$-0.0138 \pm 0.0003$</td>
<td>$-0.93 \pm 0.05$</td>
<td>$-0.41 \pm 0.01$</td>
<td>$\sim 23.9^{a}$</td>
<td>$\sim 0.8^{a}$</td>
</tr>
<tr>
<td>sample 2 A$^g$</td>
<td>$-0.00977 \pm 0.0004$</td>
<td>$-8.1 \pm 0.6$</td>
<td>$-10.0 \pm 0.6$</td>
<td>13.2 $\pm 1.2$</td>
<td>12.8 $\pm 1.4$</td>
</tr>
<tr>
<td>suspended E$^g$</td>
<td>$-0.01519 \pm 0.0007$</td>
<td>$-12.9 \pm 1.2$</td>
<td>$-13.5 \pm 0.9$</td>
<td>13.2 $\pm 1.2$</td>
<td>12.8 $\pm 1.4$</td>
</tr>
<tr>
<td>sample 2 A$^g$</td>
<td>$-0.0075 \pm 0.0007$</td>
<td>$-1.05 \pm 0.03$</td>
<td>$-0.67 \pm 0.03$</td>
<td>5.5 $\pm 1.7$</td>
<td>18.1 $\pm 6.4$</td>
</tr>
<tr>
<td>supported E$^g$</td>
<td>$-0.01402 \pm 0.0003$</td>
<td>$-1.35 \pm 0.03$</td>
<td>$-0.85 \pm 0.03$</td>
<td>9.7 $\pm 2.3$</td>
<td>22.0 $\pm 9.3$</td>
</tr>
</tbody>
</table>

$^{a}$The extracted values of the supported sample 1 require further experimental analysis with different substrates for better estimation of the errors.
and \( G \). The results for the characterized modes of samples 1 and 2 are summarized in Table 1.

When the sample is suspended over the holes, eq 2 applies in the region \( 0 < r < R \) and eq 3 in \( r > R \), where \( R \) is the radius of the hole. In this case, two additional boundary conditions are needed, namely, that the temperature profile is continuous at \( R \), and \( -2\pi R k_2 dT/dr|_{r=R} = aP \), indicating that the heat transferred out of the suspended flake must equal the absorbed laser power.

By solving the differential equation, and assuming that \( r_0 \ll R \), we get the temperature profile for suspended MoTe, namely, in the region \( 0 < r < R \)

\[
T(r) = T_s + \frac{aP}{2\pi R k} \left( \frac{G_0}{k} \right) \int_0^r \frac{1}{r} \left( 1 + \frac{r^2}{r_0^2} \right) \ln \left( \frac{R}{r} \right) dr
\]

(7)

where \( E(-r^2/r_0^2) \) is the exponential integral. The temperature profile in the region \( r > R \) is given by eq (S1) in the Supplementary Information.

We can obtain the formula for \( k_i \) by using eq 6 in 7, and then implementation of the power-dependent Raman measurement only for one objective would be sufficient, as there is only one unknown parameter. By carrying out the integration and then solving for \( k_i \), we get the formula

\[
k_i = \frac{h}{m_k^2} \left[ R_m - \frac{1}{2\pi \sqrt{k} G} \int_0^R \frac{1}{r} \left( 1 + \frac{r^2}{r_0^2} \right) \ln \left( \frac{R}{r} \right) dr \right]^{-1}
\]

(8)

where \( I = \int_0^R \exp(-r^2/r_0^2) \left( \ln(r/R) + \frac{1}{2} E(-r^2/r_0^2) \right) dr \).

The simulated temperature profiles for 37 nm MoTe are given in Figure 5, with 100x objective. Figure 5a shows the temperature profile over the 2D plane for 40 \( \mu \)W laser power, with Figure 5b limiting itself to the x-axis, indicated by the green dashed line in Figure 5a. Figure 5b shows the zoomed-in part near the edges of the Si holes, where the material changes from suspended to supported, and the temperature decays to the room temperature \( T_r = 295 \) K. The decay length of the temperature on the supported part of 2D material is determined by the thermal conductivity of the material and the interfacial thermal conductance between the material and the substrate. Still, overall the temperature of the sample is close to the room temperature at the edges due to the large diameter of the holes compared with the spot size of the Gaussian beam and the Si substrate working as an effective heat sink. Thus, the exact values of \( k_1 \) and \( G \) do not significantly affect the calculated value of \( k_i \), and the results are not substantially different if the substrate is assumed to be a perfect heat sink. However, this assumption would not be valid in the case of much smaller holes or less heat-conducting substrate and better conducting 2D material. Hence, we used the more comprehensive method by analyzing the substrate part of the MoTe, which validates our results and gives more insight into the thermal dynamics of the material.

From the Raman measurements on the suspended 17 nm MoTe, we obtain \( \chi_r = -0.0085 \pm 0.0006 \) cm\(^{-1}\)/K and \( \chi_p = -34.6 \pm 0.8 \) cm\(^{-1}\)/mW for the A1g mode and 100x objective, giving us thermal resistance \( R_m = 7.3 \times 10^5 \) K/W. Inserting this into eq 8, the thermal conductivity is extracted as \( k_1 = 3.72 \pm 0.26 \) W/mK, which is substantially smaller when compared with other 2D materials. The reason might be high strain due to mechanical exfoliation, which can cause a significant decrease in thermal conductivity. Repeating the same analysis for 37 nm MoTe, we obtain \( k_1 = 13.2 \pm 1.2 \) W/mK.

Table 1 summarizes the experimental results and the calculated thermal conductivities for supported and suspended MoTe for both 17 nm and 37 nm samples. The extracted thermal conductivities \( k_2 = 18.1 \pm 6.4 \) W/mK (supported) and \( k_2 = 13.2 \pm 1.2 \) W/m-K (suspended) for 37 nm MoTe are considerably higher than the corresponding values for the 17 nm sample. This is in agreement with the results presented for MoS\(_2\) by Yuan et al., where nonmonotonic thickness dependence of thermal conductivity was observed, and in particular, 17 nm thickness is expected to have lower thermal conductivity than relatively thicker samples due to more considerable surface scattering. The differences in thermal conductivity can also be attributed to the results of mechanical exfoliation, which are expected to vary from sample to sample. Furthermore, the interfacial thermal conductance between the MoTe and Si substrate is lower for the 37 nm sample (\( G = 5.6 \pm 1.7 \) MW/m\(^2\)K) than for the 17 nm sample (\( G = 16.2 \) MW/m\(^2\)K) in contrast to previous analysis performed on MoS\(_2\). This can be explained by the differences during the dry transfer process, as the thinner sample required an additional heating/cooling step resulting in a significantly improved contact.
between the substrate and the sample (see the Methods Section). Nevertheless, further analysis of the interfacial thermal conductance with different substrates and different MoTe₂ thicknesses are required. analysis.

### CONCLUSIONS

In summary, we used the optothermal Raman method to experimentally determine the in-plane thermal conductivities and interfacial thermal conductance of MoTe₂ thin films with thicknesses 17 and 37 nm. The laser spot sizes were experimentally obtained by the knife edge method, which was slightly different than the theoretically predicted values and was proved to be essential to extract accurate thermal conductivity for the supported MoTe₂. The thermal conductivity of MoTe₂ was shown to be considerably lower than that of other TMD materials, with 17 nm MoTe₂ having remarkably low thermal conductivities $k_1 = 3.72 \pm 0.26$ W/m-K and $k_2 = 2.35$ W/m-K for the suspended and supported parts, respectively. Overall, we performed the first experimental study of the thermal properties in suspended MoTe₂ using the optothermal Raman technique. Our experimental results provide essential input on the fundamental thermal parameters and heat dissipation of MoTe₂ and confirm this material as a potential candidate for thermoelectric applications.

### METHODS

**Sample Fabrication.** The characterized MoTe₂ flakes were first mechanically exfoliated from clean bulk crystals (HQ, Graphene, 2H phase) using the polydimethylsiloxane (PDMS)-assisted dry transfer method. The samples were transferred onto clean Si substrates with diameter holes of 9–10 μm and depth around 3 μm. The large diameter and depth of the fabricated holes ensure the elimination of possible artifacts from the influence of the substrate for the extraction of thermal conductivity. For the transfer, the positions of the Si holes and the MoTe₂ were aligned under an optical microscope (HQ, Graphene, Manual transfer system HQ2D MAN). Once they were in contact, the Si substrate and PDMS were carefully separated by moving the substrate stage. The thinner sample 1 required an additional step where the sample was heated at 60 °C so that the PDMS was softened and the MoTe₂ adhesion to the PDMS was lost. However, the thermal expansion coefficient of both materials can lead to the formation of wrinkles which may affect the thermal conductivity and interfacial thermal conductance. To preserve the quality of the material, we kept the samples in a dry nitrogen box.

**Fabrication of Si Holes.** The silicon samples were cleaned using acetone, isopropanol, and O₂ plasma. The prepatterned holes of the Si substrates were fabricated before the deposition of the MoTe₂, as follows: (a) Photoresist AZ ECI 3027 was spin-coated on the samples at 3000 rpm and baked for 60 s at 100 °C. (b) The resist was exposed using the polydimethylsiloxane (PDMS)-assisted dry transfer method. The samples were transferred onto clean Si substrates with diameter holes of 9–10 μm and depth around 3 μm. The large diameter and depth of the fabricated holes ensure the elimination of possible artifacts from the influence of the substrate for the extraction of thermal conductivity. For the transfer, the positions of the Si holes and the MoTe₂ were aligned under an optical microscope (HQ, Graphene, Manual transfer system HQ2D MAN). Once they were in contact, the Si substrate and PDMS were carefully separated by moving the substrate stage. The thinner sample 1 required an additional step where the sample was heated at 60 °C so that the PDMS was softened and the MoTe₂ adhesion to the PDMS was lost. However, the thermal expansion coefficient of both materials can lead to the formation of wrinkles which may affect the thermal conductivity and interfacial thermal conductance. To preserve the quality of the material, we kept the samples in a dry nitrogen box.

**Raman Measurement.** Raman spectra were collected using an inVia Raman spectroscopy system (Renishaw) under 526 nm laser line excitation. The laser beam focused on the sample with 100× (NA = 0.85) and 50× (NA = 0.5) microscope objectives. All measurements were calibrated with silicon samples using the characteristic phonon peaks at 520.5 cm⁻¹. After measuring the phonon for calibration, we kept the spectrometer in the corresponding position in order to avoid displacements in the spectra; thus, our experimental values have an uncertainty smaller than 0.2 cm⁻¹. The samples were placed on a temperature-controlled heating platform to measure Raman peak shifts with temperature and laser power change (Linkam Stage THMS600). The samples were left to thermalize for 20 min at each temperature. To prevent additional laser heating, the temperature-dependent measurements were performed at low power intensity and close to the minimum value of the linear region presented in the power-dependent measurements for each sample (see Figures 3, S1, S4, and S5).

### ASSOCIATED CONTENT

#### Data Availability Statement

The authors declare that the data supporting the findings of this study are available within the paper and in the Supporting Information files.

#### Supporting Information

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsami.3c06134.

The online version contains supplementary material and includes nonlinear region of the absorbed power, laser irradiation in a few-layer MoTe₂ temperature- and power-dependent measurements for suspended and supported, estimation of the error of the thermal properties, the temperature profile of sample 1, and AFM of sample 1 (PDF).

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