Thermal Conductivities and Interfacial Thermal Conductance of 1- to 3-Layer WSe$_2$

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ABSTRACT Atomically thin materials such as graphene and semiconducting transition metal dichalcogenides (TMDCs) have attracted extensive interest in recent years, motivating investigation into multiple properties. In this work, we used the opto-thermal Raman technique to measure the thermal transport properties of a popular TMDC material WSe$_2$, in single-atomic layer (1L), bi-layer (2L), and tri-layer (3L) forms. We implemented more direct measurements of the 1L-3L WSe$_2$ under study, and discovered WSe$_2$’s absorption coefficient from the measurements. In addition, by comparing the response of 1L-3L WSe$_2$ using different laser spot sizes, we are able to obtain the lateral thermal conductivity of 1L-3L WSe$_2$ and the interfacial thermal conductance to the substrate. The trend of layer dependent lateral thermal conductivity and interfacial thermal conductance of 1L-3L WSe$_2$ is discovered. For 1L-3L WSe$_2$, the room-temperature thermal conductivities are $36 \pm 12$, $23 \pm 11$, and $18 \pm 5$ W/(m·K), respectively. Crucially, the interfacial thermal conductance of 1L-3L WSe$_2$ is found to be $2.89 \pm 0.45$, $3.55 \pm 0.51$, and $3.56 \pm 0.46$ W/(m·K), with a flattened trend starting the 2L, a finding that has important implications for thermal management design and modeling of electronic devices.

TEXT

Following the interest in graphene since its first isolation by mechanical exfoliation in 2004 [1-3], the broader family of two dimensional (2D) materials has been the subject of extensive attention thanks to their unique atomically thin structure and the resulting novel physical properties [4-20]. In particular, transition metal dichalcogenides (TMDC) materials have shown unique thermal, optical and electrical properties, such as enhanced figure of merit, band structure transitions, semiconducting transport behavior, and strong photoluminescence, which are distinct from those...
of graphene [8-10, 21-30]. Thus TMDC materials are intriguing for the emerging thermal, optical, and electrical applications. Although there are adequate research results published about thermal transport properties of the common TMDC materials such as 2D MoS$_2$ and MoSe$_2$ both experimentally and theoretically [27, 29, 31-40], the thermal conductivity remains unknown for the emerging thermoelectric material WSe$_2$ in the 2D form, which is the important parameter for the thermoelectric and thermal management applications.

While electrical and optical properties of 2D WSe$_2$ have been studied extensively [41-48], there are no experimental data in the literature on thermal transport in 2D WSe$_2$. The large disparity in thermal conductivities of various morphologies of WSe$_2$ samples predicted by computational studies [49-54] ranging from 0.2 W/m·K to 22 W/m·K motivates experimental investigation and verification. Meanwhile, a low thermal conductivity of 1.2-1.6 W/m·K was found for disordered layered WSe$_2$ in the bulk form by both electrical heating method and time-domain thermoreflectance (TDTR) method [55, 56]. Whereas in a recent experiment based on TDTR, a thermal conductivity value of 42 W/m·K was found for the ordered crystalline layered WSe$_2$ in the bulk form [57]. Therefore, it is needed to experimentally investigate the thermal conductivity of WSe$_2$ in the 2D atomic-layered forms (single-layer, bi-layer, and tri-layer) to discover the intrinsic in-plane thermal conductivity values of 2D WSe$_2$, the interfacial thermal conductance with substrate, and explore their dependence with layer number.

In this letter, we demonstrate the measurement of thermal conductivities of single-layer (1L), bi-layer (2L), and tri-layer (3L) WSe$_2$ and the interfacial thermal conductance between the sample and substrate, through a well-developed measurement method for discovering thermal conductivities of thin TMDCs. We implemented more direct optical measurements of the 1L-3L WSe$_2$ to discover WSe$_2$’s absorption coefficient. The systematic experimental results enable
comparisons of experimental results between different layer numbers. The opto-thermal Raman technique has been the most successful method for measuring thermal conductivity of 2D materials down to sub-nanometer thickness [20, 27, 34]. Compared to TDTR method which is famous for its capability of measuring cross-plane and in-plane thermal conductivity of 2D materials in the bulk form [58] and electrical heating method [56] which is renowned for its reliability in high-resolution temperature calibration for 2D bulk materials, opto-thermal Raman technique is fit for measuring intrinsic thermal conductivity of 2D materials with thickness down to sub-nanometer.

In opto-thermal Raman technique, laser is focused at the center of the sample to measure the position of a Raman-active mode. As the laser power is increased, sample is heated, which enables red-shift Raman mode due to thermal softening. Thermal modeling can then be used to extract the thermal conductivity from the measured shift rate. Thermal modeling used for determination of thermal conductivity requires additional input of a number of parameters: the rate of mode softening with temperature, optical absorbance, and the thermal conductance of the supported area of the flake. The obtained room-temperature thermal conductivity and thermal interfacial conductance for the 1L-3L WSe₂ are the first reported experimental results. In summary, opto-thermal Raman technique based measurement is conducted at steady state and independent of time, during which a Raman laser beam is focused on the sample and used as both heat source and thermometer. As heat source, laser power is absorbed uniformly throughout the thickness making it an ideal method in measuring thermal conductivities of samples with the smallest existing thickness existing (sub-nanometer). In addition, the non-contact measurement method has the minimum fabrication requirement and thus maintains the pristine physical properties of the sample.

The mechanical exfoliation method was used to synthesize 1L, 2L, and 3L 2D samples from a WSe₂ bulk crystal (SPI supplies) on a SiO₂ (285 nm)/Si substrate. After repeating the exfoliation
process and achieving the desired thickness of the crystal layers on a Scotch tape strip, the exfoliated flakes were transferred onto the SiO₂/Si surface by peeling off the tape. In optical microscopy (Nikon Eclipse), the use of a SiO₂ substrate with the thickness of 285 nm optimizes the white light contrast for better identification of 2D materials of 1L, 2L and 3L. Mechanical exfoliation material synthesis method, with its cleanest fabrication nature, ensures the pristine properties and cleanliness of the sample. The layer number was reconfirmed by atomic force microscopy (AFM) (Bruker Dimension) in tapping mode, and verified by Raman spectroscopy and photoluminescence [30]. Figure 1a shows a schematic diagram of the obtained WSe₂ on SiO₂/Si substrate with Raman laser measurement by 532 nm wavelength laser. Figure 1b shows the laser profile and thermal transport schematic in 1L WSe₂ flake. The AFM image in Figure 1c shows the topography of 1L, 2L and 3L WSe₂ flakes obtained by mechanical exfoliation, with the optical microscope image in Figure 1d as a reference. The detailed AFM information is recorded in Supporting Information. The thickness of 1L-3L WSe₂ samples are reconfirmed in the optical microscope and AFM images. Figure 1e demonstrates the Raman spectrum by 532 nm wavelength laser of WSe₂, with E\textsuperscript{1}\textsubscript{2g} and A\textsubscript{1g} peaks. E\textsuperscript{1}\textsubscript{2g} peak is used as the characterization peak for thermal transport measurement due to its clear and accurate signal. Figure 1f shows photoluminescence spectra of 1L-3L by 532 nm wavelength laser, which also reconfirms the 1L WSe₂ because it has a direct bandgap which is presented as a single peak PL curve. 1L WSe₂ has only one peak at about 1.65 eV, while 2L and 3L WSe₂ has two peaks at 1.53 eV and 1.60 eV, respectively. The peak intensity of 1L WSe₂ at 1.65 eV is at least 2.5 times of that of 2L WSe₂ at 1.60 eV, in accordance with previous report [30].

During the thermal measurement, a 532 nm laser beam is focused using an objective lens on the center of the WSe₂ flake. In this configuration, laser is focused on the center of the sample area,
and the heat is transmitted along the radial direction away from the center of the sample. Here we are considering measured temperature, laser power, laser spot size, and absorption coefficient of the sample. Two kinds of objective lenses, 40× and 100×, were set to generate two laser spot sizes and thus to obtain the two unknown parameters (thermal conductivity and interfacial thermal conductance). To determine the absolute power absorbed for heat flow analysis, 1L-3L WSe$_2$ flakes were separately exfoliated onto transparent quartz substrates, and their measured absorption spectra were used to determine the frequency-dependent complex dielectric function. The dielectric functions of sample, along with that of the substrate material, were then used to calculate the absorption coefficient at 532 nm using the standard transfer matrix method. The absorption coefficient $\alpha$ of WSe$_2$ can be determined by the measured optical transmittance $T_e$ from the following equation [59]:

$$T_e = \frac{(1-R)^2 \exp(-\alpha d)}{1-R^2 \exp(-2\alpha d)},$$

where $d$ is the film thickness, and in the case of normal incidence, $R$ can be expressed simply as a function of the refractive index $n$, i.e., $R = (n - I/n + I)^2$, with $n$ reconfirmed from [26]. The obtained $\alpha$ is $894734$ cm$^{-1}$ at wavelength of 532 nm. The obtained optical absorbance of 1L-3L WSe$_2$ are $5.7 \pm 1.1\%$, $10.9 \pm 2.1\%$, and $16.0 \pm 2.8\%$ at 532 nm wavelength (Table 1).

The thermal measurement requires a careful determination of the radius of the laser beam spot ($r_0$). It is obtained by performing a micro-Raman scan across a sharp edge of a flake. Figure 2a shows Raman spectra mapping monitoring Si peak. The range between red lines is a large-scale frequency measuring range (500 cm$^{-1}$ to 550 cm$^{-1}$). Figure 2b,c shows optical microscope image and 10 $\mu$m $\times$ 10 $\mu$m micro-Raman map integrated from the 500 cm$^{-1}$ to 550 cm$^{-1}$ frequency range of the Si peak obtained. Then, the Si Raman peak intensity was obtained as a function of position,
and a Gaussian function fitted on the corresponding data points (Figure 2d,e). Derivation of peak intensity with respect to distance ($dI/dx$) can also be calculated. Gaussian function $K \cdot \exp(- (x - a)^2 / r_0^2)$ was fitted from the derivative function. $r_0$ was calculated from the fitting curves and determined as beam size radius. Through calculation, the laser spot radius was determined for $100 \times$ objective as $0.18 \pm 0.02 \, \mu m$ and $40 \times$ objective as $0.26 \pm 0.02 \, \mu m$, by micro-scanning across a long straight edge of a bulk WSe$_2$ flake. $r_0$ can also be estimated from the numerical aperture estimation: $r_0 = \lambda / (\pi \cdot NA)$. $NA$ is the numerical aperture value of the objective. $NA$ values are 0.9 and 0.75 for $100 \times$ and $40 \times$ objectives respectively, and $r_0$ was calculated as $0.19 \, \mu m$ and $0.23 \, \mu m$. This estimated value is close to the experiment results. Laser spot size is crucial to solve Bessel function in order to calculate thermal conductivity which will be discuss in the next section.

Raman spectroscopy was used to study the temperature influence of laser heating on the 1L-3L WSe$_2$ samples. We first calibrated the shift rate of the Raman $E_{12g}$ peak position with temperature by heating the entire substrate. A continuous-wave green laser beam ($\lambda=532 \, nm$) was used as the incident light. The WSe$_2$ sample was heated from 298 K to 473 K on a heating stage (Linkam THMS600). Figure 3a shows an example of the temperature dependent Raman spectra of 1L WSe$_2$. The Raman $E_{12g}$ peak follows a red shift with increasing temperature. The Raman measurements of 2L and 3L WSe$_2$ are showing the similar trend. Figure 3b is the Raman $E_{12g}$ peak position as a function of temperature of 1L WSe$_2$. The observed linear red shift with increasing frequency is due to the thermally driven bond softening, as previously observed for graphene, MoS$_2$ and MoSe$_2$[27]. Figure 3b,c,d shows the temperature dependence of the $E_{12g}$ peaks for 1L-3L WSe$_2$. Because Raman $E_{12g}$ peak has higher signal intensity, it was used for thermal transport calculations (discussed in the following paragraphs). The linear shift rate of $E_{12g}$ with temperature is defined as
the first-order temperature coefficient (Table 1). It is found that the first-order temperature coefficient decreases with layer number, which trend agrees with previous work on 1L and multilayer MoS$_2$ and MoSe$_2$ [27]. This can be explained that with the increasing layer number of 2D WSe$_2$, the temperature dependent in-plane lattice expansion coefficient has a decreasing trend.

Laser power dependent Raman spectra were taken as the second part of thermal conductivity measurements, to obtain the relation between temperature and absorbed laser power. The E$^{12g}$ peak shift was measured as a function of absorbed laser power for 1L-3L WSe$_2$ samples, using 0.18 and 0.26 μm spot sizes. Figure 4a shows the shift rates for 1L WSe$_2$. These measurements were repeated for all samples (Figure 4b,c). The obtained absorbance, temperature coefficients, and absorbed power shift rates of 1L-3L WSe$_2$ are summarized in Table 1.

Figure 1a (inlet) shows schematic diagrams of heat conduction at steady state when the WSe$_2$ sample was heated by the laser. The radial temperature distribution $T(r)$ is governed by the absorbed power and conduction through the sample to the substrate [27]. Convection through air accounts for less than 1% of the total heat conduction and is ignored (see the Supporting Information). The total absorbed laser power $P$ was determined from the laser power and the absorbance (Table 1). Assuming a Gaussian profile and a spot radius of $r_0$, this can then be used to calculate the volumetric heating power density $q'''(r)$:

$$q'''(r) = P \cdot \frac{1}{t} \cdot \frac{1}{\pi r_0^2} \exp\left(-\frac{r^2}{r_0^2}\right)$$  \hspace{1cm} (2)$$

where $t$ is the thickness of the flake. Considering the interfacial thermal conductance between WSe$_2$ and substrate, $T(r)$ is explained using a cylindrical coordinate as:

$$\frac{1}{r} \frac{d}{dr} \left( r \frac{dT(r)}{dr} \right) - \frac{d}{k_s t} (T(r) - T_a) + \frac{q'''(r)}{k_s} = 0$$  \hspace{1cm} (3)$$
Here $T_a$ is the substrate temperature, $\kappa_s$ is the thermal conductivity of the supported portion of the flake, and $g$ is the interfacial thermal conductance between the flake and the substrate. The boundary conditions $(dT)/(dr)|_{r=0} = 0$ and $T(r \to \infty) = 0$ are also applied.

The measured temperature of the flake center is determined by the measured $E^{1\,2g}$ position using the shift rate determined above (Table 1). This value reflects the local temperature distribution, weighted by the Gaussian profile of the laser spot:

$$T_m = \frac{\int_0^\infty T(r) \exp\left(-\frac{r^2}{r_0^2}\right) r dr}{\int_0^\infty \exp\left(-\frac{r^2}{r_0^2}\right) r dr}$$  \hspace{1cm} (4)

The experimental results, as shown in Figures 3 and 4, and summarized in Table 1, allow calculation of the measured thermal resistance of each sample, given as $R_m = T_m/P$. For the 1L WSe$_2$ sample, we found $R_m = (1.225 \pm 0.074) \times 10^6 \text{K/W}$ and $(0.674 \pm 0.041) \times 10^6 \text{K/W}$, for the 0.18 and 0.26 $\mu$m spot sizes, respectively. To extract values of the fitting parameters $\kappa_s$, and $g$, we followed the method of Cai et al. [34] (See the Supporting Information.) Solving Equations 2–4 yields a value of $R_m$ that is a function of $\kappa_s$ and $g$. Moreover, the ratio of measured $R_m$ for the two different spot sizes is a function of the ratio $g/\kappa_s$. We therefore used the $R_m$ ratio to obtain $g/\kappa_s$, and then used $R_m$ for a single spot size to obtain $\kappa_s$ and $g$ independently. This analysis yields $\kappa_s = 36 \pm 12 \text{W/(m\cdot K)}$ and $g = 2.89 \pm 0.45 \text{MW/m}^2\text{K}$ for the 1L WSe$_2$. The $\kappa_s$ and $g$ values were obtained by a combination of using 100× and 40× lenses (with spot sizes of 0.18 and 0.26 $\mu$m). With the same method, thermal conductivity and interfacial thermal conductance values for the other samples are given in Table 2.

The thermal conductivity of WSe$_2$ is smaller than the other three common TMDC materials MoS$_2$, MoSe$_2$ [27]. This is due to the increased atomic mass in WSe$_2$. The thermal conductivity of
WSe\textsubscript{2} decreases with the layer number. This trend of decreasing thermal conductivity is also seen in exfoliated hBN, MoS\textsubscript{2}, and MoSe\textsubscript{2} \cite{27, 60} and can be attributed to a greater phase space the intrinsic scattering mechanisms: Umklapp phonon scattering resulting from crystal anharmonicity in thicker samples. The trend of decreased $\kappa_s$ with increasing layer number reveals increased anharmonic scattering of those phonons that contribute to the measured $\kappa_s$. Because mean free path and thus the relative contribution from ballistic resistance component $R_b$ decrease with increasing layer.

All of these exfoliated thin layers, in which there are no grain boundaries, have thermal conductivity much higher than that measured for bulk materials. The total thermal conductivity is limited by the phonon Umklapp and edge boundary scattering.

This work is the first experimental work of thermal conductivities of 1-3L WSe\textsubscript{2}. Compared to the previous reported computational work, this experimental studied 2D WSe\textsubscript{2} value is larger than the computational value which is in the scale of $\sim 1 \text{ W/mK}$ \cite{49-54}. This difference is mainly because the computational model is using a nano-ribbon shaped WSe\textsubscript{2} whereas the experimental studied WSe\textsubscript{2} is in micron size. This experimental method implemented an independent check on measurements of material absorbance, spot size, and studied interfacial thermal conductance $g$, a critical parameter for heat dissipation in electronic devices which ensures the precision of the measured value.

This experiment has addressed several important problems in thermal measurement of 2D materials with atomic thickness. First, the layer dependent in-plane thermal conductivity and interfacial thermal conductance of WSe\textsubscript{2} is measured and a trend approaching a constant is observed on the sample of 3 layers. Second, it confirms the theoretical prediction that WSe\textsubscript{2} has a
relatively low in-plane thermal conductivity among the family of TMDC materials that makes it a potential candidate for the thermoelectric applications. We used a refined version of the opto-thermal Raman technique to conduct the first experimental study of thermal transport properties of 1L-3L WSe2. In particular, the calibration of optical absorption coefficient, laser spot size, and the role of thermal coupling to the substrate are considered in the measurements. These results demonstrate more robust measurements of thermal transport in 2D materials, understanding of which is necessary for device modeling, thermal management, and other applications.
FIGURES.

Figure 1. (a) Schematic of the experimental setup for the WSe$_2$ sample by opto-thermal Raman technique. (b) Schematic of Raman laser and temperature profiles. (c) Atomic force microscopy image of 1L-3L WSe$_2$. (d) Optical microscope image of 1L-3L WSe$_2$. (e) Raman spectrum of 2D WSe$_2$ with two characteristic peaks ($E_{2g}^{1}$ and $A_{1g}$). (f) Photoluminescence spectra of 1L-3L WSe$_2$. 
Figure 2. (a) The Raman spectra mapping monitoring Si peak, (b) optical microscope image and (c) 10 μm × 10 μm micro-Raman map across a sharp flake edge. The Raman intensity (green) and extracted profile of the laser beam (orange) as a function of the beam position for laser spot from 100 × objective (d) and that from 40 × objective laser spot (e).
Figure 3. (a) Raman spectra of 1L WSe$_2$ recorded at temperatures from 298 K to 473 K. The temperature dependent $E^{12g}$ Raman peak shift measured on the 1L WSe$_2$ (b), 2L WSe$_2$ (c), and 3L WSe$_2$ (d).
Figure 4. Power dependent $E'_{2g}$ Raman peak shift measured using different laser spot sizes, on the 1L WSe$_2$ (b), 2L WSe$_2$ (c), and 3L WSe$_2$ (d).
Figure 5. (a) Layer dependent in-plane thermal conductivity and (b) interfacial thermal conductance of WSe$_2$ from 1L to 3L WSe$_2$. 
TABLES.

**Table 1.** First-Order Temperature Coefficients, Absorbance, and Power Shift Rates of 1L-3L WSe$_2$

|          | Temperature coefficient (cm$^{-1}$/K) | Absorbance (%) | Absorbed power shift rate (cm$^{-1}$/µW) |
|----------|--------------------------------------|----------------|------------------------------------------|
|          |                                      |                | 0.18 µm spot                              | 0.26 µm spot                              |
| 1L WSe$_2$ | -0.0187 ± 0.0022                      | 5.7 ± 1.1      | -0.0229 ± 0.0241                          | -0.0126 ± 0.0153                          |
| 2L WSe$_2$ | -0.0147 ± 0.0019                      | 10.9 ± 2.1     | -0.0145 ± 0.0278                          | -0.0081 ± 0.0125                          |
| 3L WSe$_2$ | -0.0132 ± 0.0004                      | 16.0 ± 2.8     | -0.0127 ± 0.0004                          | -0.0072 ± 0.0002                          |

**Table 2.** Thermal Conductivities and Interfacial Thermal Conductance of 1L-3L WSe$_2$

|          | Thermal conductivity (W/m·K) | Interfacial thermal conductance (MW/m$^2$K) |
|----------|-----------------------------|---------------------------------------------|
| 1L WSe$_2$ | 36 ± 12                     | 2.89 ± 0.45                                 |
| 2L WSe$_2$ | 23 ± 11                     | 3.55 ± 0.51                                 |
| 3L WSe$_2$ | 18 ± 5                      | 3.56 ± 0.46                                 |

**ASSOCIATED CONTENT**

**Supporting Information.** The Supporting Information is available free of charge on the ACS Publications website.

Atomic force microscopy measurement of 1L-3L WSe$_2$, convection from the suspended material to the air, details of $\kappa_s$ calculation (PDF).
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