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Anisotropic thermal transport in van der Waals layered alloys $\text{WSe}_{2(1-x)}\text{Te}_{2x}$

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Transition metal dichalcogenide (TMD) alloys have attracted great interest in recent years due to their tunable electronic properties and the semiconductor-metal phase transition along with their potential applications in solid-state memories and thermoelectrics among others. However, the thermal conductivity of layered TMD alloys remains largely unexplored despite that it plays a critical role in the reliability and functionality of TMD-enabled devices. In this work, we study the composition- and temperature-dependent anisotropic thermal conductivity of the van der Waals layered TMD alloys $\text{WSe}_{2(1-x)}\text{Te}_{2x}$ in both the in-plane direction (parallel to the basal planes) and the cross-plane direction (along the *c*-axis) using time-domain thermoreflectance measurements. In the $\text{WSe}_{2(1-x)}\text{Te}_{2x}$ alloys, the cross-plane thermal conductivity is observed to be dependent on the heating frequency (modulation frequency of the pump laser) due to the non-equilibrium transport between different phonon modes. Using a two-channel heat conduction model, we extracted the anisotropic thermal conductivity at the equilibrium limit. A clear discontinuity in both the cross-plane and the in-plane thermal conductivity is observed as *x* increases from 0.4 to 0.6 due to the phase transition from the 2H to the Td phase in the layered alloys. The temperature dependence of thermal conductivity for the TMD alloys was found to become weaker compared with the pristine 2H WSe_2 and Td WTe_2 due to the atomic disorder. This work serves as an important starting point for exploring phonon transport in layered alloys. *Published by AIP Publishing.*

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Transition metal dichalcogenides (TMDs) are a family of van der Waals layered materials showing promising applications in electronics,^{1–3} photonics,^{4–6} hydrogen evolution catalysts,^{7–9} and energy conversion and storage.^{10–12} Alloys of layered TMDs are of particular interest due to their tunable properties.^{13,14} For example, electronic bandgaps can be continuously changed in several TMD alloy systems including $\text{MoS}_{2(1-x)}\text{Se}_{2x}$,^{15,16} $\text{W}_x\text{Mo}_{1-x}\text{S}_2$,^{17,18} and $\text{W}_x\text{Mo}_{1-x}\text{Se}_2$.¹⁹ With an intrinsically large electronic power factor of TMDs,^{20–22} alloying the layered TMDs could potentially lead to highly efficient thermoelectrics where the thermal conductivity can be effectively reduced due to the mass-disorder scattering.^{23,24} Along this line, Gu and Yang²⁵ performed a first-principles study on the thermal conductivity of $\text{MoS}_{2(1-x)}\text{Se}_{2x}$ monolayers and predicted an order-of-magnitude reduction in the thermal conductivity of these alloys as compared with those of the pristine MoS_2 and MoSe_2 . However, there yet exists any experimental work on anisotropic thermal transport in layered TMD alloys, especially as a function of alloy compositions.

Intrigued by the unique electronic structures^{26–28} and the abundant phase transition^{29,30} in MoTe_2 and WTe_2 , MoTe_2 or WTe_2 based TMD alloys have also attracted intensive research, especially on the manipulation of the physical properties through controlling the phase transitions. For

example, the topological electronic states of $\text{W}_x\text{Mo}_{1-x}\text{Te}_2$ can be effectively manipulated by the alloy composition.³¹ A hysteresis effect during the 1T'-to-Td phase transition of $\text{W}_x\text{Mo}_{1-x}\text{Te}_2$ was observed, and the phase change can be tuned to occur at room temperature,³² which makes it a promising material for phase change memory devices.³³ Depending on the composition, $\text{WSe}_{2(1-x)}\text{Te}_{2x}$ also exhibits a 2H-to-Td phase transition, which can be used for bandgap tuning.³⁴ Such phase transition in ternary TMD alloys could be potentially exploited for optimizing thermoelectric performances.³⁵ However, there exists no study on how such a phase transition would affect the thermal transport properties along both the in-plane and cross-plane direction.

In this letter, we study the composition-dependent and temperature dependent anisotropic thermal conductivity of the ternary alloy of layered $\text{WSe}_{2(1-x)}\text{Te}_{2x}$ using time-domain thermoreflectance (TDTR) measurements. Figures 1(a) and 1(b) show that the atomic structure of the ternary TMD alloy $\text{WSe}_{2(1-x)}\text{Te}_{2x}$ can be in both the 2H phase and the Td phase. When the Se atom occupies the majority of the chalcogen sites, the TMD alloy shows a hexagonal 2H phase. A phase transition to the orthorhombic Td phase would happen when the Te atom becomes the majority. We prepared single crystalline samples of the layered TMD alloys for the measurement of composition dependent thermal conductivity. The composition fraction *x* of the $\text{WSe}_{2(1-x)}\text{Te}_{2x}$ crystals is varied from zero to one, including the pure WSe_2 (*x* = 0), $\text{WSe}_{1.8}\text{Te}_{0.2}$ (*x* = 0.1), $\text{WSe}_{1.6}\text{Te}_{0.4}$ (*x* = 0.2), $\text{WSe}_{1.2}\text{Te}_{0.8}$ (*x* = 0.4), $\text{WSe}_{0.8}\text{Te}_{1.2}$ (*x* = 0.6), $\text{WSe}_{0.4}\text{Te}_{1.6}$ (*x* = 0.8), and

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FIG. 1. (a) Top view and side view of the structure of the layered TMD alloy in the 2H phase. The gray, blue, and yellow spheres (not to scale) represent W, Se, and Te atoms, respectively. (b) Top view and side view of the structure of the layered TMD alloy in the 1Td phase. (c) and (d) SEM and optical (inset) images of (c) 2H WSe_{1.2}Te_{0.8} and (d) 1Td WSe_{0.4}Te_{1.6}. The scale bar of SEM images is 100 μm, and each grid in the optical image is 1 mm-by-1 mm. (e) and (f) XRD and EDX (inset) characterization of (e) 2H WSe_{1.2}Te_{0.8} and (f) 1Td phase WSe_{0.4}Te_{1.6}. (g) Schematic of the TDTR system used for measuring the anisotropic thermal conductivity. Abbreviations are listed as follows: EOM, electric-optical modulator; PBS/ NPBS, polarized/non-polarized beam splitter; and $\lambda/2$, half wave plate.

WTe₂ ($x = 1.0$). These samples are synthesized by chemical vapor transport in a one-step process, and the details of synthesis are described in the [supplementary material](#) (S1). A typical structure of the 2H and 1Td alloy samples is shown in Fig. 1(c) (WSe_{1.2}Te_{0.8}) and Fig. 1(d) (WSe_{0.4}Te_{1.6}), respectively. In addition, x-ray diffraction (XRD) and energy dispersive x-ray (EDX) results show that the single crystals of WSe_{1.2}Te_{0.8} [Fig. 1(e)] and WSe_{0.4}Te_{1.6} [Fig. 1(f)] used in this work crystallize in 2H and 1Td structures, respectively, which are consistent with the nominal compositions [inset, Figs. 1(e) and 1(f)].³⁴

We measure the anisotropic thermal conductivity of layered TMD alloys using a femtosecond laser-based TDTR system as shown in Fig. 1(g). This system splits the femtosecond laser into a pump beam and a probe beam. The pump beam is modulated by an electric-optical modulator (EOM) at a typical frequency from 0.3 MHz to 10 MHz to create a periodic thermal excitation on the surface of TMD alloy samples, where the surface of TMD alloy samples is deposited with an aluminum metal thin film (~ 75 nm). This Al transducer film absorbs the pump beam, and its reflectance changes linearly with the surface temperature rise. The surface temperature change is monitored by detecting the reflected signal of the probe beam which is delayed to arrive at the sample surface by a translational delay stage. The transient surface temperature response is described by the ratio $-V_{in}/V_{out}$ between the in-phase signal V_{in} and the out-of-phase signal V_{out} , as shown in Fig. 2(a). The transient curve is fitted by solving a model for heat conduction in multi-layers using the nonlinear least squared regression to extract the thermal properties.^{36–38} Details of our TDTR system along with the data reduction method can be seen in our previous publications.^{39,40}

Since the TMD alloys are layered crystals with drastically different in-plane bonding and interlayer coupling, the thermal conductivity should be strongly anisotropic along the in-plane and cross-plane directions. Our recent work

demonstrated that the in-plane and cross-plane thermal conductivities can be separately determined by choosing appropriate operational parameters, i.e., the modulation frequency f_0 and the root-mean-squared laser spot radius w between the pump spot radius w_0 and probe spot radius w_1 , i.e., $w = \sqrt{w_0^2 + w_1^2}$.^{40,41} When the laser spot radius w is much larger than the penetration depth $d_p = \sqrt{k_r/\pi C f_0}$, with k_r the in-plane thermal conductivity, C the volumetric heat capacity, and f_0 the modulation frequency, the temperature field is quasi-one-dimensional in the cross-plane direction, and the TDTR is only sensitive to the cross-plane thermal conductivity k_z for the determination of k_z .⁴⁰ After cross-plane thermal conductivity k_z is determined, k_r can then be measured using a tightly focused laser spot. In Fig. 2(a), we show an example of measuring k_z of WSe_{1.2}Te_{0.8} with WTe₂ fraction $x = 0.4$ using a spot radius $w = 8.4$ μm. The signal $-V_{in}/V_{out}$ is sensitive to k_z only, especially when the probe delay time is shorter than 1 ns. This allows us to measure k_z independently of k_r and the interface conductance G with the Al transducer. Interestingly, we observed that the measured thermal conductivity k_z decreases as the modulation frequency increases [Fig. 2(b)], if the one-channel heat conduction model with local equilibrium assumption in Refs. 36 and 38 is used for

FIG. 2. (a) Measured TDTR signal for WSe_{1.2}Te_{0.8} using a 76 nm Al transducer, a root-mean-square spot radius $w = 8.4$ μm, and a modulation frequency $f_0 = 2.16$ MHz. The TDTR signal is dominantly sensitive to k_z . The best-fit cross-plane thermal conductivity is $k_z = 0.42$ W/mK. (b) Frequency dependent k_z of WSe₂, WSe_{1.2}Te_{0.8}, WTe_{1.6}Se_{0.4}, and WTe₂. (c) Measuring in-plane thermal conductivity of the WTe_{1.2}Se_{0.8} sample using $w = 4.2$ μm. The best-fit $k_r = 10.1$ W/mK.

the data analysis. Such frequency-dependent k_z was also observed in other layered crystals such as pure TMDs³⁹ and in black phosphorus.⁴² Jiang *et al.* explained this as the out-of-local-equilibrium phonon transport between the low frequency and high frequency phonon modes.³⁹ In the cross-plane direction, the thermal conductivity is mainly contributed by the low frequency phonons with small heat capacity, while the high frequency phonons with large heat capacity have negligible thermal conductivity due to their “flat” phonon band structure.³⁹ Such a large mismatch between heat conductivity and heat capacity of different frequency bands of phonons tends to be in non-equilibrium. When the sample is subjected to periodic laser heating, the heat is accumulated in the high frequency phonons, resulting in their higher temperature than the low frequency phonons. To describe such non-equilibrium phonon transport, we use a two-channel heat conduction model⁴³ that separates the phonon heat carriers into the high frequency channel and the low frequency channel with their own temperatures, heat capacity, and thermal conductivity. In this model, the cross-plane thermal conductivity at the equilibrium limit (k_z^{eq}) can be calculated by the summation of the thermal conductivity of both channels, indicated by the dashed lines in Fig. 2(b). [See [supplementary material](#) (S3–S5) for details of fitting, uncertainty analysis, and division of phonon channels]. After k_z^{eq} is determined, the laser spot is tightly focused to $w = 4.2 \mu\text{m}$, so that the TDTR signal becomes sensitive to both the cross-plane thermal conductivity k_z^{eq} and the in-plane thermal conductivity k_r . As shown in Fig. 2(c), the in-plane thermal conductivity of $\text{WSe}_{1.2}\text{Te}_{0.8}$ is measured to be $k_r = 10.1 \text{ W/mK}$ with a spot radius of $4.2 \mu\text{m}$ and a modulation frequency of 0.353 MHz .

Using a similar method as described above, we have measured the in-plane and cross-plane thermal conductivities of all TMD samples as summarized in Figs. 3(a) and 3(b), respectively. We compare the thermal conductivity of the pure WSe_2 and WTe_2 crystals with the results available in the literature. For WSe_2 , our measurements for both $k_r = 40 \text{ W/mK}$ and $k_z = 2.45 \text{ W/mK}$ are consistent with our previous work³⁹ and the first principles calculations by Lindroth and Erhart.⁴⁴ However, the measured k_z is significantly higher than the TDTR measurements by Chiritescu *et al.*⁴⁵ and Muratore *et al.*,⁴⁶ probably because they neglected non-equilibrium transport. For WTe_2 , our measurement for the in-plane thermal conductivity ($k_r = 13.5 \text{ W/mK}$) agrees well

with the measurement by Zhou *et al.* ($k_r = 15 \text{ W/mK}$).⁴⁷ Interestingly, the k_r value of WTe_2 measured by TDTR is even higher than the first principles calculation of lattice thermal conductivity. There are two possible reasons for such discrepancy. First of all, the electrons might have a notable contribution to the total thermal conductivity measured by the TDTR since WTe_2 is a semi-metal,⁴⁸ while it was completely neglected by the first principles calculations of lattice thermal conductivity. The other possible reason is likely due to the computational error. WTe_2 has a rather complicated atomic structure. However, in the calculation by Liu *et al.*,⁴⁹ the authors used a relative small supercell ($2 \times 2 \times 1$ unit cells) for calculating the force constants to mitigate the large computation cost, which might lead to an underestimated thermal conductivity.⁴⁴ In the cross-plane direction, our measurement is $k_z = 1.34 \text{ W/mK}$, which agrees well with both the first principles calculations⁴⁹ and the measurements by others.^{47,50,51} For both the in-plane and the cross-plane direction, the thermal conductivity reduces as the composition fraction x or $(1-x)$ in $\text{WSe}_{2(1-x)}\text{Te}_{2x}$ approaches 0.5. The sharp change in thermal conductivity in both in-plane and cross-plane directions is observed as x increases from 0.4 to 0.6. This is due to the phase transition from the 2H to the Td phase as x increases from 0.4 to 0.6. Since phase transition has also been used for enhancing the thermoelectric figure of merit (ZT) for other chalcogenides such as SnSe ⁵² and Cu_2Se ,⁵³ such a phase transition of 2H to Td could potentially result in a higher ZT factor due to the decreased thermal conductivity both along the in-plane direction and the cross-plane direction.

In Fig. 4, we show the temperature-dependent k_r and k_z of the 2H $\text{WSe}_{1.2}\text{Te}_{0.8}$ ($x=0.4$) and Td $\text{WSe}_{0.4}\text{Te}_{1.6}$ ($x=0.8$), in comparison to the pure 2H WSe_2 and Td WTe_2 , respectively. In the entire temperature range from 80 K to 300 K, both k_r and k_z of 2H $\text{WSe}_{1.2}\text{Te}_{0.8}$ are greatly reduced compared with the pure WSe_2 due to the alloy scattering, and a similar reduction is also observed in $\text{WSe}_{0.4}\text{Te}_{1.6}$ in the

FIG. 3. Composition dependent (a) in-plane thermal conductivity and (b) cross-plane thermal conductivity of $\text{WSe}_{2(1-x)}\text{Te}_{2x}$ measured by TDTR. The thermal conductivity of pristine WSe_2 ($x=0$) and WTe_2 ($x=1$) is compared with first-principles calculations by Lindroth and Erhart⁴⁴ and Liu *et al.*⁴⁹ and the measurement by Jiang *et al.*,³⁹ Chiritescu *et al.*,⁴⁵ Muratore *et al.*,⁴⁶ Zhou *et al.*,⁴⁷ and Brixner⁵⁰ and Jana *et al.*⁵¹

FIG. 4. Temperature dependent (a) in-plane thermal conductivity and (b) cross-plane thermal conductivity of 2H WSe_2 and $\text{WSe}_{1.2}\text{Te}_{0.8}$ ($x=0.4$) and temperature dependent (c) in-plane thermal conductivity and (d) cross-plane thermal conductivity of WTe_2 and $\text{WSe}_{0.4}\text{Te}_{1.6}$ ($x=0.8$). The thermal conductivity of WSe_2 is taken from Ref. 39.

Td phase compared with WTe_2 . As shown in Figs. 4(a) and 4(c), the $k_r(T)$ curve flattens below 150 K for both 2H $\text{WSe}_{1.2}\text{Te}_{0.8}$ and Td $\text{WSe}_{0.4}\text{Te}_{1.6}$, while k_r kept increasing with the temperature for the pristine WSe_2 and WTe_2 . Since the alloy scattering due to mass disorder is temperature independent,⁵⁴ it becomes more dominant at lower temperature when the intrinsic three phonon scattering is much weaker at cryogenic temperature, contributing to the weaker temperature dependence of k_r below 150 K. It is interesting to note that the $k_z(T)$ curve of $\text{WSe}_{0.4}\text{Te}_{1.6}$ is much flatter than the $k_z(T)$ curve WTe_2 , as shown Fig. 4(d). Since $\text{WSe}_{0.4}\text{Te}_{1.6}$ has randomly distributed Se defects, the distribution pattern of the defects is expected to be very different in each monolayer, which breaks the periodicity in the cross-plane direction. As a result of such atomic disorder, the vibration modes are greatly localized in the cross-plane direction, resulting in the much lower k_z value and a much flatter $k_z(T)$ curve than the pristine TMD crystal. For the same reason, the $k_z(T)$ curve of $\text{WSe}_{1.2}\text{Te}_{0.8}$ showed a similar shape compared to $\text{WSe}_{0.4}\text{Te}_{1.6}$, as shown in Figs. 4(b) and 4(d). However, the temperature dependence of k_z of WSe_2 is very different from WTe_2 . Instead of increasing with decreasing temperature, the $k_z(T)$ curve of WSe_2 showed a peak near 150 K. Suggested by Lindroth and Erhart,⁴⁴ and Jiang *et al.*,³⁹ there could exist stacking faults that induce boundary scattering on the length scale of $\sim 150\text{ nm}$,⁴⁴ resulting in such a peak in the $k_z(T)$ curve and much lower k_z compared with first principles calculations. However, such a peak in the $k_z(T)$ curve is absent in WTe_2 . Such a different behavior of temperature dependent k_z can be explained by the different distribution of phonon mean free paths (MFPs) in WTe_2 and WSe_2 . As suggested by the first principles calculations, half of k_z of WTe_2 is contributed by phonons with MFPs between 200 nm and $1\text{ }\mu\text{m}$,⁴⁹ but 50% of k_z of WSe_2 is contributed by phonons with MFPs above $1\text{ }\mu\text{m}$.⁴⁴ Due to the much longer MFPs in WSe_2 than WTe_2 , the stacking faults are expected to have a much more pronounced effect on k_z in WSe_2 than WTe_2 .

In summary, we applied the ultrafast laser based TDTR technique to study the temperature-dependent anisotropic thermal transport of layered TMD alloys $\text{WSe}_{2(1-x)}\text{Te}_{2x}$. We observed that the cross-plane thermal conductivity depends on the modulation frequency of the TDTR pump beam due to the non-equilibrium transport between different phonon modes in the cross-plane direction. A two-channel heat conduction model is used to extract the cross-plane thermal conductivity at the thermal equilibrium limit. The in-plane thermal conductivity is then determined using a tightly focused laser spot with a radius of $4.2\text{ }\mu\text{m}$. Both the in-plane and cross-plane thermal conductivities are reduced at a higher alloy mixing level as the composition fraction x or $(1-x)$ in $\text{WSe}_{2(1-x)}\text{Te}_{2x}$ approaches 0.5. A clear discontinuity in both the cross-plane and the in-plane thermal conductivity is observed as x increases from 0.4 to 0.6 due to the phase transition from the 2H to the Td phase in the layered alloys. We also found that the temperature dependence of thermal conductivity for the TMD alloys becomes weaker compared with the pristine 2H WSe_2 and Td WTe_2 due to the atomic disorder. This work serves as an important starting point for exploring phonon transport physics in layered alloys.

See [supplementary material](#) for the detailed description of sample preparation, two-channel heat transfer model, data processing, and uncertainty analysis.

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