High Thermoelectric Performance in Hexagonal 2D PdTe$_2$ Monolayer at Room Temperature

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Figure S1. Phonon dispersion curve of (a) \( h \)-PdTe\(_2\) monolayer (b) \( p \)-PdTe\(_2\).
Figure S2. Total energy difference ($\Delta E$) vs strain ($\Delta l/l_0$) of (a)-(b) hexagonal structure along the AC and ZZ directions and (c)-(d) pentagonal structure along the $x$ and $y$ directions.
Figure S3. The linear fitting of the variation of the energy of the valence band maxima (VBM) and conduction band minima (CBM) of the PdTe$_2$ monolayer: (a)-(b) hexagonal structure along the AC and ZZ directions and (c)-(d) pentagonal structure along the x and y directions.
Figure S4. Temperature dependent carrier mobility ($\mu$) of the (a) hexagonal structure (b) pentagonal structure and relaxation time of (c) hexagonal structure (d) pentagonal structure.
Figure S5. Comparison of the electrical thermal conductivity obtained from Wiedemann-Franz law and Boltzmann transport equation (BTE) for (a)-(b) hexagonal structure along the AC and ZZ directions and (c)-(d) pentagonal structure along the x and y directions. Here, we illustrated the results at 300 K.
Figure S6. Calculated phonon lifetime as function of frequency of the PdTe$_2$ monolayer: (a) hexagonal structure and (b) pentagonal structure

In the hexagonal structure, the transvers (TA) and out of plan (ZA) acoustics branch had a large life time compared with that of the longitudinal acoustic branch (LA) at low frequency (0~2 THz). The TA branch reached the maximum value of 120 (ps) ~ 0.1 THz while the optical branch appeared at high frequency (> 3 THz) and it was almost negligible compared with that of the acoustic branch. In the pentagonal structure, the phonon lifetime of the acoustic branch was substantially decreased and also the life time of the optical branch appeared at low frequency with considerable values. Such behavior can be understood from the phonon dispersion curve (Fig.1S (a)-(b)). For instance, the hexagonal structure had very small phonon band gap between the acoustic and optical branches at low frequency while the hybridization between the acoustic and optical branches appeared in the pentagonal structure. This hybridization led to reduce the phonon life time in pentagonal compare to the hexagonal structure.
Figure S7. The calculated phonon group velocity as function of frequency of the PdTe$_2$ monolayer: (a-b) hexagonal structure along the AC and ZZ direction and (c-d) pentagonal structure along the $x$ and $y$ direction.

In the hexagonal structure, the phonon group velocity was dominated by the acoustic mode at low frequency (0–2 THz). Here, the longitudinal acoustic (LA) branch had larger value than the transvers (TA) and the out of plan (ZA) acoustic branch. On the other hand, the optical branch appeared at high frequency with lower magnitude. Such behavior can be understood from the phonon spectrum density (Fig.1-b). We also found that the phonon group velocity showed almost the same behavior in both the ZZ and AC directions and this is the reason why the lattice thermal conductivity had almost no anisotropy. In the pentagonal structure, the optical branch appeared at rather low frequency (~1.5THz) due to the hybridization of the optical and acoustic branch in the phonon dispersion (Fig.S1 (b)). Both LA branch and the optical branch had larger group velocity along $y$ direction and this feature can explain the anisotropy of the lattice thermal conductivity in the pentagonal structure.
Figure S8. Calculated ZT using the empirical law Wiedemann-Franz law to estimate the electronic thermal conductivity at room temperature as function of carrier concentration of the PdTe$_2$ monolayer: (a)-(b) hexagonal structure along the AC and ZZ directions and (c)-(d) pentagonal structure along the x and y directions.
Figure S9. Calculated open circuit electronic thermal conductivity $K_{e\text{-open}}$ using the correction term ($K_{e\text{-open}} = K_e - T\sigma_S^2$) at 300K, 500K and 700K as function of carrier concentration of the PdTe$_2$ monolayer: (a)-(b) hexagonal structure along the AC and ZZ directions and (c)-(d) pentagonal structure along the $x$ and $y$ directions.
Figure S10. Calculated open circuit ZT_{open} at 300K, 500K and 700K as function of carrier concentration of the PdTe$_2$ monolayer: (a)-(b) hexagonal structure along the AC and ZZ directions and (c)-(d) pentagonal structure along the x and y directions.