Supporting Information

Thermoelectric Performance of 2D Tellurium with Accumulation Contacts

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Supplementary Note 1: Temperature coefficient calibration

In order to convert the resistance of the Pt/Ni nano-strip thermometer, the temperature coefficient was first calibrated for a broad temperature range. The resistance was measured with lock-in amplifier using standard AC method. The sample was cooled down in a dilution refrigerator from room temperature to helium temperature. The temperature versus resistance curve is shown in Figure S1, which exhibits great linearity for a broad temperature range from 300 K to 30 K. The temperature coefficient, defined by the following equation: \( \frac{dR}{R} = \alpha T \), is extracted to be 0.0019 at the vicinity of room temperature, which can be used to convert the resistance of the thermometer into temperature.

![Figure S1](image)

**Figure S1** | **Calibrating the temperature coefficient of Pt thermometers.** The resistance versus temperature curve shows good linearity for a broad temperature range.
The temperature coefficient is extracted from the equation: \( \frac{dR}{R} = \alpha T \) to be 0.0019 at the vicinity of room temperature.

**Supplementary Note 2: Tuning the transistor performance with different metal contacts**

To understand how the work function of contact metal will affect the transistor performance, we fabricated multiple transistors with different contact metals on the same flake to eliminate the flake-to-flake variation. The device schematics was presented in Figure S2(a). A 2D Te flake was transferred onto a heavily doped silicon wafer as back gate with 90 nm SiO\(_2\) insulating layer. Two high work function metal (Pd, work function \( \sim 5.2\text{eV} \)) and two low work function metal (Cr, work function \( \sim 4.6 \text{eV} \)) was patterned with even spacing and deposited with electron beam evaporator. We now have a Pd-Te-Pd transistor, a Cr-Te-Cr transistor, and a Pd-Te-Cr Schottky diode. The transfer curves of these three devices were measured with the modulation of the back gate and plotted in the same figure in Figure S3(b). We notice that the Pd-contacted device shows strongest p-type behavior and largest drain current since the work function is aligned close to the valence band and there are no obstacles for hole transport. The Cr-Cr shows ambipolar behavior and the drain current is much smaller compared to Pd device since both the electron and hole current branch suffered from the large Schottky barrier. The Pd-Cr Schottky current shows asymmetric device performance: when device was forward biased, the hole current can be injected through Pd contact smoothly whereas when the device was reversely biased the current will be hindered by Cr contact.
Figure S2] Multiple channel transistors with different metal contacts on the same flake. (a) Schematic image of device structure. Two Pd contacts and two Cr contacts were placed on the same Te flake with even spacing. (b) Transfer curves of Pd-Pd transistor, Cr-Cr transistor, and Pd-Cr diode with forward and reverse biases.

Supplementary Note 3: Te Schottky diodes
Owing to the controllability of Te-to-metal contact behavior, we can achieve high-performance Te Schottky diodes. Figure S3 shows a Te Schottky diodes with Pd and Ti contacts respectively. A large rectification ratio over $10^3$ was measured at room temperature.
**Figure S3| Te-based Schottky diode devices.** (a) Schematic image of Schottky device structure. Pd and Ti contacts were fabricated onto 2D Te flakes. (b) I-V curves of a Schottky diode plotted in log scale. The rectification ratio is over $10^3$. Inset: the I-V current plotted in linear scale.

**Supplementary Note 4: Photovoltaic measurement based on Te Schottky diodes**
The Schottky diodes can be used as solar cells. Here we also explore the photovoltaic effect of Te Schottky devices. The Pd-Te-Cr diode devices with channel length of 5 μm were fabricated. 633 nm laser illuminated the center of the flake and I-V curves with and without the laser was plotted in the inset of Figure S4(a). Particularly for solar cells, we are interested in two figures-of-merit, the short-circuit current and open-circuit voltage which can be determined from the intercept of I-V curve, as shown in Figure S4(a). The output power of the solar cell $P_d$ can be calculated by multiplying the current and voltage which peaks at $\sim 0.25$ nW at 0.7 mV, as shown in Figure S4(b).
Figure S4| Demonstration of solar cell device based on Te Schottky diodes. (a) I-V curve of a Pd-Cr diodes with and without laser illumination. The open-circuit voltage and short-circuit current can be extracted from the intercept of the red I-V curve. Inset, the I-V curve on a broader sweep range. (b) The output power of the solar cell $P_d$ can be calculated by multiplying the current and voltage which peaks at $\sim 0.25$ nW at 0.7 mV.
Supplementary Note 5: In-plane thermal conductivity measured using Micro-Raman method

The thin-film tellurium sample (35-nm) for thermal conductivity measurement is suspended on a silicon trench (5-µm wide, 13-µm deep) by the PDMS stamping method\textsuperscript{1} after using the Langmuir–Blodgett transfer process\textsuperscript{2}. A 633-nm He-Ne laser beam is diffracted through a slit, and then focused using a 100x objective into a laser focal line. The length of the laser focal line is 7.5 µm, and is along the direction of the silicon trench, and the width is 0.45 µm, positioned at the center of the suspending region. The laser focal line heats up the tellurium film, creating a temperature gradient and inducing Raman scattering at the same time. The Raman scattering signal is collected and sent to a spectrometer (Horiba LabRam). By calibrating the Raman peak shift with temperature, the Raman signal can be used as a thermometer. Laser absorption by the tellurium film is determined by measuring reflection and transmission. The thermal conductivity can then be extracted using a heat transfer model with the knowledge of laser absorption, Raman shift/temperature, and geometry. This method is adapted from our previous works for measuring in-plane thermal conductivity of 2D materials\textsuperscript{3–5}.

Figure S5(a) shows the measured Raman spectrum of the suspended 35-nm tellurium thin-film. The Raman peak shift of $A_1$ mode vs. temperature is shown in Figure S5(b) and is used as the thermometer, due to its higher temperature sensitivity and good linearity. The sample is then heated using the laser focal line, and the Raman shift vs. laser power is showed in Figure S5(b). A numerical model is setup to calculate the temperature rise under certain laser power, with varied thermal conductivity assumed (Figure S5(c)).
Comparing to the numerical model (Figure 3(b)), the thermal conductivity is extracted to be 1.50 W/m-K. The detailed studies on thermal transport of Te films including first-principle calculations are being prepared for another publication.

**Figure S5** Measurement of in-plane thermal conductivity using micro-Raman thermometry. (a) Typical Raman spectrum of thin-film tellurium. (b) A$_1$ mode Raman shift vs. incident laser power. (c) Temperature distribution across the trench with 10 µW laser heating, from the numerical simulation. The inset cartoon: schematics of suspended Te thin film heated at the center.
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