Solution-Synthesized High-Mobility Tellurium Nanoflakes for Short-Wave Infrared Photodetectors

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26 **Abstract**

- 27 Two-dimensional (2D) materials, particularly black phosphorous (bP), have
- 28 demonstrated themselves to be excellent candidates for high-performance
- 29 infrared photodetectors and transistors. However, high-quality bP can be
- 30 only obtained via mechanical exfoliation from high temperature- and high-
- 31 pressure-grown bulk crystals and degrades rapidly when exposed to ambient
- 32 conditions. Here, we report solution-synthesized and air-stable quasi-2D
- tellurium (Te) nanoflakes for short-wave infrared (SWIR) photodetectors. We

perform comprehensive optical characterization via polarization-resolved transmission and reflection measurements, and report the absorbance and complex refractive index of Te crystals. It is found that this material is an indirect semiconductor with a bandgap of 0.31 eV. From temperaturedependent electrical measurements, we confirm this bandgap value and find that 12 nm thick Te nanoflake show high hole mobilities of 450 and 1430 cm² V⁻¹ s⁻¹ at 300K and 77K, respectively. Finally, we demonstrate that despite its indirect bandgap, Te can be utilized for high-performance SWIR photodetectors by employing optical cavity substrates consisting of Au/Al₂O₃ to dramatically increase the absorption in the semiconductor. By changing thickness of the Al₂O₃ cavity, the peak responsivity of Te photoconductors can be tuned from 1.4 µm (13 A/W) to 2.4 µm (8 A/W) with a cut-off wavelength of 3.4 µm, fully capturing the SWIR band. An optimized room temperature specific detectivity (D^*) of 2×10^9 cm Hz^{1/2} W⁻¹ is obtained at a wavelength of 1.7 µm.

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Narrow bandgap semiconductors, with bandgaps in the range of 0.7 to 0.3 eV, have been heavily investigated for numerous applications. These include high-speed and high-performance transistors, which typically require materials with high mobility and low effective mass. 1,2 Additionally, photodetectors and light sources which can operate in the short-wave infrared (SWIR, 1.4 μm – 3 μm) band are heavily utilized for imaging and optical communication. $^{3-5}$ This area has typically been dominated by III-V semiconductors such as indium gallium arsenide (InGaAs) and indium

gallium arsenide phosphide (InGaAsP), which have excellent performance but require complex growth and fabrication procedures.^{6,7} Numerous research groups have investigated techniques to achieve III-V semiconductors through non-epitaxial growth on silicon or by layer transfer.⁸⁻¹¹ However, these techniques still introduce significant complexity and may require high growth temperature. Moreover, key limitations still exist in InGaAs-based devices; an example being edge-recombination in scaled InGaAs photodiodes which limits pixel sizes in focal plane arrays to dimensions of 5-6 µm, well above the diffraction limit.¹² As such, there is a pressing need to find suitable materials that can either improve the performance and/or reduce the costs of these devices.

One such potential material system is two-dimensional (2D) materials, which have been heavily studied over the past decade. One of the key advantages of 2D materials, which enables their high performance, is their naturally terminated surfaces; *i.e.* since the out-of-plane bonds in these materials are van der Waals rather than covalent. As such, they do not suffer from the surface-induced performance degradation commonly observed in three-dimensional (3D) semiconductors, which require surface passivation (typically in the form of cladding layers). This has enabled extraordinary electronic and optical properties of these materials such as photoluminescence quantum yields approaching unity and their exceptional thickness scalability down to the monolayer limit. Devices which have exploited the advantages and physics of 2D systems have demonstrated dramatic scaling as well as new devices paradigms. However, edges in 2D

layers still act as recombination centers and are analogous to a surface in a 3D crystal structure.²⁰ A recent work by Wang *et al.* demonstrated the solution synthesis of air-stable quasi-2D Te nanosheets and its high-performance for field-effect transistors.²¹ Note that Te is a true one-dimensional (1D) system, which can potentially overcome some of these limitations since it consists of van der Waals bonded molecular chains.²² As such these material systems naturally possess no dangling bonds when scaled, with the exception of the ends of the molecular chains.²² Importantly, this material also possesses a small, thickness tunable bandgap enabling its potential use in SWIR photodetectors. Recent theoretical works have predicated an indirect gap of 0.35 eV in bulk and a direct gap of 1.04 eV in the monolayer case.^{23,24}

Here, we systematically study the optical and electrical properties of solution-synthesized quasi-2D Te nanoflakes. From polarization-resolved IR transmission and reflection measurements, we experimentally extract the bandgap, absorbance, and complex refractive index of this material. Temperature-dependent electrical measurements were performed to verify the optically measured bandgap values as well as determine the carrier transport properties as a function of temperature in quasi-2D Te nanoflakes. Finally, we demonstrate SWIR photoconductive detectors based on thin (16-20 nm) quasi-2D Te nanoflakes. Although this material has low absorbance in the SWIR (6×10^5 cm⁻¹ for wavelengths in the range of 2 to 3.5 μ m) induced by the indirect bandgap, the Te nanoflake-based photodetectors exhibit high photoresponsivity and specific detectivity by leveraging optical cavities

engineered to maximize absorption at various technologically important wavelengths in the SWIR range.

RESULTS and DISCUSSION

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Material Synthesis and Characterization.

Quasi-2D Te nanoflakes with varying thickness in the range of 10-200 nm were synthesized by a hydrothermal method via reduction of sodium tellurite with hydrazine hydrate in the presence of poly(-vinyl pyrrolidone) in an aqueous alkaline solution at 180°C for certain varying times (details are provided in methods).^{21,25} The crystal structure of Te is shown in Figure 1a-c. The Te crystallizes in a structure composed of Te atomic chains in a triangular helix that are stacked together via van der Waals forces in a hexagonal array (Figure 1a). In this structure, Te atoms only form covalent bonds to the two nearest neighbor Te atoms in the helical chain (Figure 1b). Therefore, Te is a true 1D system rather than a 2D van der Waals crystal. When it is viewed from x-axis, the zig-zag layers are seen to be stacked together via van der Waals forces to form a 3D structure (Figure 1c). Figure 1d shows the transmission electron microscope (TEM) image of a typical Te nanoflake with a length of 15 µm and width of 4 µm. The corresponding highresolution TEM (HR-TEM) image shows the continuous crystal lattice of the Te nanoflake and the measured the lattice constant is ~ 2 Å (Figure 1e), which is assignable to the (003) planes of the Te crystal. The corresponding selected area electron diffraction (SAED) pattern of the Te nanoflake shows bright diffraction spots with a rectangular shape, in which the nearest two spots to

the diffraction center correspond to the (110) and (001) planes of the Te crystal (Figure 1f). Both the HR-TEM and SAED pattern results indicate that the solution-synthesized Te nanoflake is single-crystalline. The optical image shows that Te nanoflakes have an irregular shape with length of tens of micrometers, width of few micrometers and thickness of 10-30 nm (Figure 2a). Note that there are always some Te nanowires along Te nanoflakes in the growth solution, but Te nanowires can be partially removed from the solution by proper centrifugation (see the details in Methods). An atomic force microscope (AFM) image of a Te nanoflake with a thickness of 16.1 nm is shown in Figure 2b. Raman spectrum of tellurium nanoflakes gives three vibrational modes at 92, 121, 141 cm⁻¹ (Figure 2c) corresponding to the E₁-TO, A₁, and E₂ peaks, which are consistent with previous literature reports.^{21,26} The X-ray diffraction (XRD) peaks of Te nanoflakes match well with the simulated reference (Figure 2d). Both Raman and XRD analysis further confirm the successful preparation of Te nanoflakes. 21,25

Electrical Properties.

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We now turn our attention to the electrical properties and performance of the quasi-2D Te nanoflakes. To this end, back-gated field-effect transistors were fabricated on Si/50 nm SiO₂ by patterning Pd (40 nm thick) electrodes on Te nanoflakes with thickness ranging from 10-20 nm using electron-beam lithography.²¹ The $I_{\rm d}$ - $V_{\rm d}$ characteristics of a 12.3 nm thick quasi-2D Te field-effect transistor, as well as an optical image of the measured device (L=6.8 µm, W=1.5 µm, where L and W are length and width respectively) are shown in Figure 3a. The device shows dominantly p-type transport

characteristics and a peak current density of 116 $\mu A/\mu m$ at a V_d = -1 V and V_g 153 = -20 V. It should be noted that in some devices (Figure S1) ambipolar 154 155 transport characteristics are observed, indicating that via proper contact engineering n-type transport could be obtained. Additionally, this device 156 shows a current on/off ratio of $\sim 3 \times 10^3$, which is expected for a material with 157 a small bulk bandgap of ~ 0.31 eV. The output characteristics of this device 158 are shown in Figure 3b, and show current saturation at high drain bias, as 159 well as linear behavior at low drain biases, indicating low contact resistance. 160 We then utilize the square law model to calculate the effective mobility (μ_{Eff}) 161 in this device as a function of gate field (Figure 3c) using $\mu_{Eff} = (dI_d/dV_d)/\dot{\epsilon}$, 162 where C_{ox} is the gate capacitance (69.1 nF cm⁻² in the case of 50 nm SiO₂) 163 and V_t is the threshold voltage. We extract a high peak μ_{Eff} of 619 cm²V⁻¹s⁻¹ 164 for holes, which is very competitive with high-performance p-type 165 semiconductors with similar bandgaps such as InGaSb and bP, especially 166 considering that the material is prepared by a solution-based method. 27-29 167 The thickness-dependent transport properties of quasi-2D Te nanoflakes 168 were also investigated over a range of 11 to 36 nm as shown in Figure 3d 169 and 3e. As the overall thickness of the Te nanoflakes is reduced a dramatic 170 improvement in the ratio of the on/off currents from $\sim 30 \times$ to $\sim 4000 \times$ can be 171 172 observed, which can be attributed to reduced gating efficiency in the device. Furthermore, over this measured thickness range, we find that there is 173 negligible variation in the peak effective mobility which has an average value 174 of 424±74 cm²V⁻¹s⁻¹ over thirty-nine measured samples. 175

To better understand the transport in this material, we performed temperature-dependent electrical characterization of the devices. I_d - V_q characteristics of quasi-2D Te nanoflakes with different thicknesses measured over a temperature range of 77K to 350K are shown in Figure 4a and Figure S1. A clear increase in the current on/off ratio can be observed when the device is cooled, which is consistent with small bandgap materials, where thermal generation dramatically increases the carrier densities at elevated temperatures. From the temperature-dependent measurements, we extract the μ_{Eff} at a gate overdrive of -10 V as a function of temperature for different layer thicknesses as shown in Figure 4b. For all thicknesses, the mobility increases as the temperature is reduced, with the 12 nm thick device showing a μ_{Eff} of 1430 and 450 cm²V⁻¹s⁻¹ at 77K and 300K respectively. For all three thicknesses, we observe the temperature dependence of mobility can be fitted with a power law, $\mu_{\rm Eff} \propto T^{-\gamma}$, where $\gamma = -1.03$ in our case. This indicates that the mobility in this material is limited by phonon scattering and not ionized impurities.³⁰ Additionally, we estimate the bandgap using temperature-dependent measurements on a device prepared on a 260 nm gate oxide. In this case, the minimum drain currents, $I_{d,min}$, is determined by thermal activation of carriers over the contact Schottky barrier height.³¹ The resulting data, shown in Figure 4c, can be fit using:

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where T is the temperature, E_g is the transport bandgap, and k is the Boltzmann constant. We extract an E_g of 280 meV for a 11.8 nm thick

sample, and it is important to note that this method is expected to underestimate the bandgap due to contributions from trap states.

Optical Properties.

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To identify the optical bandgap of the guasi-2D Te nanoflakes, we performed polarization-resolved transmission and reflection measurements on Te nanoflakes which were transferred onto KBr. Polarization-resolved transmission and reflection spectra measured on a 111 nm thick sample are shown in Figure 5a. In order to accurately extract the absorption in the sample, the optical path was kept constant for both the transmission and reflection measurements, as annotated in the Figure 5a. Transmission and reflection spectra were taken in reference to the blank KBr substrate and an Au mirror, respectively. The total absorption and the corresponding absorption coefficient in the material can then be calculated as 100%-Reflection-Transmission as shown in Figure 5b and 5c. For light polarized along the direction of the Te molecular wires (defined as 90°), we measure a strong absorption that can be fitted to a direct bandgap (Figure 5c and Figure S2) at 0.71 eV. However, when the light is polarized perpendicular to the axis of the Te molecular wires (defined as 0°) a significantly weaker absorption edge is observed at lower wavelengths. From similar analysis, the transition can be fitted to an indirect bandgap at 0.31 eV. Importantly, while the absorption coefficient for wavelengths below 1.6 μ m is high (4.5×10⁶ cm⁻¹ 1), the absorption for longer wavelengths (1.6 μ m < λ < 3.4 μ m) is an order of magnitude lower $(6 \times 10^5 \text{ cm}^{-1})$. As such, in order to effectively utilize this material in the full SWIR band, it is necessary to increase the absorption *via* proper optical engineering.³²

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To this end, we extracted the complex refractive index (n, k) which can be used to estimate the total absorption in the guasi-2D Te layers in combination with optical cavities, back-reflectors, or anti-reflection coatings. The refractive index was extracted using a previously established technique for nanoscale materials for unpolarized light, where reflection is measured as a function of oxide or sample thickness.³³ In our case, quasi-2D Te nanoflakes of thicknesses varying from 16 nm to 180 nm were drop-casted on Au substrates, and the reflection spectra were measured using an Fourier transform infrared spectrometer (FTIR). The resulting reflection data are plotted as a function of thickness and wavelength. (Figure 6a and Figure S3). To extract the complex refractive index from the reflection measurements, we utilize the transfer matrix method to fit a refractive index to the measured reflection versus thickness at each wavelength.³⁴ The extracted n and k values are plotted in Figure 6b; the extracted extinction coefficient clearly shows absorption resulting from both the indirect and direct transitions at the expected wavelengths and corroborates the results from absorption measurements shown in Figure 5c.

Optical Cavity Enhanced Gated-Photoconductors.

Based on the optical properties and electrical properties of quasi-2D Te, the device structure shown in Figure 7a was chosen to optimize photoconductive detectors targeting the SWIR spectrum. Here, we selected thicknesses in the range of 16-20 nm for the quasi-2D Te layers, which

provides desired electrical characteristics, the most important of which being a low dark current. To increase the absorption, an optical cavity consisting of an optically thick Au film (100 nm) and an Al_2O_3 dielectric spacer layer was utilized. The predicted absorption for an 18 nm thick Te layer as a function of wavelength and Al_2O_3 thickness was calculated using the transfer matrix method and is shown in Figure 7b. From these simulations, it can be clearly seen that by adjusting the thickness of the Al_2O_3 layer, the peak absorption wavelength can be tuned over a large range and the total absorption in the material can be increased by over two orders of magnitude relative to a sample on Si/SiO_2 (Figure S4). Additionally, the optical cavity simultaneously acts as the gate stack in this structure.

Figure 7c shows optical images of devices fabricated on three different Al_2O_3 spacer thickness (550 nm (i), 150 nm (ii), and 350 nm (iii)), which are selected to provide optimized absorption at 1.4 µm, 1.8 µm, and 2.2 µm, respectively (reflection spectra of devices on these substrates are shown in Figure S5). The spectral responsivity of these devices to SWIR illumination was characterized using an FTIR, by placing the devices in place of the internal detector and focusing the light source of the FTIR on the device with a CaF_2 lens. To calibrate the measurements, the internal Deuterated Triglycine Sulfate (DTGS) in the FTIR, which has a wavelength independent responsivity, was used to determine the relative intensity of the light source while a NIST traceable Ge photodiode was used to determine the absolute intensity of the illumination (further measurement details are provided in the methods and ref. 35). The final responsivity (R) is then calculated as $R(\lambda)$ =

 $I_{ph}(\lambda)/P_{in}(\lambda)$, where I_{ph} is the photocurrent and P_{in} is the incident optical power on the device. The spectral responsivity for representative devices fabricated on the three different optical cavity substrates are shown in Figure 7d. Devices fabricated on these three substrates were measured at a drain bias of $V_d = 5$ V, and the gate voltage was adjusted to maximize the responsivity. The measured responsivity curves closely match the calculated absorption curves which further confirms the estimated refractive index values.

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Gate-dependent Photoresponse, Detectivity, and Frequency Response.

To further characterize the performance of these photodetectors, additional measurements were performed on an 18.8 nm thick device fabricated on a Au/150 nm Al_2O_3 substrate. The I_d - V_g characteristics of a device measured at 78K and 297K without the presence of a cold shield is shown in Figure 8a. The gate and drain bias dependence of the device at an incident wavelength of 1.7 µm are shown in Figure 8b and 8c, respectively. We can see that the photoresponse can be strongly modulated by the gate bias and is maximized when the device is biased such that drain current is minimized, similar to what has previously been observed in bP photoconductors^{35,36}. This can be seen in the clear peak responsivity of 27 A/ W at 78K, while the responsivity at 297K saturates with increasing gate biases to a peak measured value of 16 A/W. Additionally, the device shows an approximately linearly increasing responsivity at low V_d , and begins to show saturation at high V_d . The full spectral responsivity of the device is shown in Figure 8d. For $V_d=5$ V, this detector shows a peak responsivity at λ = 1.7 μ m of 27 A/W and 16 A/W at 78K and 297K, respectively.

To calculate the specific detectivity of this device, the noise current density is measured under bias conditions which give maximum responsivity at an integration time (Δf) of one second and is shown in Figure 8e. The noise currents clearly show the expected 1/f (where f is frequency) flicker noise which is typically attributed to charge trapping/de-trapping. Utilizing the noise and responsivity measurements, we calculate the specific detectivity (D^*) using:

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$$D^{i} = \frac{\sqrt{A\Delta f}}{NEP} = \frac{R\sqrt{A\Delta f}}{i_{p}}$$

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303 where A is the detector area, NEP is noise equivalent power, and i_n is the noise current. In the ideal case for photoconductors, the i_n is limited by shot 304 noise and can be calculated from dark current using: $\langle i_s^2 \rangle = 2 q I_{G-R} \Delta f$. This 305 typically overestimates noise current; thus, we utilized the experimental 306 noise currents taken at a modulation frequency of 1 kHz, which was selected 307 to minimize the contribution of 1/f noise. The specific detectivity as a 308 function of wavelength is shown in Figure 8f for both 78K and 297K. At room 309 temperature these devices show a peak D^* of 2.9×10^9 cmHz^{1/2}W⁻¹. This value 310 dramatically increases at low temperatures due to the large decrease in the 311 noise current as well as the improved responsivity to a peak value of 312 $2.6 \times 10^{11} \text{ cmHz}^{1/2}\text{W}^{-1}$. 313

To characterize the speed of this device, a directly modulated laser diode ($\lambda = 1.55~\mu m$) was utilized to excite the photoconductor. The response

of the device to a low-frequency and high-frequency square wave are shown in Figure 9a and 9b, respectively. From both of the step responses shown in Figure 9a and 9b, a slow and fast component of the rise/fall time can be observed. To more clearly characterize this behavior, the photoresponse was measured with a sinusoidally modulated incident light over a wide frequency range (100 mHz to 100 kHz) as shown in Figure 9c. Over this measured spectral range, a clear 3dB cutoff wavelength cannot be observed, indicating that it is higher than the measured range. However, the device shows a weak increase in response for light modulated at low frequencies; i.e. the photoresponse at a 100 kHz shows only a ~5× drop relative to the low frequency (~0.1 Hz) photoresponse. It is likely that this behavior is a result of photoconductive gain present in the device which consistent with the high responsivity and the short carrier transit time (τ_{tr}), which is calculated to be 0.4 ns according to $\tau_{tr} = L^2/(\mu V_d)$, where L is channel length (8.9 μ m) and μ in this case is 392 cm²V⁻¹s⁻¹. The exact mechanisms of the gain likely is a result of the population and depopulation of trap states, which show saturation at high illumination powers.³⁹⁻⁴¹ This is particularly likely given the fact that the Te nanoflakes in this work were prepared using solution-based synthesis.

Polarization-Resolved Photoresponse.

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Finally, we investigated the polarization-dependent photoresponse of Te nanoflake-based photodetectors. We chose a device fabricated on a 350 nm thick Al_2O_3 substrate as an example to measure the responsivity, which provides strong absorption enhancement at wavelengths covering indirect gap band edge. Figure 10a shows the responsivity of an 18.8 nm thick Te

photoconductor measured at various incident polarization angles, with 0° and 90° corresponding to light polarized perpendicular and parallel to the molecular wires, respectively. A clear drop in the responsivity for wavelengths greater 1.5 μ m can be observed when the incident light is polarized at 90°, consistent with the indirect band edge. A polar plot showing the responsivity as a function of polarization at wavelengths of 3 μ m and 1.5 μ m is shown in Figure 10b, and clearly shows the highly anisotropic responsivity of the indirect gap, while the direct gap shows minimal dependence on the polarization of the light.

CONCLUSION

In summary, we have performed a systematic study on the electrical and optical properties of solution-synthesized quasi-2D Te, an indirect, small bandgap (0.31 eV) semiconductor. The Te nanoflake-transistor shows high effective hole mobilities of 450 and 1430 cm² V¹ s¹ at 300K and 77K respectively. The absorbance and complex refractive index of Te were also extracted based on the polarization-resolved transmission and reflection measurements. Furthermore, the Te gated photodetectors exhibit high responsivities over the entire SWIR range with a cut-off wavelength of 3.4 μm . Importantly, using the extracted optical parameters, we dramatically enhanced the responsivity and specific detectivity of Te photodetectors using optical cavity substrates. We show that the peak wavelength can be tuned from 1.4 to 2.4 μm by changing the thickness of the Al₂O₃ cavity on Au. This simple substrate engineering technique can be a general way to significantly enhance the performance of other 2D material-based

photodetectors. In addition, due to the anisotropic crystal structure of the Te nanoflakes, the devices possess a polarization-sensitive SWIR photoresponse. Our electrical and optoelectronic study indicate that Te is a promising material for high-performance and low-cost electronic and optoelectronic devices. Finally, this material has a high potential for wafer-scale production either through the assembly of solution-processed nanoflakes to form semi-continuous thin films or through techniques such as chemical vapor deposition or epitaxial growth.

METHODS

Chemicals

Sodium tellurite (Na₂TeO₃), hydrazine monohydrate (N₂H₄•H₂0) and ammonium hydroxide solution (NH₃•H₂0, 28%) were purchased from Sigma-Aldrich (USA). Poly(-vinyl pyrrolidone) (PVP, average M.W. 58,000) was purchased from Alfa Aesar (USA). All the chemicals were used as received without further purification. Milli-Q water (Milli-Q System, Millipore) was used in all experiments.

Synthesis of tellurium nanoflakes

Tellurium nanoflakes were synthesized by a hydrothermal method according to a previously reported procedure with slight modifications. 21,25 In a typical procedure, 1.5 g of PVP was first dissolved in 16 mL DI water and 46 mg of Na₂TeO₃ was then added and dissolved into the PVP solution to form a clear solution. Then, 1.66 mL of ammonium hydroxide solution and 0.838 mL of hydrazine monohydrate was added into the above solution in sequence. The solution was then transferred into a 25 mL Teflon-lined stainless steel

autoclave. The autoclave was sealed well and then placed in an oven. The autoclave was heated up to 180°C from room temperature with ramp rate of 3 °C/min and then maintained at 180°C for 4 hours. After, the autoclave was removed from the oven and cooled to room temperature by running water immediately. The resulting product was purified and washed with DI water three times by centrifugation at 3000 r.p.m for 2 min. After washing and purification, the final tellurium solution is a silver-gray color. Note that there are always some Te nanowires coexisting along with Te nanoflakes and it is challenging to fully separate the nanowires from the nanoflakes. The final product was redispersed in pure ethanol before being transferred to the target substrate for characterization and device fabrication by drop-casting. Maintaining the autoclave in oven at 180°C for 4h will produce the tellurium nanoflakes with a typical thickness in the range of 10-30 nm, with typical lateral dimensions of 10-50 µm. It is important to point out that the thickness of tellurium nanoflakes can be roughly controlled by controlling the reaction time (4-30 hours) at 180°C. The thickness of tellurium nanoflakes can be increased by prolonging the reaction.

Device Fabrication

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Te nanoflakes were transferred onto p⁺⁺ Si/SiO₂ chips with a 50 nm oxide or back reflector Al_2O_3/Au chips with varying thicknesses of Al_2O_3 by drop-casting the tellurium ethanol solution. Back reflector substrates were prepared by evaporating Ti/Au/Ti (10 nm/200 nm/1 nm) on Si substrates, followed by atomic layer deposition of Al_2O_3 (Cambridge Nanotech). The final Al_2O_3 thickness was determined by ellipsometry. Suitable Te nanoflakes were

then located on an optical microscope and devices were fabricated using standard e-beam lithography techniques. After development, 40 nm thick Pd was evaporated as electrode contacts *via* e-beam evaporation and lift-off was performed using acetone.

Material Characterization

Transmission electron microscopy (TEM) images were performed at the National Center for Electron Microscopy at Laurence Berkeley National Laboratory using a FEI Titan 60-300 microscope operated at 200kV. Tapping mode atomic force microscope (AFM) measurements were conducted on a Dimension ICON AFM (Bruker, Germany) under ambient conditions. A AXS D8 Discover GADDS X-ray diffractometer (Bruker, Germany) equipped with a Vantec-500 area detector and a Co K α (λ =1.7903 Å) X-ray source was used to measure the powder X-ray diffraction (XRD) pattern. The Raman spectrum was measured on Horiba Jobin Yvon LabRAM ARAMIS automated scanning confocal Raman microscope system with the excitation line of 532 nm (Horiba, Ltd. Japan). The Raman band of a silicon wafer at 520 cm⁻¹ was used as the reference to calibrate the spectrometer.

Optical/Electrical Characterization

Samples for absorption/reflection measurements were transferred onto KBr substrates using a previously established dry transfer technique and were measured in an FTIR microscope (ThermoFisher).³⁵ Electrical measurements were performed in a cryogenic protestation (LakeShore) with a B1500a Semiconductor Parameter Analyzer (Agilent). Spectral photoresponse measurements were performed in a FTIR spectrometer (ThermoFisher) with a

customized Janis cryostat equipped with a CaF₂ window. Devices were wire bonded in a 24 pin chip carrier and evacuated to a base pressure of $< 10^{-5}$ Torr. The cryostat was then placed at the auxiliary exit port of the FTIR, and the exciting illumination from the tungsten lamp source was focused on the sample using either a CaF₂ lens or a 15× Schwarzschild objective. The optical velocity of the FITR was set to either 0.1515 or 0.4747 cm s⁻¹; the resulting modulation frequency range (7.5 kHz to 300 Hz) is within the operating regime of the frequency response. The photocurrent from the Te devices was then sent to a current amplifier (Stanford Research Systems), which also provided the bias voltage. The resulting signal was subsequently returned to the external detector interface of the FTIR. The relative intensity of the illumination source was measured using the internal Deuterated Triglycine Sulfate (DTGS) detector of the FTIR with identical measurement conditions. The photoresponse was calibrated using a NIST traceable Ge photodiode (ThorLabs) with a 100 µm aperture placed in the same position as the device under measurement. The frequency response of the devices was measured using a directly modulated 1550 nm laser diode. Further details and a schematic of the measurement setup are provided in ref. 35.

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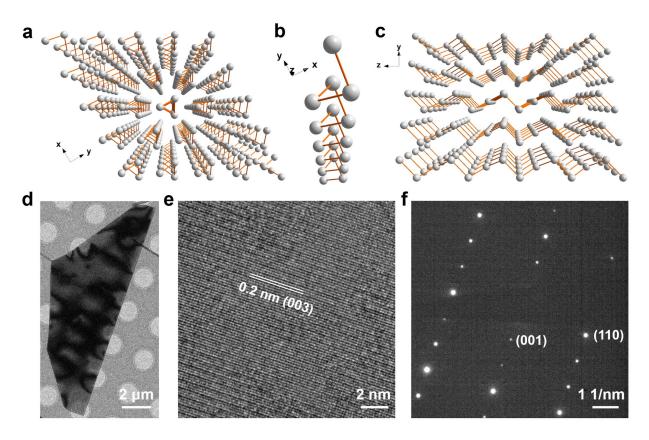


Figure 1. Crystal structure of tellurium: (a) viewed from x-axis, (b) single-molecular chain and (c) viewed from z-axis. (d) TEM image, (e) corresponding HR-TEM image and (f) SAED pattern of a typical tellurium nanoflake.

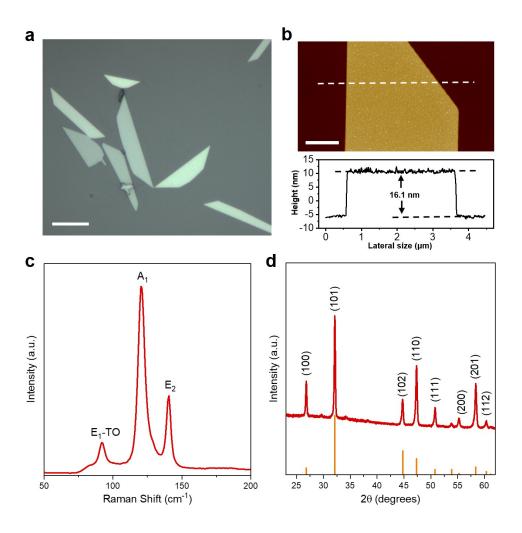


Figure 2. (a) Optical microscope image of tellurium nanoflakes deposited on a SiO_2/Si substrate, scale bar is 10 μ m. (b) AFM image of typical tellurium nanoflake (top) and the corresponding height profile (bottom), scale bar is 1 μ m. (c) Raman spectrum of tellurium nanoflakes. (d) XRD pattern of dried tellurium nanoflake powder.

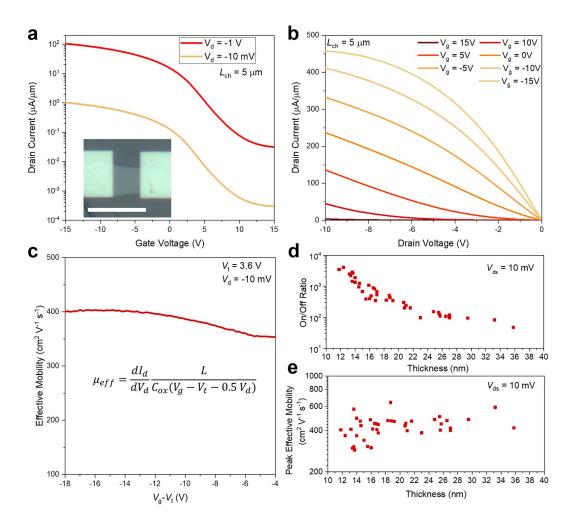


Figure 3. (a) Back-gated I_d - V_g characteristics of a 12.3 nm thick Te nanoflake with Pd contacts on 50 nm SiO₂ measured at room temperature; inset shows optical image of the device, scale bar is 10 μ m. (b) I_d - V_d measured for the same device as shown in panel (a). (c) Effective mobility as a function of gate overdrive voltage for the device shown in panel (a) under a V_d of -10 mV, giving a peak mobility value of 619 cm⁻² V⁻¹ s⁻¹. (d) Thickness dependence of the on/off ratio measured for Te nanoflakes of varying thickness in the range of 12-36 nm. (e) Thickness dependence of the peak effective mobility measured for Te nanoflakes of varying thickness in the range of 12-36 nm.

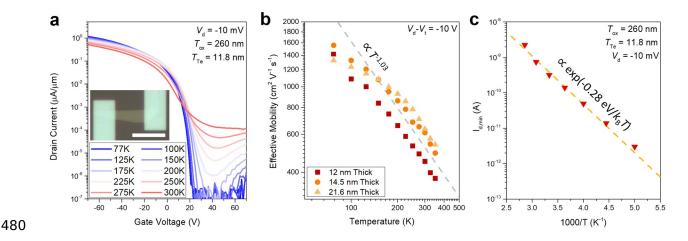


Figure 4. (a) Temperature dependent I_{d} - V_{g} characteristics of a 12 nm thick Te nanoflake, measured at a low V_{d} of -10 mV. Scale bar is 5 μ m. (b) Effective mobility for quasi-2D Te nanoflakes with varying thickness as a function of temperature extracted from panel (a) and Figure S1 at a constant gate overdrive voltage of -10 V, dashed line shows a power law fit. (c) Arrhenius plot showing the minimum drain source current ($I_{d,min}$) as a function of temperature extracted from the data shown in panel (a).

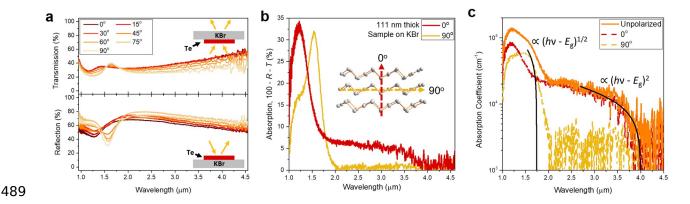


Figure 5. (a) Polarization resolved transmission and reflection spectra measured for a 111 nm thick Te nanoflake transferred on a KBr substrate, insets show the optical configuration used during measurements. (b) Absorption spectra for the nanoflake measured in panel (a), calculated from 100%-*T-R*. Inset shows the Te crystal directions of 0° and 90°. (c) Absorption coefficient of tellurium for unpolarized light, as well as light aligned parallel to z-axis (0°) and y-axis (90°) of Te crystal; Tauc plots and optical bandgap extraction for light polarized perpendicular and parallel to the wire axis are shown in Figure S2.

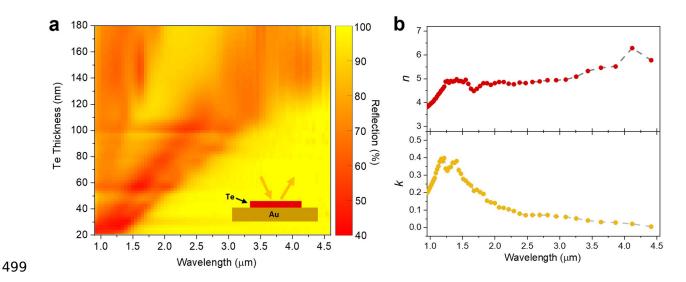


Figure 6. (a) Reflection of quasi-2D Te nanoflakes on Au substrates, with Te thickness ranging from 20 nm to 180 nm. Plot is obtained from interpolation of spectra taken on 18 Te crystal nanoflakes of varying thicknesses, shown in Figure S3. (b) Complex refractive index of quasi-2D Te nanoflakes calculated by fitting measured reflection data as a function of thickness.

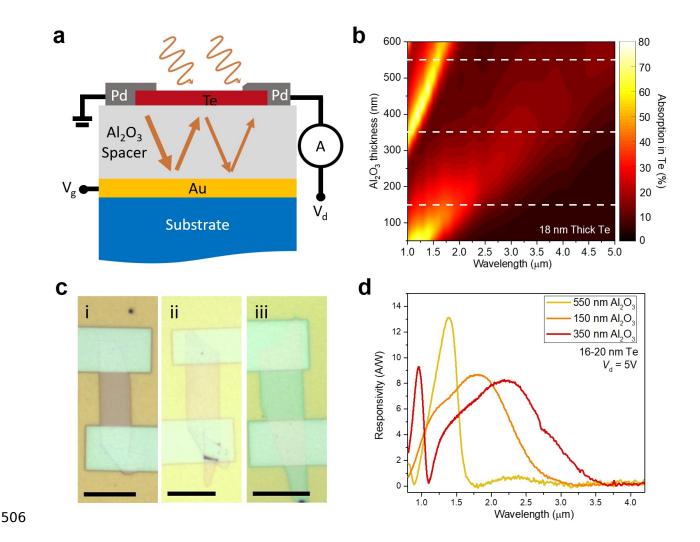


Figure 7. (a) Schematic showing the optical cavity structure used to fabricate SWIR photoconductors from quasi-2D Te nanoflakes in this study. An evaporated Au film acts as the gate electrode and back-reflector, while a Al_2O_3 dielectric layer acts as a $\sim \lambda/4$ spacer as well as the gate oxide. (b) Calculated absorption in the Te layer, as a function of wavelength and Al_2O_3 thickness for a fixed semiconductor thickness of 18 nm. (c) Optical images of SWIR photoconductors fabricated on optical cavities with Al_2O_3 thickness of 550 nm (i), 150 nm (ii), and 350 nm (iii); scale bar is 10 μ m. (d) Measured

- responsivities of quasi-2D Te nanoflakes fabricated on optical cavities with
- 516 different Al_2O_3 thicknesses.

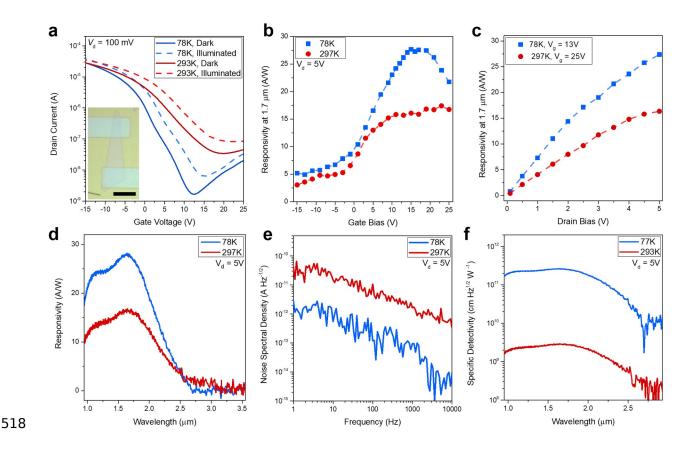


Figure 8. (a) $I_{\rm d}$ - $V_{\rm g}$ characteristics of an 18.8 nm Te photoconductor measured at 297K and 78K under illumination and in the dark. Inset shows optical image of the measured device; scale bar is 10 μ m. (b) Responsivity at 1.7 μ m measured at 297K and 78K as a function of gate bias, at a constant drain bias of $V_{\rm d}=5$ V. (c) Responsivity at 1.7 μ m measured at 297K and 78K as a function of drain bias at an optimized gate bias ($V_{\rm g}=13$ V at 78K and $V_{\rm g}=25$ V at 297K). (d) Spectral responsivity per watt of a Te photoconductor measured at 78K and 297K under optimized gate bias and $V_{\rm d}=5$ V. (e) Noise spectrum measured at room temperature for a Te photoconductor operated with optimized gate bias and a $V_{\rm d}=5$ V and optimized gate bias. (f) Specific detectivity of Te photoconductors with optimized thickness; measurements were performed under gating conditions which maximized detectivity for the

specific device at room temperature. Note that an Au/Al_2O_3 substrate with an Al_2O_3 thickness of 150 nm was used for the measurements.



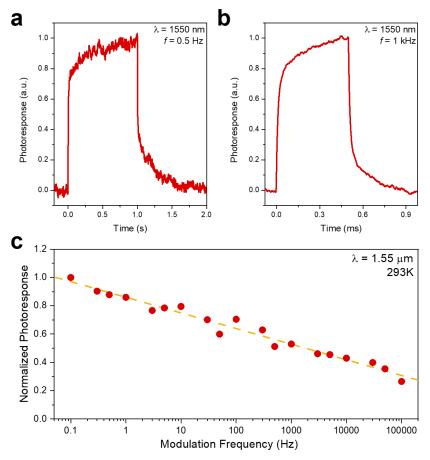


Figure 9. (a) Room-temperature step response of an 18.8nm thick Te photoconductor, excited by a 1550-nm laser modulated by a 0.5 Hz square wave. (b) Room-temperature step response of a Te photoconductor, excited by a 1550 nm laser modulated by a 1 kHz square wave. (c) Normalized photoresponse of a Te photoconductor *versus* modulation frequency; the device is excited using a sinusoidally modulated 1550 nm laser. Note that an Au/Al_2O_3 substrate with an Al_2O_3 thickness of 150 nm was used for the measurements.

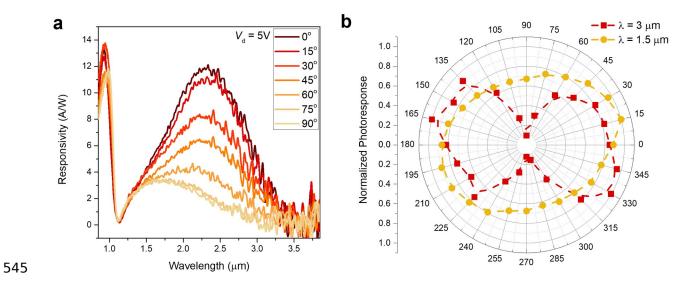
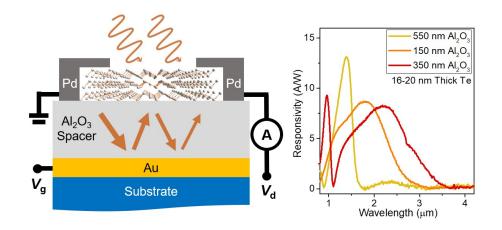


Figure 10. (a) Polarization resolved photoresponse of an 18.8 nm thick quasi-2D Te nanoflake fabricated on an optical cavity with a 350 nm thick Al_2O_3 . (b) Polar plot showing the relative response of the device shown in panel (a) at wavelengths of 3 μ m and 1.5 μ m as a function of polarization.

TOC Graphic



554 ASSOCIATED CONTENT

Supporting Information

- 556 The Supporting Information is available free of charge on the ACS
- 557 Publications website.
- Temperature dependent electrical characterization of additional
- devices. Tauc plot used to extract direct and indirect bandgap of Te.
- Reflection spectra used to create plot in Fig. 6a. Photoresponse
- measurements for devices prepared on Si/SiO₂ substrates. Reflection
- spectra taken on Te photoconductors.

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