Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C. This journal is © The Royal Society of Chemistry 2021

Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C. This journal is © The Royal Society of Chemistry 2021

## **Electronic Supplementary Information**

## High-quality two-dimensional tellurium flakes grown by

## high-temperature vapor deposition

Baoquan Huang,<sup>a</sup> Zehong Lei, <sup>a</sup> XuanHao Cao,<sup>a</sup> Aixiang Wei,<sup>a</sup> Lili Tao,<sup>a</sup> Yibin Yang,<sup>a</sup> Jun Liu,<sup>a</sup> Zhaoqiang Zheng<sup>a</sup> and Yu Zhao\*<sup>a</sup>

Guangdong Provincial Key Laboratory of Information Photonics Technology, Guangdong Provincial Key Laboratory of Functional Soft Condensed Matter, School of Material and Energy, Guangdong University of Technology, Guangzhou 510006, People's Republic of China

\*E-mail: zhaoyu@gdut.edu.cn



**Fig. S1.** AFM of the Te flake with a thickness of 1.4 nm, corresponding to three layer thick.



**Fig. S2.** Angle-resolved Raman spectra. (a)-(b) The scattering polarization direction is parallel to the incident polarization direction. (c)-(d) The scattering polarization direction is perpendicular to the incident polarization direction.

Theoretically, 2D Te crystal belongs to  $D_3^4$  space group and the anisotropic Raman intensity can be fitted by Raman tensor [S1]

$$R(A_{1}) = \begin{bmatrix} a & 0 & 0 \\ 0 & b & 0 \\ 0 & 0 & c \end{bmatrix}$$
(1)  
$$R(E_{2}) = \begin{bmatrix} 0 & d & e \\ d & 0 & 0 \\ e & 0 & 0 \end{bmatrix}$$
(2)

where a, b, c, d, and e are Raman tensor elements. The polarized Raman

intensity can be fitted as follows:

$$I(A_1), \| = |asin\theta^2 + ccos^2\theta|^2$$
(3)

$$I(E_2), \| = |2esin\theta cos\theta|^2$$
(4)

where  $\theta$  is the angle between the longer side of the crystal and the incident light polarization direction.



**Fig. S3.** XPS of (a) all elements, (b) tellurium element and (c) indium element. The XPS measurement is performed after an etching on Te surface for 360 s.



**Fig. S4.** The time-resolved photoresponse, power-dependent responsivity and detectivity curves under 532, 635 and 808 nm laser illumination, respectively.



**Fig. S5.** The photocurrent of Te flake FETs in response to a pulse of laser illumination under the ambient and vacuum condition. (a) Illumination of 405 nm laser. (b) Illumination of 532 nm laser.



Fig. S6. XPS measurement of Te flakes with and without 320 s etching.



Fig. S7. The energy alignment of the band structure between the Te and superficial  $TeO_2$ .

we propose a possible mechanism based on the Te/TeO<sub>2</sub> heterinterface to explain the unique photoresponse feature. By taking the electron affinity of Te from reference [S2] and bandgap value of 2D Te from reference [S3], as well as the experimental band parameters of 2D TeO<sub>2</sub> from reference [S4], we construct the energy alignment of the band structure between the Te and superficial TeO<sub>2</sub>, as shown in Fig. S7. As the Te has been widely reported as highly photo-responsive material, we believe the photo-induced carrier is primarily produced from Te flakes. When the excited laser energy (e.g. 635 nm and 808 nm) is smaller than the gap energy between the minimum of the conduction band (E<sub>c</sub>) of TeO<sub>2</sub> and the maximum of the valence band (E<sub>v</sub>) of Te (about 2.09 eV), the excitation and recombination of photo-induced electron-hole pair occur only in the direct-band-gap Te, resulting in a fast response time as exhibited in Fig. 3 (c)(d) in manuscript. However, when the excited laser energy (e.g. 405 nm and 532 nm) is larger than 2.09 eV, the photo-induced electron has energy high enough to transit from the valence band of Te to the conduction band of the TeO<sub>2</sub>. In this case, the photo-induced electron is separated from the photo-induced hole in Te and transported physically to TeO<sub>2</sub>, which lead to the prolonging of the lifetime of electron. Also, the electron excited to TeO<sub>2</sub> will partially recombine with hole in valence band of TeO<sub>2</sub>, again prolonging the recombination process of the photo-induced electron-hole pair.

For the excitation of 405 nm, electron is excited to a higher energy level above the  $E_c$  of TeO<sub>2</sub>, which needs more time to relax to a lower level. In this process, most of the electron in high energy level drift to the electrode before they relax to  $E_c$ , resulting in a faster response of the photocurrent when compared with those exited to a lower energy level (e.g. exited by 532 nm). Therefore, as the 532 nm laser has energy very close to the required minimum energy of 2.09 eV, the photocurrent of Te flakes under 532 nm illumination exhibits very slow response.



**Fig. S8.** The power-dependence sensitivity curve under 405 nm laser illumination with different power density. Inset: the enlarged partial curve. The sensitivity can be expressed as

$$S = \frac{I_{Ph}}{I_{dark}P}$$
(5)

where the  $I_{Ph}$  is photocurrent,  $I_{dark}$  is dark current and P is the light density.



**Fig. S9.** (a) The photoresponsivity and (b) detection rate under different wavelengths laser illumination with different light density.



Fig. S10. The photocurrent of the FETs based on Te flakes (a) under a continuous 50 cycle of 635nm layer illumination and (b) before and after a ten-days' exposure in the air.  $V_{ds} = 0.2 \text{ V}$ ,  $V_g = 0 \text{ V}$ .

## References

[S1] A. S. Pine and G. Dresselhaus, Phys. Rev. B,1971, 4, 356-371.

[S2] D. R. Lide, CRC Handbook of Chemistry and Physics, 2010, 90nd Edition

- [S3] Z. Shi, R. Cao, K. Khan, A. K. Tareen, X. Liu, W. Liang, Y. Zhang,
- C. Ma, Z. Guo, X. Luo and H. Zhang, Nano-Micro Lett., 2020, 12, 99
- [S4] A. Zavabeti, P. Aukarasereenont, H. Tuohey, N. Syed, A. Jannat, A.
- Elbourne, K. A. Messalea, B. Y. Zhang, B. J. Murdoch, J. G. Partridge, M.
- Wurdack, D. L. Creedon, J. V. Embden, K. K. Zadeh, S. P. Russo, C. F.

McConville and T. Daeneke, Nat. Electron., 2021, 4, 277-283