Supporting Information

Ultrathin tellurium dioxide: Emerging direct band gap semiconductor with high-mobility transport anisotropy

Shiying Guo,[a] Zhen Zhu,[b] Xuemin Hu,[a] Wenhan Zhou,[a] Xiufeng Song,[a] Shengli Zhang,[a,*] Kan Zhang,[a] Haibo Zeng[a,*]

[a]Key Laboratory of Advanced Display Materials and Devices, Ministry of Industry and Information Technology, College of Material Science and Engineering, Nanjing University of Science and Technology, Nanjing 210094, China

[b]Materials Department, University of California, Santa Barbara, CA 93106, USA

*Corresponding author: zhangslvip@njust.edu.cn and zeng.haibo@njust.edu.cn

Table S1 The relaxed crystal parameters of three polymorphs for bulk TeO_2: tetragonal α-TeO_2 (Paratellurite, $P4_12_12$), orthorhombic β-TeO_2 (Tellurite, Pbca), and

a metastable phase γ-TeO_2 ($P2_12_12_1$).

Phases	α	β	γ
Space group	$P4_12_12$	Pbca	$P2_12_12_1$
a(Å)	4.97	5.54	4.47
b(Å)	4.97	5.73	5.11
c(Å)	7.62	12.22	8.80

Table S2 The calculated cohesive energies of three polymorphs for bulk TeO_2 with two methods: PBE and PBE+vdw interactions. Our results show that the paratellurite and tellurite phases have very similar cohesive energies, which are larger than that of the metastable γ-TeO_2 phase, in consistent with the experiments.

Phases	α	β	γ
$E-E_\alpha$	0 meV	-11 meV	+0.4 meV
$E-E_\alpha$(+vdW)	0 meV	+3 meV	+60 meV

Table S3 Lattice parameters a and b of TeO_2 crystals. The lattice parameters (a and b) decrease progressively while thinning the thickness of TeO_2 crystal.

N_L	1	2	3	4	5	6	bulk
a (Å)	5.459	5.503	5.518	5.525	5.529	5.532	5.541
b (Å)	5.726	5.727	5.731	5.732	5.733	5.733	5.734

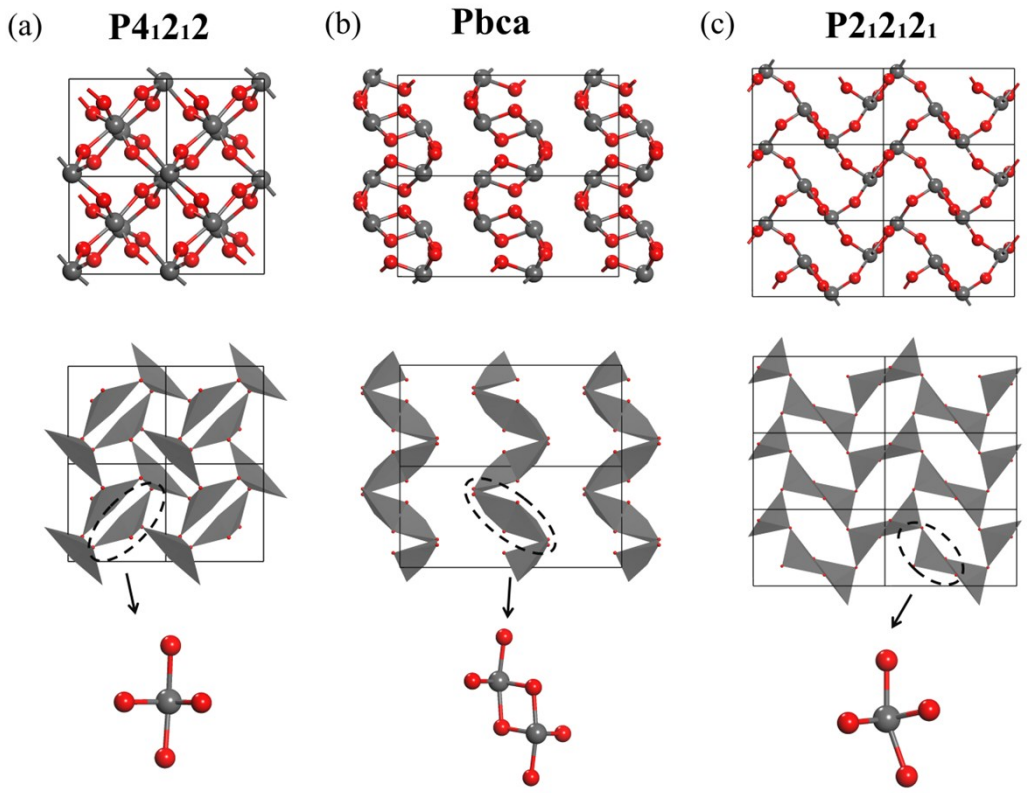

Fig. S1 The relaxed three polymorphs for bulk TeO_2: tetragonal α-TeO_2 (Paratellurite, $P4_12_12$), orthorhombic β-TeO_2 (Tellurite, Pbca), and a metastable phase γ-TeO_2 ($P2_12_12_1$).

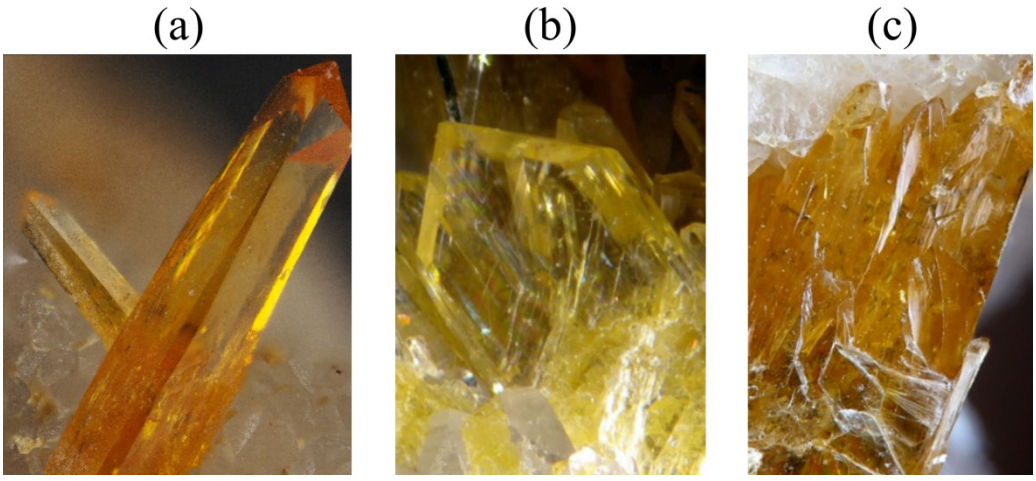

Fig. S2 (a-c) Natural mineral tellurite of layered β-TeO_2. Reproduced from https://www.mindat.org/min-3905.html.

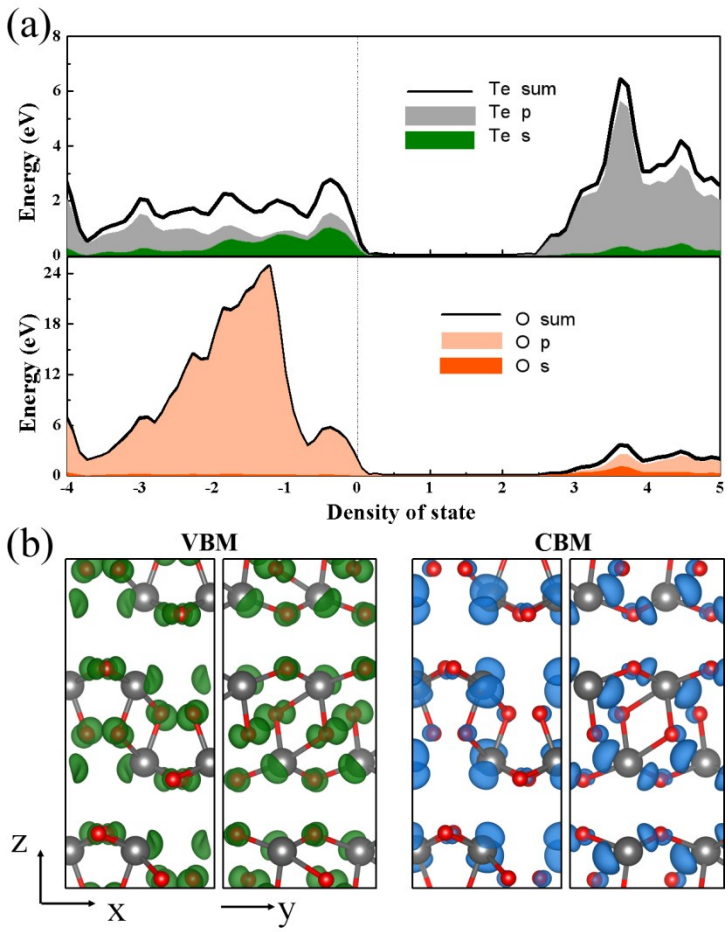

Fig. S3 (a) Projected density of states (PDOS) of Te and O atoms in monolayer TeO_2. (b) and (c) Isosurfaces of partial charge densities corresponding to the VBM and CBM.

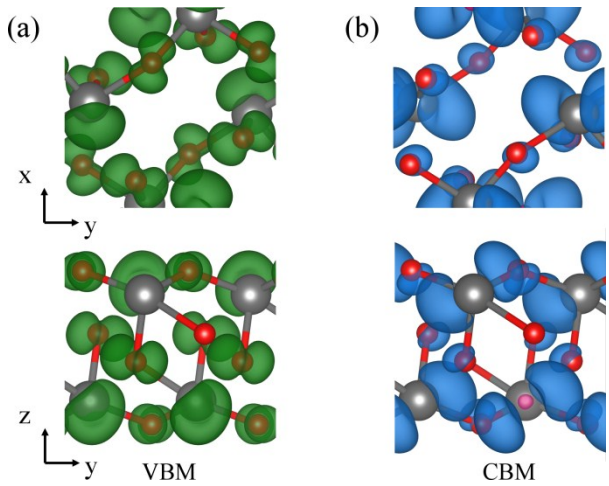

Fig. S4 Isosurfaces of partial charge densities corresponding to the VBM (a) and CBM (b) in monolayer TeO_2. The isosurface value is 0.03 e/Å^3.

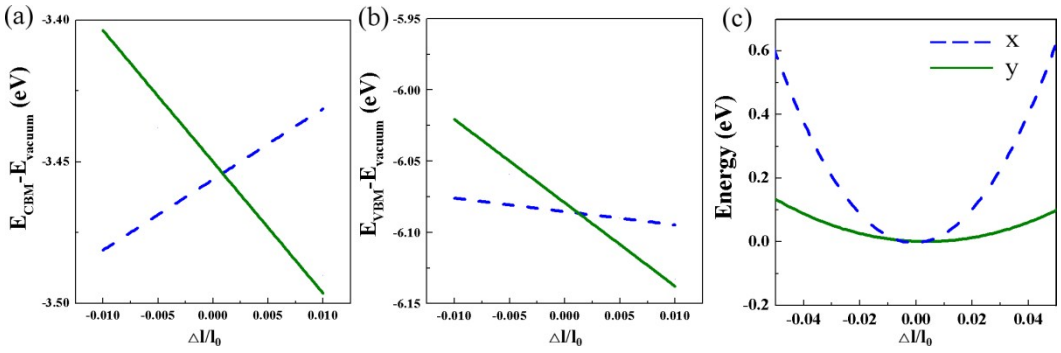

Fig. S5 Shifts of CBM (a) and VBM (b) under uniaxial strain along the x and y directions for monolayer TeO_2. (c) Relations between strain and total energy.

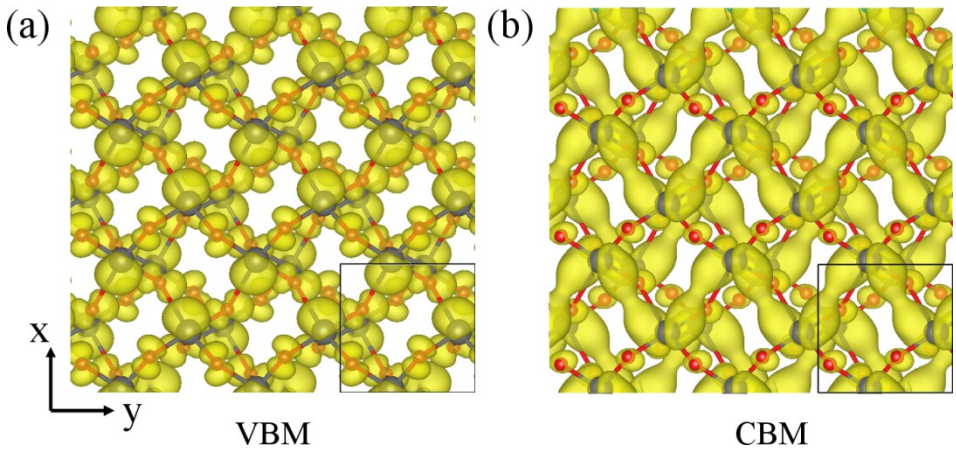

Fig. S6 Isosurfaces of partial charge densities corresponding to the VBM (a) and CBM (b). The isosurface value is 0.002 e/bohr^3.

Table S4 The calculated effective mass (m*) and mobility (μ) of electrons and holes along Γ-X and Γ-Y directions for monolayer TeO_2 at 300 K.

Carrier type	m* x	m* y	μ_{x_2D} ($cm^2V^{-1}s^{-1}$)	μ_{y_2D} ($cm^2V^{-1}s^{-1}$)
e^-	2.36	0.36	790~820	1200~2100
h^+	0.50	0.54	8400~9100	180~250