Research article

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Ultra-strong anisotropic photo-responsivity of bilayer tellurene: a quantum transport and time-domain first principle study

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Abstract: Unlike traditional two-dimensional layered materials, tellurium (Te) shows its one-dimensional van der Waals structure and triggers intensive researches. Through a density functional theory coupled with the nonequilibrium Green's function calculation, we found that bilayer tellurene has a broad photoelectric response over the range from the visible to the near-infrared region. Besides, bilayer tellurene photodetector presents an ultra-strong anisotropic photo-responsivity and an ultra-high extinction ratio (~ 2812 at the photon energy of 3.4 eV) because of its non-layer/chain structure, which is superior to the antimonene (~ 145) and phosphorene/blue phosphorene bilayers (~ 240). Time-domain first principle study further reveals that the ultra-strong anisotropy comes from the transition of Te 5p bonding orbitals along or perpendicular to the chain directions. With these excellent optoelectronic merits, bilayer tellurene may become a promising candidate for next-generation photoelectronic devices.

Keywords: bilayer tellurene photodetector; quantum transport; TD-DFT; ultra-high extinction ratio; ultra-strong anisotropy; non-layer structure.

1 Introduction

Two-dimensional (2D) materials have evoked intensive studies of application in electronic and optoelectronic devices, e.g. solar cell [1, 2], transistors [3, 4], light-emitting diodes [5], photodetector [6–8], and spintronic device [9, 10]. Compared with their bulk counterpart, the unique crystal structure endows 2D materials with striking properties [11]. Gradually, a large number of other 2D materials, such as group-VA layered materials [12], transition metal chalcogenides [13], perovskites materials [14], and indium monochalcogenides [15], emerged in frontier research. Despite the intriguing properties, the restrictions in the synthesis condition [16, 17], potential of scaling up, and environmental stability [18, 19] of synthesized materials become an obstacle for the progress of 2D materials. Therefore, it is necessary to find a new material displaying better stability at room temperature and lower cost of preparation.

Tellurene is a good candidate for such materials [20]. Monolayer tellurene is only stable in β -phase, which has mirror symmetry, and Te atoms are connected only by covalent bonds [21]. Bilayer tellurene is stable in α -phase which demonstrates one-dimensional-like nonlayered structure. Therefore, the bilayer tellurene was chosen in this work. A large variety of schemes have been developed to synthesize a large-area tellurium nanosheet displaying good air-stability over 2 months, such as substrate-free solution [22], liquid exfoliation method [23], and van der Waals epitaxy [24]. It has been reported that tellurene presents extremely high hole mobility on the order of 10^5 cm²/V s and eccentrically robust optical absorption in the close-packed nonbond direction [21]. In recent experiments, tellurene has been verified to be a promising candidate for application, such as field effect transistor [25], photodetector [26], and lithium-tellurium batteries [27, 28]. For example, Wang et al. [22] pronounced that the tellurene transistor shows a high on/off ratio on the order of 10^6 , heavy drain current of over 300 mA/mm, and field-effect mobility of approximately 700 cm²/V s. Conversely, Du et al. [29] demonstrated that the distinct

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magneto-resistance and phase coherence length obtained in three directions proves strong anisotropy in magnetic transport. However, the anisotropic photoresponse of bilayer tellurene was seldom reported either experimentally or theoretically. Besides, little attention has been paid to the physical origin of anisotropy in the photocurrent of the nano- and optoelectronic devices based on bilayer tellurene. Therefore, new investigation of the anisotropic photocurrent of bilayer tellurene is highly demanded.

In this work, density functional theory (DFT) combined with the Keldysh nonequilibrium Green's Function theory (NEGF-DFT) was employed to resolve the photoelectric performance of bilayer tellurene photodetector. Time-dependent DFT (TD-DFT) was used to explore the physical origin of the ultra-strong anisotropy of photoresponse. The largest extinction ratio indicates strong polarization sensitivity and anisotropy between the covalent bond and nonbond directions. These wide-range and strong anisotropy make bilayer tellurene potential candidate for photonic and polarizer and plasmonic devices.

2 Method

2.1 Quantum transport calculations

The photocurrent calculation was performed based on the DFT coupled with the nonequilibrium Green's function [30, 31], as implemented by Nanodcal package. In this work, we proposed a two-probe bilayer tellurene photodetector. The device is divided into three parts: central scattering region and left and right electrodes, which extend to $\pm\infty$. For the quantum transport system, the ground-state self-consistent Hamiltonian H_0 is calculated based on DFT. The electron-photon interaction is considered as a perturbation, which is added to the H_0 . Hence, the total Hamiltonian is given by the following:

$$\hat{H} = \hat{H}_0 + \frac{e}{m_0} \mathbf{A} \cdot \hat{\mathbf{p}} \quad (1)$$

where $\mathbf{A}$ is the electromagnetic vector potential and $\hat{\mathbf{p}}$ is the momentum of electron. The current flowing through the device can be obtained by the Landauer-Büttiker equation, which is given by the following:

$$I(V_b) = \frac{2e}{\hbar} \int T(E, V_b) [f(E - \mu_L) - f(E - \mu_R)] dE \quad (2)$$

where μ_L and μ_R are the electrochemical potentials of the left and right leads. The $f(E - \mu_{L/R})$ is the Fermi distribution

function. $T(E, V_b)$ is the transmission function around the Fermi level, describing the probability of an electron with a given energy that transfers from the left (right) electrode through the central region and into the right (left) electrode under a certain bias voltage. In particular, transmission under a zero bias is given by $T(E) = \text{Tr}(\Gamma_1 G \Gamma_2 G^\dagger)$, where G is the Green's function.

2.2 TD-DFT calculations

The TD-DFT calculations are performed using the CASTEP code [32] with the OTFG ultrasoft pseudopotential. The Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation [33] is adopted to describe the exchange-correlation energy. The energy cutoff of the plane wave basis set is chosen to be 400 eV. The Brillouin zone of the bilayer tellurene is sampled with a Monkhorst-Pack grid of $10 \times 10 \times 10$ and $16 \times 16 \times 1$, respectively. The self-consistency threshold of the total energy is set to 10^{-6} eV/atom. All the systems are allowed to fully relax to look for a ground state until the maximum energy, force, stress, and displacement converge to 10^{-5} eV/atom, 0.03 eV/Å, 0.05 GPa, and 0.001 Å, respectively. Periodic boundary conditions are applied throughout the study, with the bilayer structure represented by a periodic array of nonlayered slab separated by a 20 Å thick vacuum region. The damped Grimme scheme is considered to describe the dispersion interaction at the PBE level [34]. The hybrid functional (HSE06) method [35, 36] is adopted to obtain more accurate bandgap.

TD-DFT is carried out to explore the excitonic effect in the bilayer tellurene, as TD-DFT simulation can obtain accurate results for the excited state dynamic process [37]. TD-DFT calculation considers the time variation of the exchange-correlation energy, which is beyond the random-phase approximation. Eight low-lying excitons were considered in the calculation. The low-lying excitons reveal the physical origin of the energy-dependent photoresponse and the anisotropy caused by transition dipole polarizations.

3 Results and discussion

3.1 Bilayer tellurene photodetectors in three transport directions

Figure 1 shows the two-probe bilayer tellurene photodetector. The device consists of the central scattering region, left lead, and right lead. Three transport directions (y -axis) were considered: covalent bond 1-direction, close-packed

Figure 1: Illustration of bilayer tellurene photodetector.

(A) The model of the bilayer tellurene photodetector. (B) The diagram of the helical chain of tellurium along the 1-direction and the definition of three transport directions. (C) The lattice structure of bulk tellurium. The top and side views of bilayer tellurene photodetector in the (D) covalent bonded 1-, (E) close-packed nonbond 2-, and (F) nonbond 3-directions. The left and right electrodes are enclosed with black arrow, and the rest area is the central scattering region that the incident light illuminates. The polarization angle θ is the angle of light polarized orientation with respect to its transport direction.

nonbond 2-direction, and nonbond 3-direction. The 1-direction is along the atomic chain of bilayer tellurene (black arrow). The 2-direction is along the van der Waals direction on the close-packed plane (blue arrow), where atoms have the mirror symmetry in the xy -plane. The 3-direction is along the nonbond direction on the non-close-packed plane (green arrow). A linear-polarized light is applied in the z -direction. The linear polarization angle of incidental light is the angle θ with respect to its transport direction in the xy -plane.

The left and right leads were applied by the bias voltages ranging from 0.1 to 1.0 V to obtain a continuous

photo-induced current. The linearly polarized light with photon energies of 0.7–3.4 eV is used to irradiate the whole scattering region, which covers the range of E_g (Supporting Information). The polarization vector of linearly polarized light can be written as follows:

$$\mathbf{e} = \cos\theta\mathbf{e}_1 + \sin\theta\mathbf{e}_2 \quad (3)$$

where θ is the angle of the polarization direction with respect to the vector $\mathbf{e}_1$. The $\mathbf{e}_1$ and $\mathbf{e}_2$ are set to the transport and periodic directions in the systems.

The α -phase bilayer tellurene (space group $P2_1$) is considered in this work. The DFT-optimized lattice parameters, bond lengths, and the distance of chain at the PBE+D level are $a=b=4.4593$ Å ($a=b=4.4593$ Å), $c=5.9282$ Å ($c=5.9282$ Å), and $d_{\text{Te-Te}}=2.8898$ Å, respectively, for the bilayer tellurene (bulk tellurium).

3.2 Linear-polarized angle-resolved photocurrent

Figure 2 shows the dependence of photocurrent on the polarization angle θ with the photon energies of 1.8, 2.0, and 2.4 eV in the 1-direction (chain direction), close-packed nonbond 2-direction, and nonbond 3-direction at the bias voltage of 0.2 V. The photocurrent I_{ph} in the 1-, 2-, and 3-direction can be well fitted by an approximate $\cos(2\theta)$ dependence, demonstrating anisotropic photocurrent between the covalent bond and nonbond directions in the bilayer tellurene. Phenomenologically, the photocurrent of the materials with C_2 symmetry under the irradiation of linearly polarized light is relevant to either $\cos(\theta)$ or $\sin(\theta)$, which exactly agrees with the results. This distinction gives rise to strong polarization sensitivity and anisotropy between these two high-symmetry directions.

Besides, it can be seen that the phase of $I_{\text{ph}}(\theta)$ at the photon energies of 1.8 and 2.0 eV is sensitive to 1-direction and 2-direction, whereas the phase of $I_{\text{ph}}(\theta)$ for the photon energy of 2.4 eV is relatively independent. As the HOMO-LUMO energy gap locates in approximately 1.75 eV, the electron transitions of 1.8 and 2.0 eV correspond to the transfer from the HOMO to the LUMO containing doubly degenerate states at Γ point. The major contribution to the HOMO and LUMO is the p_x and p_z orbitals (x , y , and z

are defined in Figure 1E), which are continuous in the surface along the chain. Hence, the phase of $I_{\text{ph}}(\theta)$ at the photon energies of 1.8 and 2.0 eV is sensitive to 1-direction and 2-direction. However, the electron transition from HOMO-1 to LUMO+3 at around 2.47 eV is dominated by p_y orbital, which is perpendicular to the chain (nonbond direction). In contrast, the p_x , p_y , and p_z orbitals in the 3-direction are not continuous. The electrons transporting in the 3-direction suffer a great obstacle, leading to the photocurrent retaining value less than 20. Additionally, the Hartree potential profile (Supporting Information) verifies the fluctuating potential barriers in the 3-direction.

3.3 Photon energy-dependent photocurrent and bias-induced saturation

Figure 3A–C compare the maximum photocurrent $I_{\text{m}}(\theta)$ vs. photon energy E for bias voltages 0–1.0 V along the 1-, 2-, and 3-directions. In 1-direction, at first, $I_{\text{m}}(\theta)$ increases with the photon energy and then experiences several peaks in the 1.3–2.5 eV range and finally shows a sudden decline until the 3.4 eV. In the 2-direction, there is a rise of the $I_{\text{m}}(\theta)$ at the range of 0.7–1.3 eV, after which $I_{\text{m}}(\theta)$ fluctuates in the range of 1.3–2.2 eV and then falls steadily. The peak region (1.3–2.2 eV) is narrower compared with the 1-direction. In the 3-direction, $I_{\text{m}}(\theta)$ for most of bias voltages sees a gradual growth and reaches its maximum value and then decreases modestly. Overall, the photoresponse can be observed in the near-infrared, compared with the bismuth quantum dots (200–600 nm) [38]. However, $I_{\text{m}}(\theta)$ at the bias of 0.5 V has relatively large values and shows several peaks throughout the photon energies of 0.7–3.4 eV. To explain this phenomenon, we investigated the

Figure 2: The dependence of photocurrent on polarization angle in bilayer tellurene. The photocurrent as a function of the polarization angle θ in the (A) covalent bond 1-direction, (B) close-packed nonbond 2-direction, and (C) nonbond 3-direction of bilayer tellurene for the photon energies of 1.8, 2.0, and 2.4 eV at 0.2 V. The lines are the fitted results.

Figure 3: The dependence of maximum photocurrent on photon energy and bias voltage in bilayer tellurene.

The maximum photocurrent as a function of (A–C) photon energy and (D–F) bias voltage in the covalent bond 1-, close-packed nonbond 2-, and nonbond 3-directions of bilayer tellurene, respectively.

transmission coefficient $T(E)$ at 0.5 V, as well as 0.3 and 0.9 V for comparison, as shown in Figure S3 (Supporting Information). We found that $T(E)$ at 0.5 V has the largest value compared with other bias. As the current depends on the integral of $T(E)$ within the bias window, $I_m(\theta)$ at 0.5 V has the largest values and shows peaks within the corresponding energy range. In general, $I_m(\theta)$ experiences an increase with the bias voltage and then reaches its largest value and finally declines instead. That is, the maximum photocurrent saturates at a certain photon energy in the visible range.

Figure 3D–F describe the dependence of $I_m(\theta)$ on the bias voltages for different photon energies. $I_m(\theta)$ for small photon energies monotonically increases with the bias, such as 0.7 and 0.8 eV in the 1- and 2-directions. For most photon energies, $I_m(\theta)$ shows an upward trend and reaches

its maximum value at a certain bias voltage. For instance, 2.4 eV, $I_m(\theta)$ in the 1-direction grows sharply and obtains its peak value at 0.9 V and then decreases; in the 2-direction, $I_m(\theta)$ for a photon energy of 3.1 eV remains stable and sharply increases at 0.5 V, and then it decreases rapidly instead. Along the 3-direction, some $I_m(\theta)$ reaches its largest value greater than 70 at a bias of 0.5 V. This means that the maximum photocurrent saturates at a certain bias voltage.

3.4 Ultrahigh extinction ratio and strong anisotropy

To further evaluate the polarization sensitivity of the bilayer tellurene, an extinction ratio is defined as $I_{\parallel}/I_{\perp}$ or

Figure 4: The photon energy-dependent extinction ratio in bilayer tellurene. The variation in the extinction ratio of photocurrent with the bias voltages for different photon energies in the (A) covalent bond 1-, (B) close-packed nonbond 2-, and (C) nonbond 3-directions of bilayer tellurene, respectively. The inset (A–C) is the photocurrent as a function of polarization angle θ at their respective largest extinction ratio.

$I_{\perp}/I_{\parallel}$, which is the ratio of I_{ph} in the covalent bond 1- and close-packed nonbond 2-direction (nonbond 3-direction) with different bias voltages for each photon energy, as shown in Figure 4. Here, $I_{\parallel}$ and $I_{\perp}$ are the photocurrents at $\theta = 0^{\circ}$ and $\theta = 90^{\circ}$ with respect to the transport direction, respectively. In the 1-direction, it is clear that some photon energies have large extinction ratios, such as $I_{\parallel}/I_{\perp}$ of 35 for a photon energy of 3.0 eV at a bias of 0.4 V, $I_{\parallel}/I_{\perp}$ for a photon energy of 2.7 eV of 10 at a bias of 0.5 V and of 17 at a bias of 0.7 V. But in some certain energies such as 1.8, 2.4, and 3.2 eV, $I_{\parallel}/I_{\perp}$ values are less than 5, indicating those transition modes are relatively independent of θ . In the 2-direction, large $I_{\perp}/I_{\parallel}$ over 10 are achieved at some

photon energies. For instance, $I_{\perp}/I_{\parallel}$ is 60.29 at a bias of 0.2 V for the photon energy of 0.7 eV and 98.19 at 0.9 V for the photon energy of 2.7 eV.

It can be seen that in the 3-direction, most of photon energies have large extinction ratios, such as 1.0, 2.5, and 3.0 eV. Strikingly, an extremely large $I_{\parallel}/I_{\perp}$ of 2812 is obtained for the photon energy of 3.4 eV at a bias of 0.3 V. These large $I_{\parallel}/I_{\perp}$ derive from the almost zero value of photocurrent $I_{\parallel}$ for these photon energies in the 3-direction, as shown in Figure 4C inset. This means that there is a strong anisotropy between the 3-direction and its perpendicular direction-intrachain direction in the bilayer tellurene. This largest calculated extinction ratio is much higher than the extinction ratios of the bilayer phosphorene/blue phosphorene photodetector (134 at 1.0 eV with V_g and 273 at 2.8 eV without V_g) [39] and antimonene (145 at 3.2 eV) [40] in the calculations. Anisotropy in the photocurrent was observed in the PbS nanowire FETs with an extinction ratio of 2.38 [41]. Black phosphorus photoelectronic device achieved a large experimental extinction ratio of 5 [42]. Sb_2Se_3 nanoflake was also investigated in the experiment, exhibiting a superior extinction ratio of 16 [43]. Thus, bilayer tellurene shows the strongest polarization sensitivity and anisotropy than other 2D materials.

3.5 The permitted transmission channels

Figure 5 illustrates contour maps of transmission coefficient $T(E)$ as a function of energy and bias voltage in the 1-, 2-, and 3-directions, respectively (See Figure S4 also). It is noted that the area enclosed by black lines (Figure 5) is the region of bias window where the current depends on the integral of the $T(E)$. For zero bias, there is no transmission channel in the bias window. Obviously, in the 1- and 2-directions, the peak appears in the bias window, and the peak value of transmission function enhances as the bias voltage increases. This means that the integral area of the $T(E)$ rises, leading to a greater photocurrent with a higher photon energy in the transport direction. In contrast, for the 3-direction, the integral of $T(E)$ shows an upward trend with the increase of bias until 0.5 V, and then it decreases instead (Supporting Information). Besides, the peak position of the main peak of $T(E)$ moves to lower energy level as the bias increases, corresponding to the shift of a particular energy level. The Fermi level of the bilayer tellurene changes with bias voltage. This causes the movement of a particular energy level of $T(E)$ within the bias window, which affects the magnitude of photocurrent at a certain bias.

Figure 5: The dependence of transmission coefficient on bias voltage and photon energy. Contour maps of transmission coefficient (lg) as a function of energy varying from -0.5 to 0.5 eV and bias voltage of 0 – 1.0 V in the (A) covalent bond 1-, (B) close-packed nonbond 2-, and (C) nonbond 3-directions of bilayer tellurene photodetector. The area enclosed by black lines is the region of bias window.

Figure 6: TD-DFT resolved anisotropy in electron transitions of bilayer tellurene. (A) The band structure of bilayer tellurene. The blue and red arrows in (A) indicate the electron transitions from Γ_1 to Γ_2 and from Γ_1 to Γ_3 . (B) Total density of states of bilayer tellurene. The shadow areas denote the peaks of DOS. (C) The Brillouin zones of the bilayer tellurene. The top, side, and top views of contributing molecular orbitals at the (D) HOMO, (E) LUMO, (F) HOMO-1, and (G) LUMO + 3 energy bands. Axes x , y , and z define the directions of p orbitals. Red arrows indicate the electron transport direction.

3.6 The origin of strong anisotropy revealed by time-domain first principle study

Figure 6A shows the band structure of bilayer tellurene. As the momentum of photon is rather small, the Γ point plays an important role in the electron transition. The energy band around the Γ point is adopted to explore the physical origin of the anisotropic photocurrent. Electron transitions mainly occur between the valence band and the doubly degenerate valence bands (blue rectangle, $\Gamma_1-\Gamma_2$) and the higher energy band (red rectangle, $\Gamma_1-\Gamma_3$) at the conduction band (Figure 6A). Figure 6B shows the total density of states (DOS) of the bilayer tellurene. There are two main DOS peaks that are contributed by the energy bands around the Γ point in the DOS profiles: one is at around 1.5 eV in the conduction band, the other one is at around -1.0 eV in the valence band. A high electron transition probability can be obtained between these two DOS peaks, corresponding to a large photocurrent. Figure 6C shows the Brillouin zone and the path of band structure ($\Gamma \rightarrow M \rightarrow Y \rightarrow \Gamma \rightarrow X \rightarrow H' \rightarrow M$). $\Gamma \rightarrow X$ corresponds to the direction along the chain and $\Gamma \rightarrow Y$ is the direction across the chain. From the band structure, the effective mass and mobility can be obtained (see Supporting Information for details). The effective masses along the chain are $m_{e-||}^* = 0.496m_0$ and $m_{h-||}^* = 0.295m_0$ (m_0 is the free-electron mass). For across the chain direction, the effective masses for e and h are both negative, $m_{e-\perp}^* = -0.664m_0$ and $m_{h-\perp}^* = -0.351m_0$. The negative m^* indicates that the external electric field does negative work to the carrier, which means that the carrier can hardly be driven by the external electric field. The carrier mobility for electrons along the chain $\mu_{||}(e)$ is $2.1 \times 10^4 \text{ cm}^2/(\text{V s})$, whereas the calculated μ for holes $\mu_{||}(h)$ is $2.31 \times 10^3 \text{ cm}^2/(\text{V s})$. For the direction across

Table 1: The TD-DFT derived electron transitions and their respective excited states.

Excited states	Contributing energy band	E_{photon} (eV)	λ_{photon} (nm)
1	HOMO $\rightarrow$ LUMO	1.923	645
2	HOMO $\rightarrow$ LUMO + 1	1.949	636
3	HOMO $\rightarrow$ LUMO + 2	1.990	623
4	HOMO-1 $\rightarrow$ LUMO	2.140	579
5	HOMO-1 $\rightarrow$ LUMO + 1	2.161	574
6	HOMO-1 $\rightarrow$ LUMO + 2	2.254	550
7	HOMO $\rightarrow$ LUMO + 3	2.323	534
8	HOMO-1 $\rightarrow$ LUMO + 3	2.467	503

the chain, the mobility $\mu_{\perp}(e)$ and $\mu_{\perp}(h)$ are supposed to be extremely low. This means that there is a strong anisotropy in transport mobility of the bilayer tellurene.

To further investigate which molecular orbitals contribute to the electron transitions, electron transitions around the Γ point are calculated and are assigned to respective energy levels through the TD-DFT calculation, as shown in Table 1. The corresponding molecular orbitals (Figure 6D–G) are calculated with the eigenvalues of energy bands and can be written as the superposition of atomic orbitals:

$$\begin{aligned}
 \Gamma_{\text{HOMO}} &= 0.359\text{Te}(5p_x) + 0.351\text{Te}(5p_z) \\
 \Gamma_{\text{LUMO}} &= -0.271\text{Te}(5p_x^*) - 0.605\text{Te}(5p_z^*) \\
 \Gamma_{\text{HOMO-1}} &= 0.554\text{Te}(5p_y) \\
 \Gamma_{\text{LUMO+3}} &= -0.629\text{Te}(5p_y^*)
 \end{aligned} \quad (4)$$

Both the p_x and p_z atomic orbitals contribute to LUMO, LUMO + 1, and LUMO + 2 energy levels. It is obvious that when a photon with ~ 1.9 eV irradiates the scattering region, the electrons transit from the HOMO to LUMO,

Figure 7: Anisotropy excitons in bilayer tellurene.

The polarization dependence of excitons for the energies of (A) 1.923 and (B) 2.467 eV to mimic the absorption spectra of bilayer tellurene. The polarization angle θ is set to the light polarization with respect to the covalent bond 1-direction.

which are both dominated by p_x and p_z atomic orbitals. As the p_x and p_z orbitals locate in the plane along the chain, the transition orbital dipoles and the linearly polarized light are coupled well, leading to the maximum photocurrent at a photon energy of ~ 1.9 eV. As the energy of incident light increases, the major contribution to the energy band becomes p_y orbital, which is perpendicular to the chain direction, resulting in low transition probability, that is, relatively small photocurrent. Thus, when the bilayer tellurene absorbs the photon of a certain energy, the p orbitals make a contribution to these electron transitions.

To further verify the anisotropic polarization dependence, we investigate the polarization-dependent oscillator strength of the absorption coefficient of the excited excitons with the energies of 1.923 eV (HOMO $\rightarrow$ LUMO) and 2.467 eV (HOMO-1 $\rightarrow$ LUMO+3), as shown in Figure 7. It can be seen that for the exciton with 1.923 eV, the absorption coefficient has a maximum value observed at the light polarization θ of either 90° or 270° (close-packed nonbond 2-direction). The largest absorption value obtained along the chain can be explained as the coupling of $5p_x$ and $5p_z$ orbitals and the electric field of incident light. In contrast, the absorption coefficient for the exciton of 2.467 eV achieves its maximum value at the polarization angle of either 0° or 180° (covalent bond 1-direction), corresponding to the contributing $5p_y$ atomic orbital across the chain in the xy quasi-plane. Hence, it can be concluded that the absorption coefficient is light polarization dependent and has a period of 180° , exhibiting strong anisotropically optical property in the xy quasi-plane, which is in excellent agreement with the results calculated before. Having the similar nonlayered structure with bilayer tellurene, selenium nanoflake has been reported to possess stable mode-locked performance [44], indicating the great potential for the exploration of bilayer tellurene.

4 Conclusions

In conclusion, we have revealed the physical origin of strong anisotropy in the photocurrent in the bilayer tellurene based on DFT with Keldysh nonequilibrium Green's Function calculation. The photocurrent has an approximate cosine dependence on the polarization angle in these three transport directions. The anisotropy in the photocurrent derives from the difference of intrinsic atomic orbitals that make a contribution to the energy levels. Moreover, the largest extinction ratio of 2812 is achieved at 0.3 V for a photon energy of 3.4 eV in the bilayer tellurene photodetector because of its one-dimensional-like nonlayered

structure. Additionally, time-dependent first principle calculation resolves that the major contribution to the electron transitions is the p orbitals. These results describe a high polarization sensitivity, a broadband photoresponse, and a large anisotropy in the bilayer tellurene photodetector.

Supporting Information: Supporting Information is available free of charge on the website.

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