Supplementary Information (SI) for Physical Chemistry Chemical Physics.
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Supporting information file for:

Ferroelastic Phase Transition-Modulated Electronic Transport and Photoelectric Properties in Monolayer 1T' $ZrCl_2$

Yuehua Xu, He Sun, Jindian Chen, Qianqian Long, and Haowen Xu*

School of Microelectronics and Control Engineering, Changzhou University, Changzhou 213164, Jiangsu, China.

Corresponding author: yhxu@cczu.edu.cn

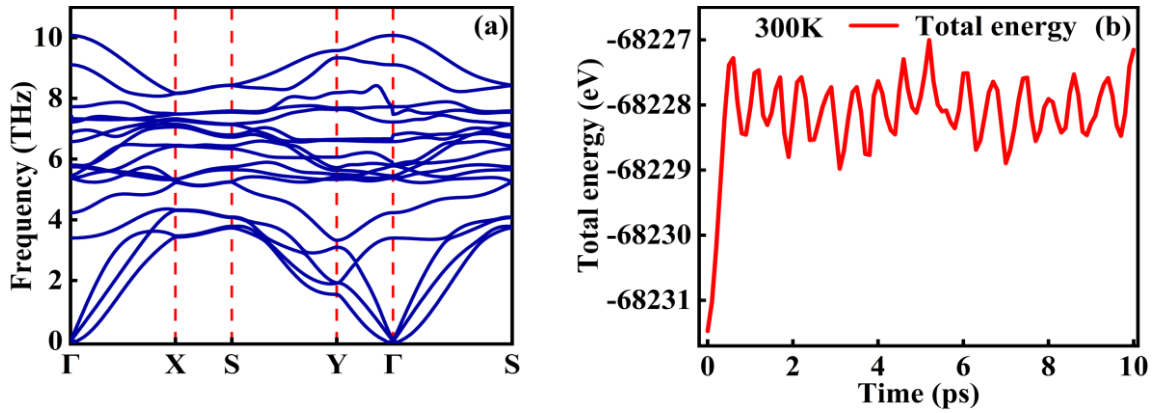

Fig. S1 (a) The phonon spectrum of the O1 variant and (b) The energy fluctuation plot from molecular dynamics with a time step of 1 *fs* at a temperature of 300 K.

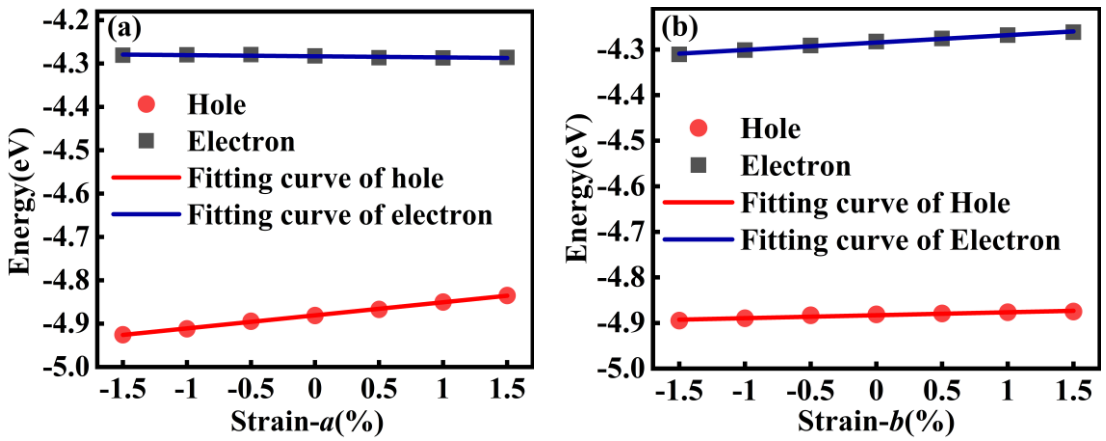

Fig. S2 Fitting plot of the CBM and VBM variations of the O1 variant under stress applied in (a) the *a* direction and (b) the *b* direction.

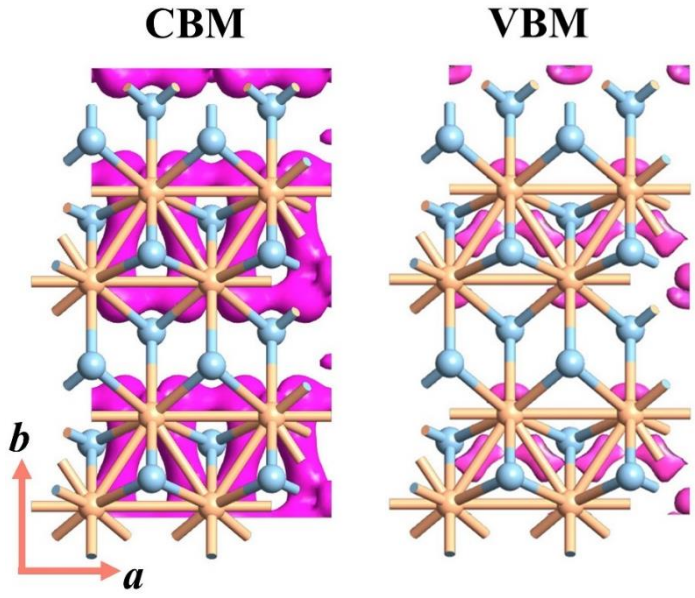

Fig. S3 Top views of the spatial structure of wave functions for the monolayer O1 variant of $ZrCl_2$ at the *k* points corresponding to the CBM (left) and VBM (right), respectively. An isosurface of 0.00007 $Å^{-3}$ is adopted.

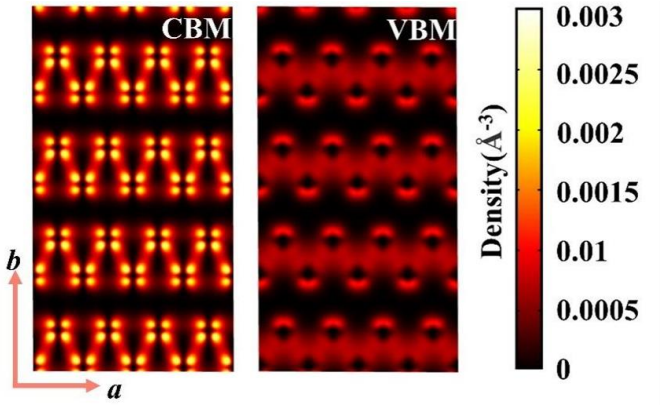

Fig. S4 The partial charge density at CBM (left panel) and VBM (right panel) of the O1 variant.

The reason for using the DFT-PBE method to calculate the photoelectric properties

(1) The high doping concentration (10^{14}/cm²) in our system:
- Significantly enhances screening effects.
- Weakens electron-hole interactions.
- Reduces the excitonic effects that typically dominate in some pristine 2D materials.[1-3]

(2) The built-in electric field in our *p-i-n* structure:
- Promotes rapid separation of photogenerated carriers.
- Prevents the formation of stable exciton states.
- Makes the system's response more dependent on carrier transport than excitonic effects.

(3) Previous studies have demonstrated that in 2D systems, the band gap increase due to electron-electron interactions (GW correction) is often compensated by the decrease from electron-hole interactions (excitonic effects), making DFT-GGA a reasonable approximation. For instance, studies of graphdiyne showed that the GW correction of +0.66 eV was largely offset by the BSE correction of -0.55 eV, resulting in optical spectra that were better approximated by DFT calculations.[4]

(4) Our approach is supported by extensive literature showing excellent agreement between DFT-GGA calculated photocurrents and experimental measurements for similar 2D systems. For example, photoresponsivity calculations using DFT-GGA for MoS_2, WSe_2, and BP photodetectors closely matched experimental values, validating the reliability of this method for photocurrent predictions in 2D materials.[5-8]

(5) The computational efficiency of PBE allows us to handle the large supercells required for our device simulations while maintaining reasonable accuracy. Full GW+BSE calculations for our system would be computationally prohibitive without providing significant additional insights into the photocurrent characteristics.

(6) Our focus is primarily on the relative differences in photoresponse between different crystal orientations and variants, where systematic errors in absolute energies are less critical. In our device configuration, the photocurrent generation is mainly determined by:
- Carrier generation.
- Carrier separation by the built-in field.
- Carrier transport These processes are well-captured by our current computational approach, and the suppressed excitonic effects due to high doping and strong built-in field suggest that GW+BSE calculations would not significantly alter our main conclusions about the device's photoresponse characteristics.

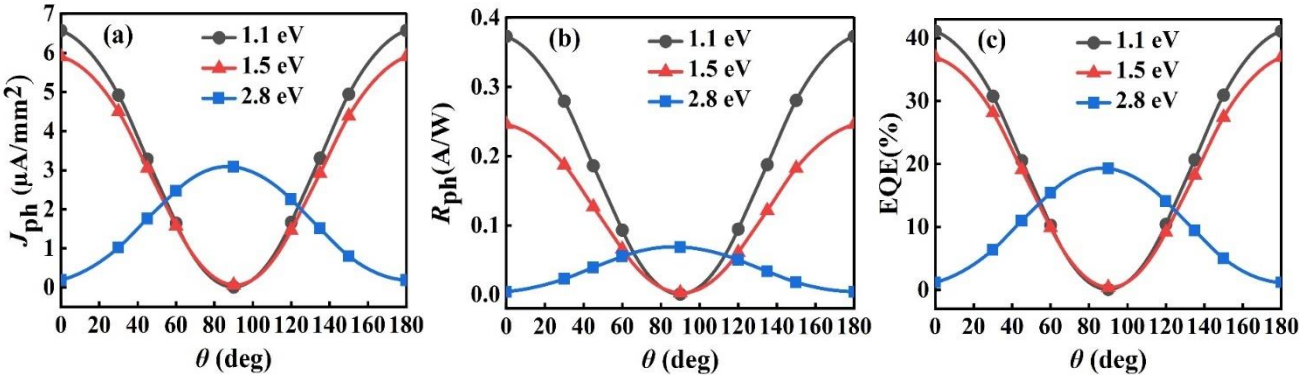

Fig. S5 (a) J_{ph}, (b) R_{ph}, and (c) EQE as functions of θ under photon energies of 1.1, 1.5, and 2.8 eV, with the linearly polarized light incident along the *a* directions for the O1 variant.

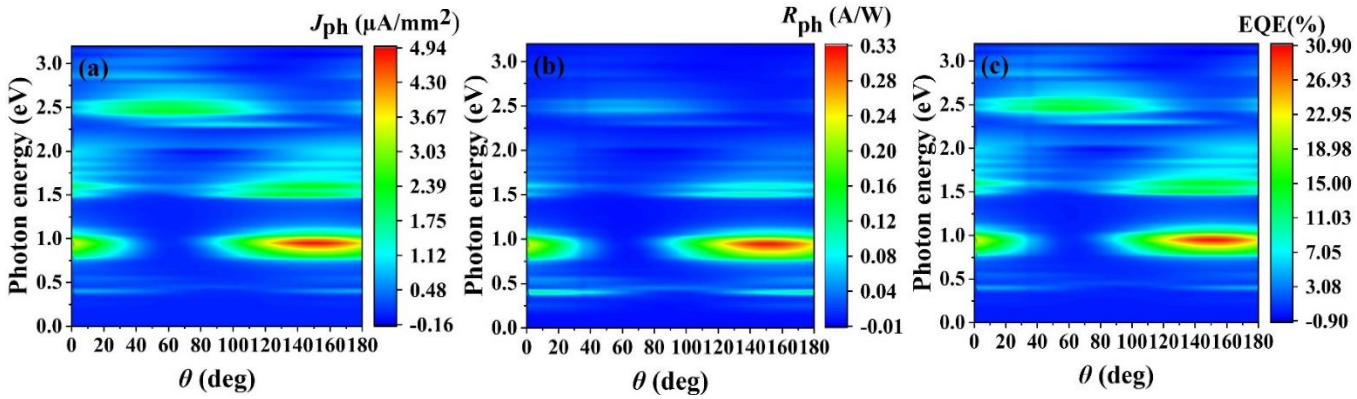

Fig. S6 (a) J_{ph}, (b) R_{ph}, and (c) EQE of the O3 variant with respect to photon energy and θ for the linearly polarized light incident along the *b* direction.

References

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