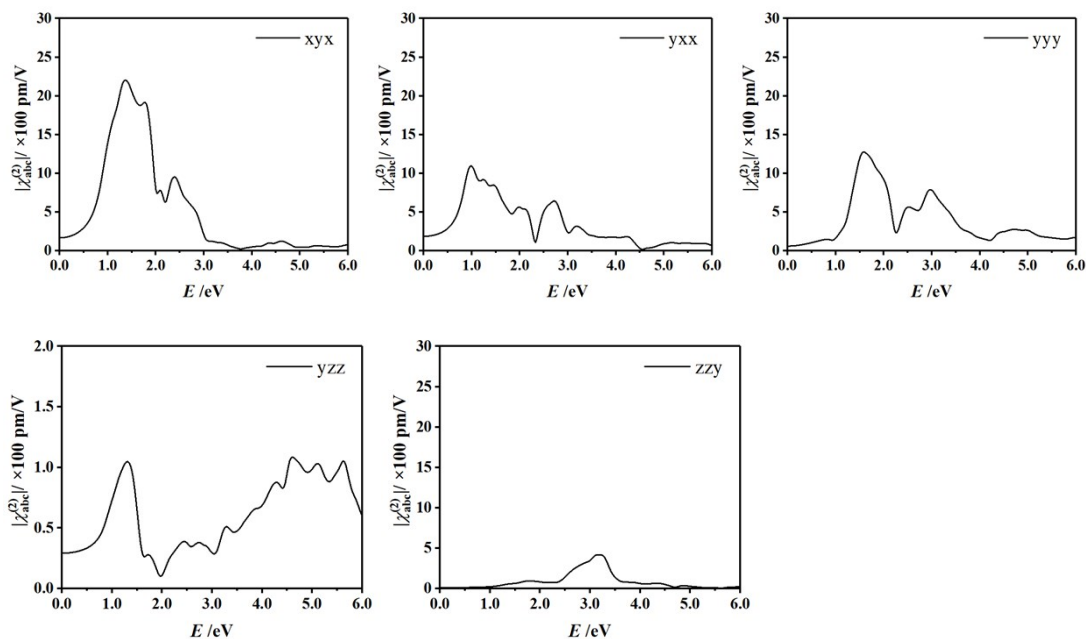


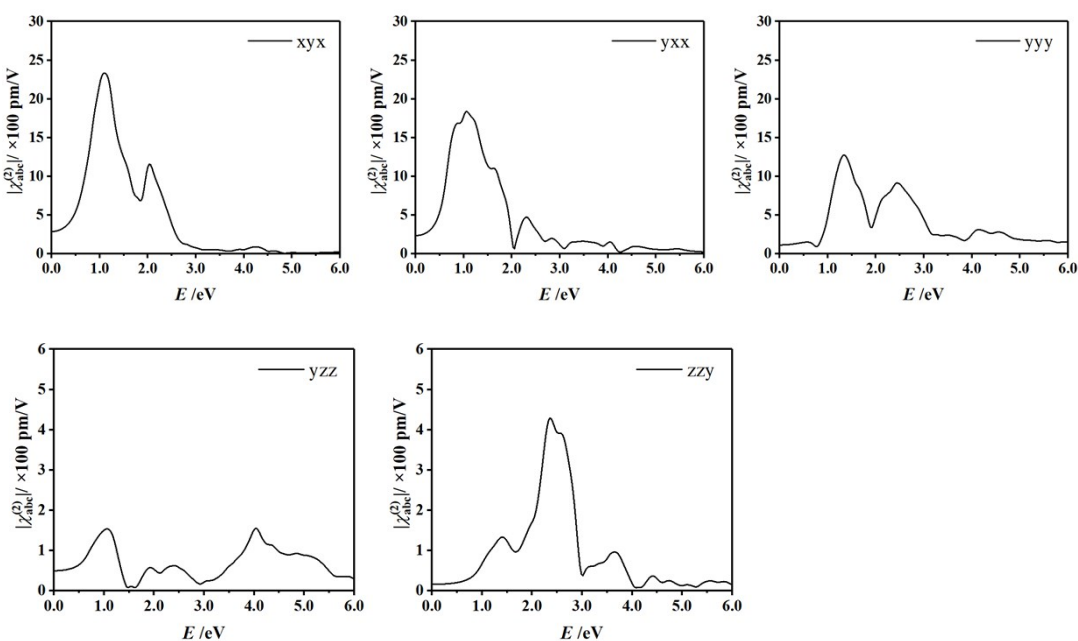
## Supplementary materials for the manuscript “Giant and anisotropic second harmonic generation of V-V binary phosphorene derivative with permanent dipole”

1. Nonzero components of the common 2D materials calculated using the method in the manuscripts, (in the unit of pm/V)

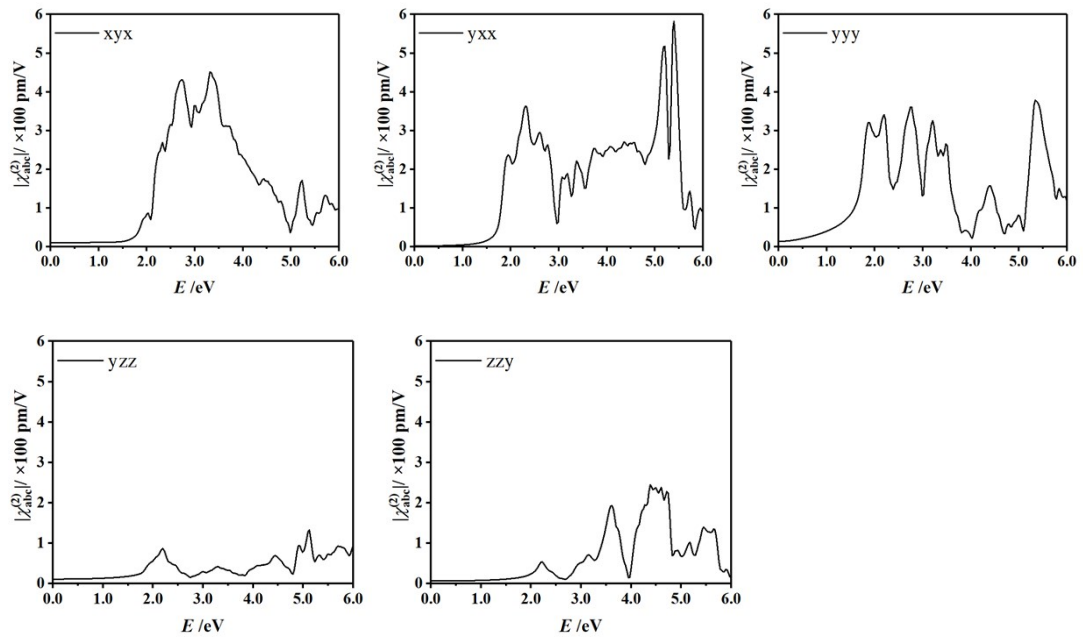
### Monolayer GeSe: 5 nonzero components



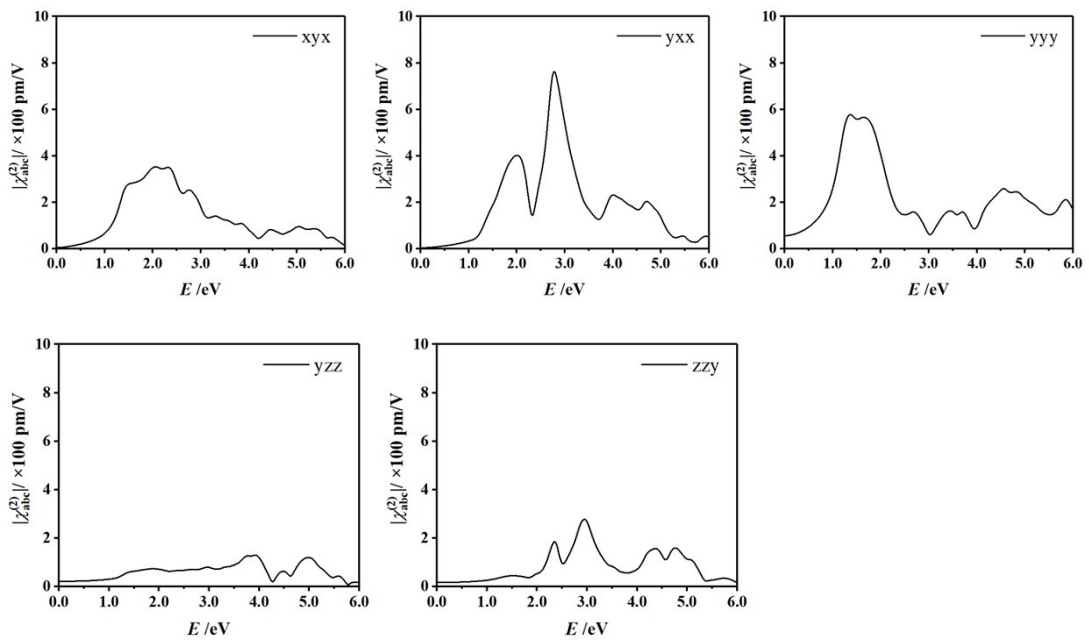
### Monolayer SnSe: 5 nonzero components



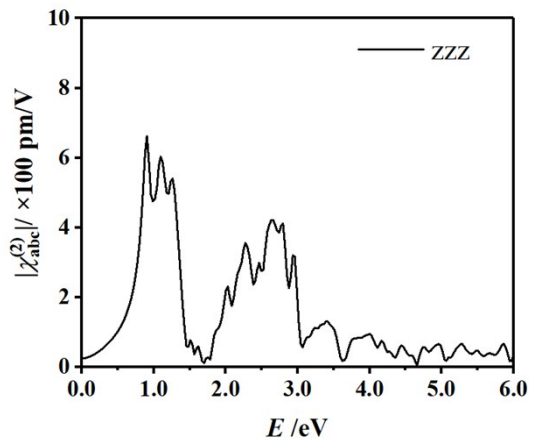
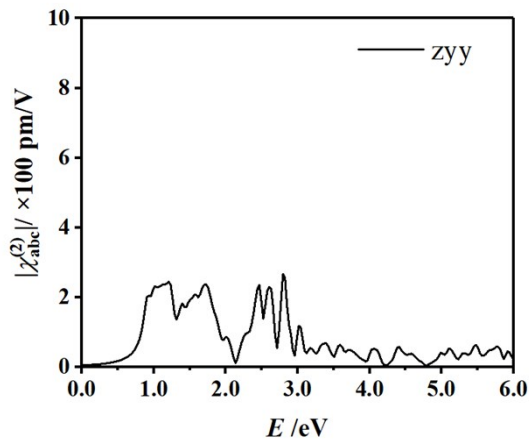
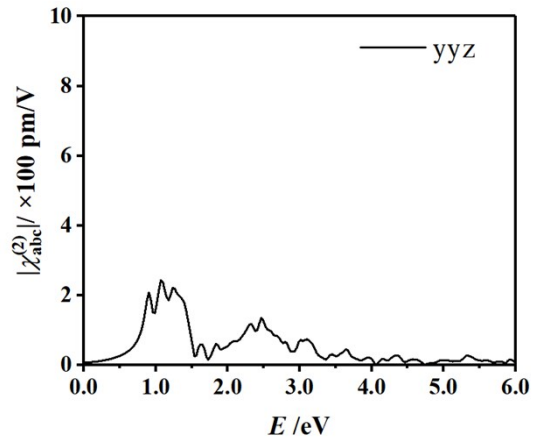
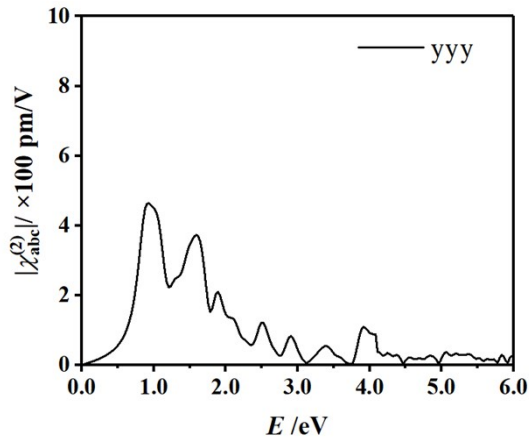
Monolayer GeS: 5 nonzero components



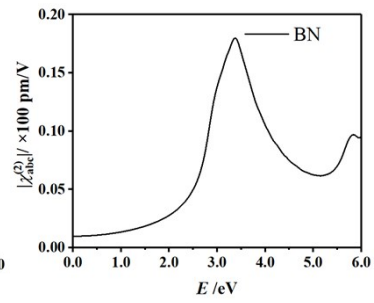
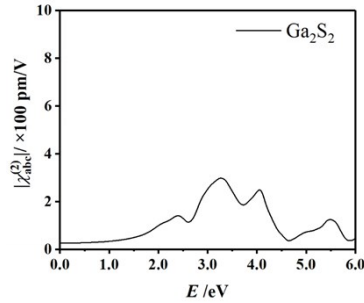
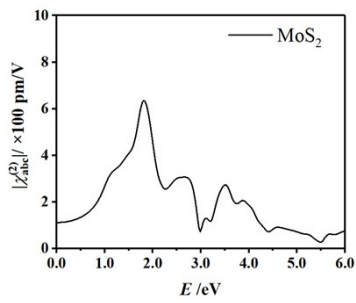
Monolayer SnS: 4 nonzero components



Monolayer MoSSe: 4 nonzero components



Monolayer MoS<sub>2</sub>/Ga<sub>2</sub>S<sub>2</sub>/h-BN: Only in-plane component  $\chi_{yyy}^{(2)}$



## 2. The phonon dispersion of $\alpha$ -PAs and $\alpha$ -PN materials

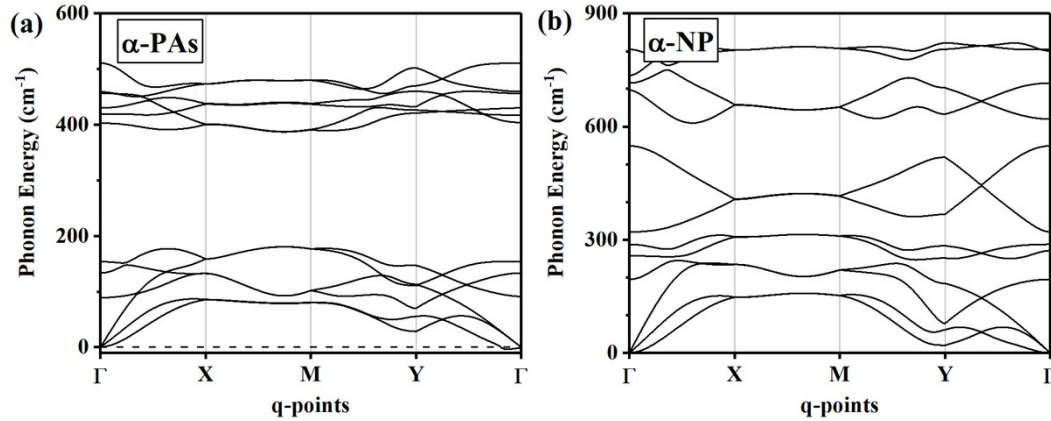


Figure S2 The phonon dispersion of  $\alpha$ -PAs and  $\alpha$ -PN materials

To perform the dynamical stability, we calculated the phonon dispersion of both  $\alpha$ -PAs and  $\alpha$ -PN materials by Quantum Espresso, using 45 Ry cutoff energy and  $6 \times 6 \times 1$  q-mesh and k-mesh. The phonon dispersions are shown below and added into supplementary information. No soft mode was found with phonon energies lower than 0 for monolayer NP. Although one of the branches (out of plane acoustical modes) becomes soft and get imaginary frequencies near  $\Gamma$  point for PAs, such instability may be an artifact related to mesh size in the calculations. It can be removed by defects or finite size effects.<sup>[1-2]</sup> As a result,  $\alpha$ -PAs and  $\alpha$ -PN materials are dynamically stable. This result agrees with Yu's work<sup>[3]</sup> and the supplementary information of Nie's work<sup>[4]</sup>, in which the phonon dispersion of 2D V-V binary materials with  $\alpha$  and  $\beta$  phase are listed.

### References

- [1] S. Cahangirov, M. Topsakal, E. Aktürk, H. Şahin, and S. Ciraci, *Physical Review Letters*, 2009, 102, 236804.
- [2] H. Şahin, S. Cahangirov, M. Topsakal, E. Bekaroglu, E. Akturk, R. T. Senger, and S. Ciraci, *Physical Review B*, 2009, 80, 155453.
- [3] W. Yu, C. Niu, Z. Zhu, X. Wang and W. Zhang, *Journal of Mater. Chem. C*, 2016, 4, 6581-6587.
- [4] Y. Nie, M. Rahman, P. Liu, A. Sidike, Q. Xia, and G. Guo, *Physical Review B*, 2017, 96, 075401.