

Electronic supplementary information for

**Stacking Induced Indirect-to-direct Bandgap Transition in Layered Group-IV  
Monochalcogenides for Ideal Optoelectronics**

Ji-Hui Yang<sup>a</sup> and Xin-Gao Gong<sup>ab</sup>

<sup>a</sup>Department of Physics, Key Laboratory for Computational Science (MOE), State Key  
Laboratory of Surface Physics, Fudan University, Shanghai 200433, China

Email: [jihuiyang2016@gmail.com](mailto:jihuiyang2016@gmail.com), [jhyang04@fudan.edu.cn](mailto:jhyang04@fudan.edu.cn)

<sup>b</sup>Collaborative Innovation Center of Advanced Microstructures, Nanjing 210093, Jiangsu, China

Table S1: lattice parameters and bandgaps of monolayer MX and MX bilayers with different stacking orders. ‘D’ represents direct bandgaps and ‘I’ represents indirect bandgaps.

Monolayer	a (Å)	a (Å)	Bandgap (eV)
GeS	3.657	4.495	2.41 (I)
GeSe	3.981	4.287	1.59 (D)
SnS	4.084	4.276	2.20 (I)
SnSe	4.293	4.383	1.32 (I)
Bilayer GeS			
AA	3.663	4.418	2.01 (I)
AA'	3.659	4.456	1.63 (I)
AB	3.671	4.432	2.16 (D)
AB'	3.651	4.497	2.10 (I)
AC'	3.675	4.393	1.64(I)
Bilayer GeSe			
AA	3.967	4.257	1.41 (D)
AA'	3.928	4.401	1.24 (I)
AB	3.947	4.347	1.63 (D)
AB'	3.929	4.398	1.58 (D)
AC'	3.968	4.287	1.32 (D)
Bilayer SnS			
AA	4.027	4.482	1.85 (I)
AA'	4.081	4.170	1.59 (I)
AB	4.041	4.359	1.93 (D)
AB'	4.012	4.447	1.56 (I)
AC'	4.066	4.237	1.29 (I)
Bilayer SnSe			
AA	4.266	4.417	1.40 (D)
AA'	4.272	4.333	1.12 (D)
AB	4.249	4.490	1.41 (D)
AB'	4.209	4.538	1.40 (I)
AC'	4.061	4.182	0.91 (I)

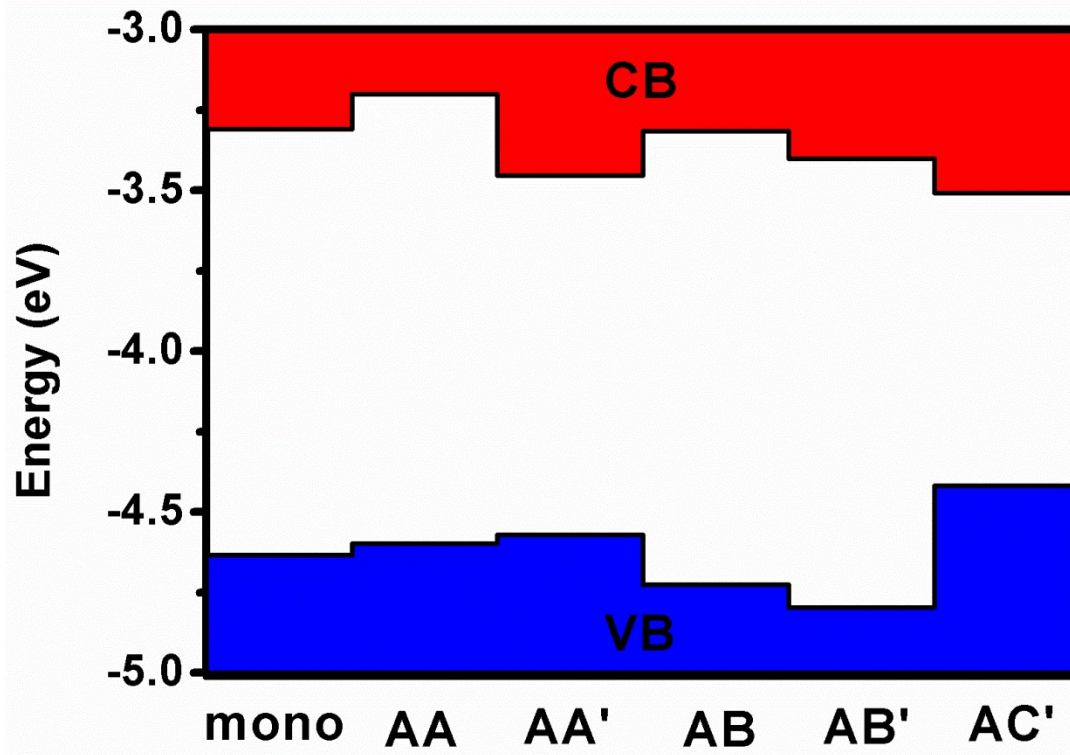


Figure S1. Band alignments between monolayer and bilayer SnSe with different stacking orders. The vacuum level is set as zero.

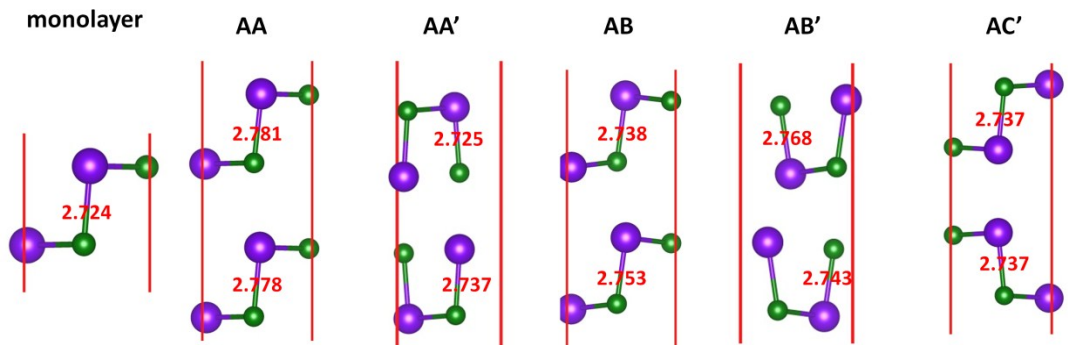


Figure S2. Vertical bond lengths in monolayer and bilayer SnSe with different stacking orders.

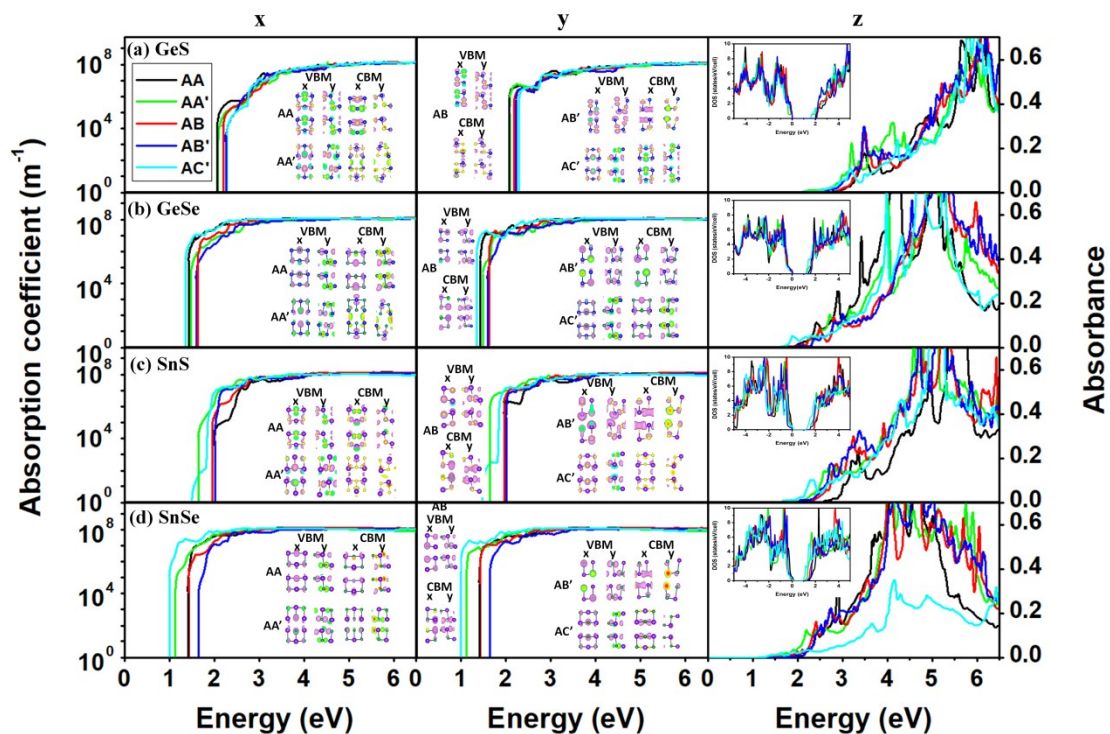


Figure S3. (a)-(d) Calculated in-plane optical absorption coefficients (for x and y panels) and out-of-plane absorbance (for z panel) of MX bilayers with different stacking orders. The insets of x and y panels indicate the partial charge densities of the VBM and CBM states with the contour set as  $0.005 \text{ e}/\text{\AA}^3$ . The insets of z panel show the density of states of MX bilayers.