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Supplementary material

Effect of stacking order and in-plane strain on the electronic properties of bilayer GeSe

Yuliang Mao,†[a] CongshenXu,[a] JianmeiYuan,[b] Hongquan Zhao[c]

[a] Hunan Key Laboratory for Micro–Nano Energy Materials and Devices, School of Physics and Optoelectronic, Xiangtan University, Hunan 411105, China.

[b] Hunan Key Laboratory for Computation and Simulation in Science and Engineering, School of Mathematics and Computational Science, Xiangtan University, Hunan 411105, China.

[c] Chongqing Institute of Green and Intelligent Technology, Chinese Academy of Sciences, 266 Fangzheng Ave, ShuiTu technology development zone, Beibei District, Chongqing, China.

† Corresponding author, E-mail address: ylmao@xtu.edu.cn

1. Phonon spectrum of AA-, AB-stackingbilayer GeSe

To further validate the dynamical stabilities of the bilayer GeSe for AA-, AB-stacking order, we calculate the phonon spectrum of them. As shown in Fig. S1, no imaginary frequency is found in the phonon spectrum, which proves that AA-, AB-stacking is stable.

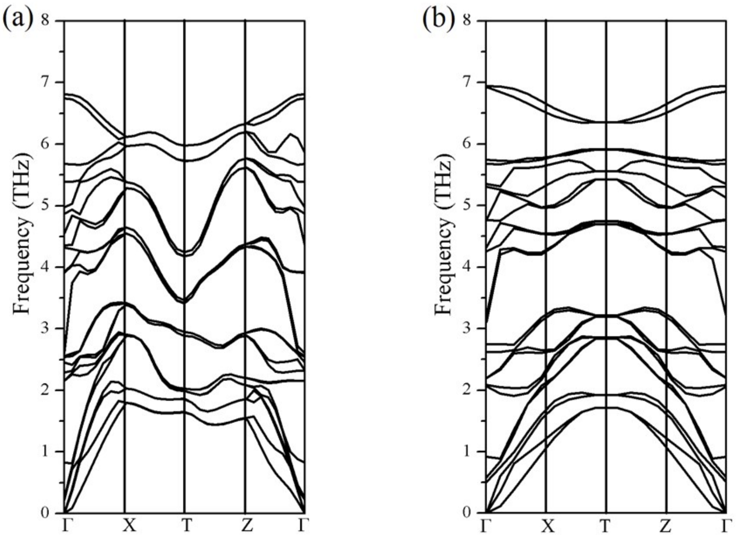

Fig. S1. Phonon spectrum of different stacking bilayer GeSe, (a) AA-stacking, (b) AB-stacking.

2. Tuning the band gap of AA-, AC-, AD-stacking bilayer GeSe by strain

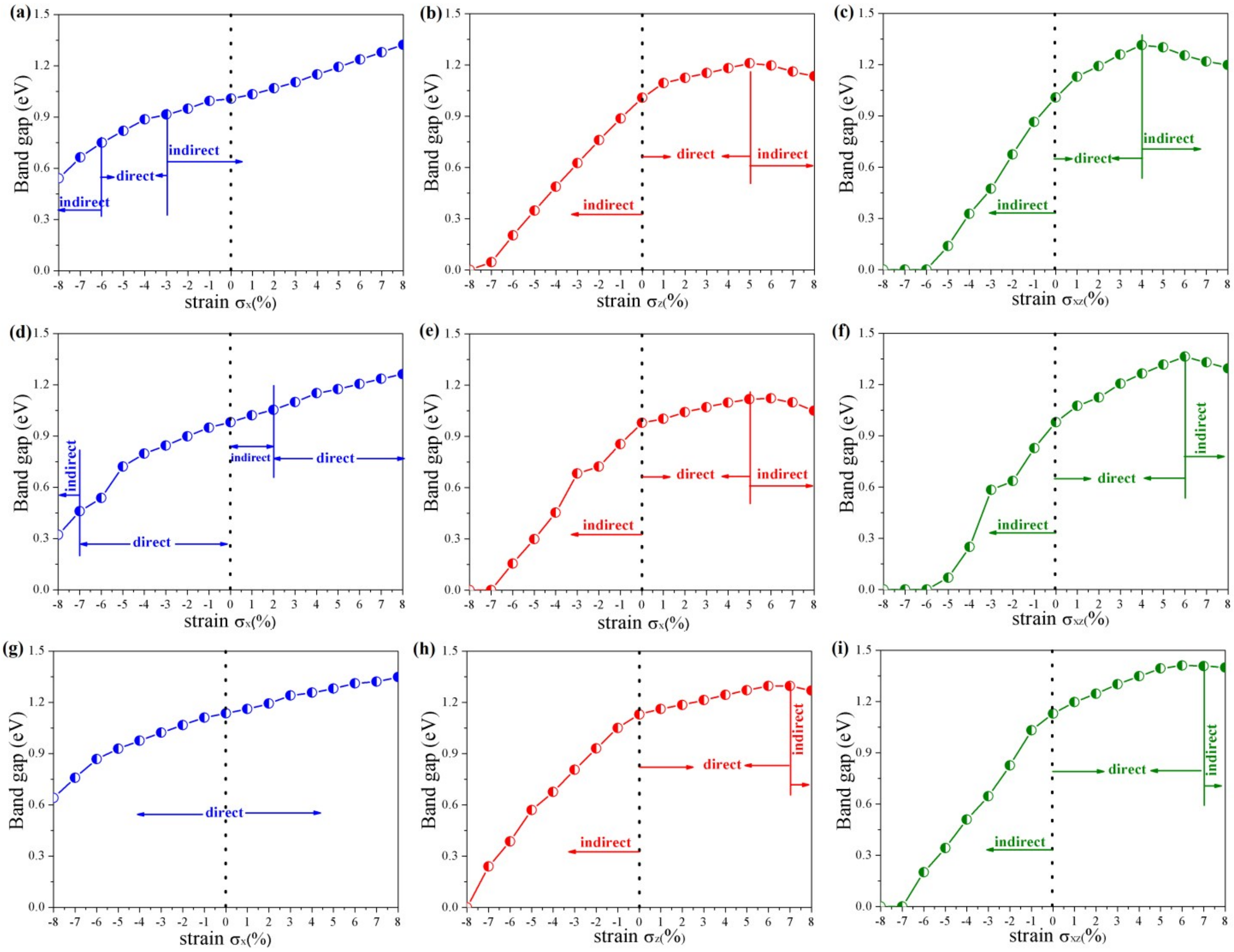

Fig. S2.The variation of band gap under in-plane strains.The value range of σ is from -8% to 8%. The positive and negative value corresponds to the stretching and compression strain, respectively. (a)-(c) indicate the results of AA stacking bilayer under strains along different directions, (a) σ_x, (b) σ_z, and (c) σ_{xz}, respectively. (d)-(f) indicate the results of the AC stacking bilayer under strains along different directions, (d) σ_x, (e) σ_z, and (f) σ_{xz}, respectively. (g)-(i) indicate the results of the AD stacking bilayer under strains along different directions, (g) σ_x, (h) σ_z, and (i) σ_{xz}, respectively.

In Fig. S2, we show our calculated results for the variationsband gap in the AA-, AC-, AD-stacking configurations under in-plane strain. As can be found in Fig. S2(a), when applying strain along x direction from -8% to 8%, the band gap of the AA-stacking bilayer GeSe is almost linear increased from 0.66 eV to 1.32 eV. Moreover, when the AA-stacking structure is stretched along x direction, the AA-stacking configuration maintains the feature of indirect band gap. However, if the compressive strain from -6% to -3% along x direction is applied, the indirect band gap of AA-stacking configuration is tuned to direct band gap. For the strain along z direction in the AA-stacking configuration, as can be found in Fig. S2(b), when compressive strain is applied, the band gap of AA-stacking is linear increased and is always kept indirect band gap. However, when AA-stacking configuration is stretched along z direction from 0% to 5%, the band gap of AA-stacking keeps direct band gap and is increased from 0.94 eV to 1.21 eV. While for the stretched strain form 5% to 8% along z direction, the band gap is decreased from 1.21 eV to 1.13 eV and is changed to indirect band gap. The variation of band gap of AA-stacking bilayer GeSe under biaxial strains almost similar with that under the strain along z direction. Their slight difference is that the range of increased direct band gap is from 0% to 4% when applying biaxial strain.

In Fig. S2(d, e, f), we show the variations of band gap of AC-stacking configuration under in-plane strain. As shown in Fig. S2(d), the band gap of AC-stacking is increased from 0.46 eV to 1.26 eV when the strain from -8% to 8% along x direction is applied. In addition, the inherent indirect band gap of AC-stacking is converted to direct band gap when applying -5%-0% compressed strain or 3%-8% stretched strain. While applying stretched strain along x direction from 0%-2% or compressed strain from -6% to -8%, the AC-stacking is indirect band gap. For the z-uniaxial strain, the variation of band gap is very similar with that in AA-stacking GeSe bilayer, as can be found in Fig. S2(e). The band gap of AC-stacking is also linear increased when applying strain from -8% to 5% and is decreased under strain from 5% to 8%. The type of band gap is indirect when applying strain from -8%-0% and 5%-8%. However, it will be changed to direct band gap when applying strain from 0%-5%. As shown in Fig. S2(f), the variation of band gap under the applied biaxial strain in AC-stacking is almost similar with that under the applied strain along the z direction. The band gap is increased from 0 eV to 1.36 eV when applying biaxial strain from -8% to 6% and is later slightly decreased from 1.36 eV to 1.29 eV when biaxial strain is increased from 6% to 8%. The direct band gap of AC-stacking appears under stretched strain from 0% to 6%. When the biaxial strain is applied from -8% to 0% and 6% to 8%, the AC-stacking keeps indirect band gap.

The variations of band gap of AD-stacking configuration under in-plane strain are plotted in Fig. 2(g, h, i). In Fig. S2(g), we find that the band gap of AD-stacking is almost linearly increased and maintains direct when applying strain along x direction from -8% to 8%, which is very similar with that in AB-stacking. For the single axial strain along z direction, the variation of band gap is also similar with that in AB-stacking. As can be found in Fig. S2(h), the band gap is increased linearly from 1.13 eV to 1.41 eV when applying stretched strain from 0% to 7% and maintains direct band gap in this range. However, when the stretched strain continues to be increased from 7% to 8%, the band gap is decreased and has been changed to indirect. While the compressed strain is applied from 0% to -8%, the band gap is rapidly decreased to 0 eV and always keeps indirect band gap under the compressed strains. Under biaxial strain, the tuning situation of band gap in AD-stacking GeSe bilayer is similar with that in the case under the tuning along z-direction.

In summary, our results show that the inherent indirect band gap of AA-stacking can be tuned to direct under small strain along biaxial or unaxial z direction. However, the tuned range of the direct band gap is narrow under the applying in-plane strains. Moreover, when the stretched strain is larger than 5% along zigzag direction and the stretched strain is larger than 4% along biaxial direction, the band gap is found decreasing. The variation of band gap in AC-, AD-stacking is similar with that in AB-stacking when applying in-plane strain. The transition between the direct to indirect band gap is observed under some special values of strain, which does not exist in the case of AB-stacking. For example, in the AC-stacking case, the transition from direct to indirect band gap is always found only if the compressive uniaxial strain along the armchair direction is larger than -7%.

3. about the van der Waals functionals

Table S1. The lattice constants and band gap of AB-stacking bilayer GeSe obtained from different van der Waals (vdWs) functionals. The symbol * indicates the direct band gap, otherwise it is indirect. The values of previous reports obtained from PBE [Ref. S1], the experimental data for bulk [Ref. S2], as well as the data from Ref. S3 are given for comparison.

Functional	a(Å)	b(Å)	Band gap(eV)
PBE	4.31	3.96	1.03*
PBE [Ref. S1]	4.31	3.97	1.02*
vdW-DF	4.74	3.91	1.49
vdW-DF2	4.82	3.96	1.55
DFT-D2	4.40	3.88	0.98
TS	4.37	3.94	1.06*
Bulk(Exp) [Ref. S2]	4.37	3.81	1.08
Exp. [Ref. S3]			*

Before our calculations, we have tested the accuracy of the vdW types to obtain the suitable lattice constants and band gap of AB-stacking GeSe. The obtained results from using different vdW-correlation functions such as vdW-DF [S4], vdW-DF2 [S5], semiempirical Grimme's DFT-D2 [S6] and Tkatchenko-Scheffler (TS) dispersion corrections [S7], are summarized in Table S1. Without vdW-correction, the obtained lattice constants are a=4.31 Å and b =3.96 Å, respectively. We also obtained a direct band gap with the energy gap of 1.03 eV, which is agree with previous report [S1]. When the vdW-DF and vdW-DF2 functionals are used, the lattice constant has a large change comparing to the experimental data of bulk GeSe. Moreover, the type of band gap is different with reported experimental result [S3] in bilayer GeSe, which possesses a direct band gap. The lattice constants obtained from DFT-D2 and TS functionals are close to the experimental data of bulk GeSe. However, the type of band gap is indirect when using DFT-D2 function. By our test, only the calculation using TS functional predicts the direct energy gap. Hence, we choose TS functional to discuss the stacking and strain effect on the studied GeSe bilayers.

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