Supplementary material

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

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1. Phonon spectrum of AA-, AB-stacking bilayer 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

![Graphs showing band gap variation under in-plane strains](image)

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) \( \sigma_x \), (b) \( \sigma_z \), and (c) \( \sigma_{xz} \), respectively. (d)-(f) indicate the results of the AC stacking bilayer under strains along different directions, (d) \( \sigma_x \), (e) \( \sigma_z \), and (f) \( \sigma_{xz} \), respectively. (g)-(i) indicate the results of the AD stacking bilayer under strains along different directions, (g) \( \sigma_x \), (h) \( \sigma_z \), and (i) \( \sigma_{xz} \), respectively.

In Fig. S2, we show our calculated results for the variation of band 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
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 $a = 4.31 \text{ Å}$ and $b = 3.96 \text{ Å}$, 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.

References


