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# Supporting Information

## Bandgap Transition and Second-Harmonic Generation in ABC and ACB stacked $\gamma$ -Trilayers of Janus-Ga<sub>2</sub>SeTe

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### Note 1. Stacking orders of GaSe, GaTe, and Ga<sub>2</sub>SeTe

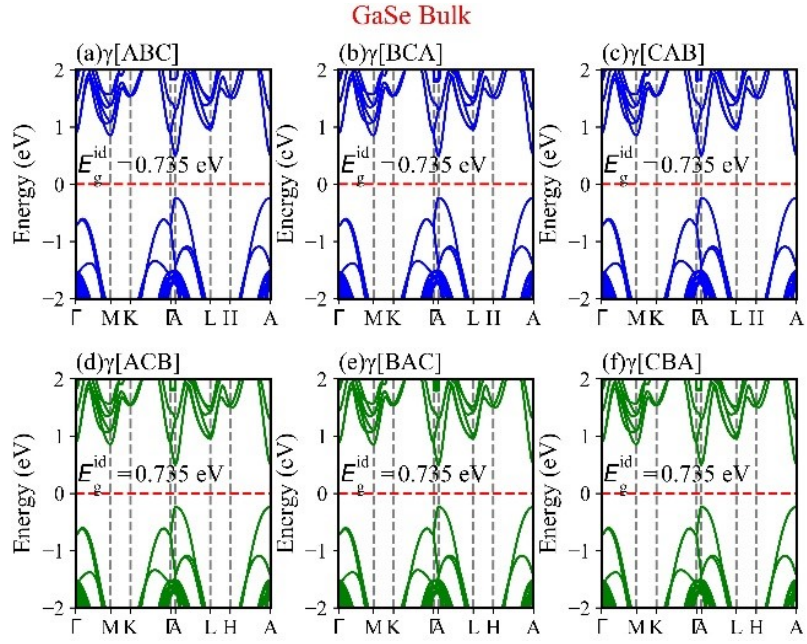
As shown in **Figs. S1** and **S2**, the band structures of bulk GaSe and GaTe show the negligible dependence on their stacking sequences, where both systems maintain the indirect bandgaps—approximately 0.74 eV for GaSe and 0.25 eV for GaTe—across all six configurations. By contrast, bulk Ga<sub>2</sub>SeTe exhibits the clear stacking-dependent electronic structures, which can be classified into two distinct phases of  $\gamma$ [ABC]-type (including equivalent [BCA] and [CAB]) and  $\gamma$ [ACB]-type (including equivalent [BAC] and [CBA]) (**Fig. S3**). These phases differ in their bandgaps: the  $\gamma$ [ABC] phase has a direct bandgap of 0.34 eV, while the  $\gamma$ [ACB] phase possesses a larger direct bandgap of 0.41 eV.

Importantly, the same trend persists in the few-layer limit (**Figs. S4-6**). PBE calculations confirm that GaSe, GaTe, and Ga<sub>2</sub>SeTe trilayers show similar stacking-related band-structure variations as their bulk counterparts. Therefore, these results demonstrate that the influence of  $\gamma$  phase stacking sequence on electronic properties is pronounced in non-centrosymmetric materials, such as Janus-Ga<sub>2</sub>SeTe, whereas centrosymmetric systems like GaSe and GaTe remain largely unaffected.

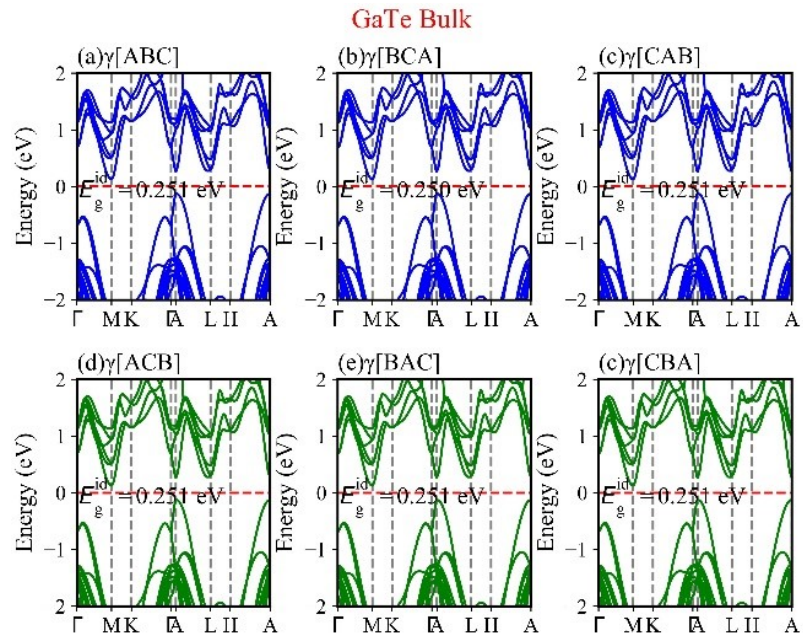
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\*Corresponding authors.

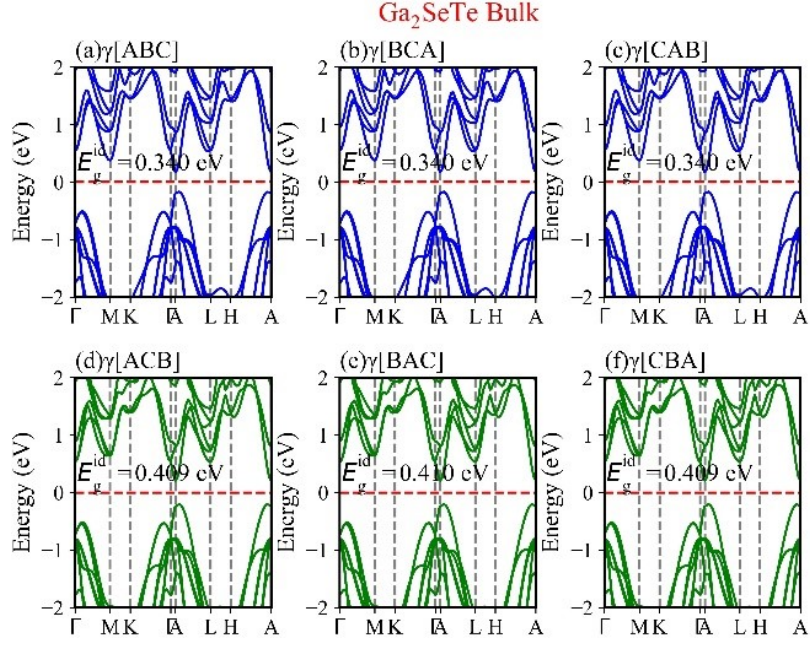
E-mails: [iamleili@nwpu.edu.cn](mailto:iamleili@nwpu.edu.cn) (L. Li); [jzshang@cczu.edu.cn](mailto:jzshang@cczu.edu.cn) (J. Shang).



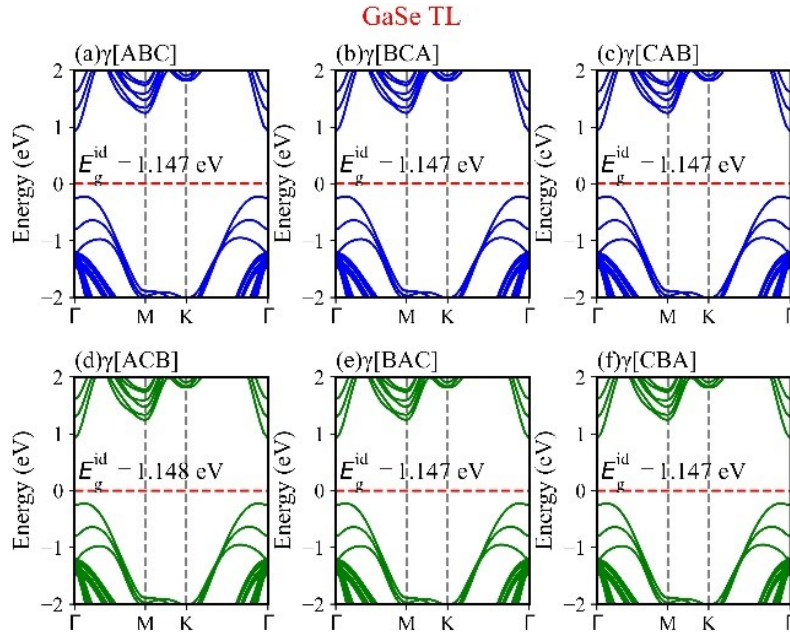
**Fig. S1** Band structures of GaSe bulk with different stacking orders (PBE method).



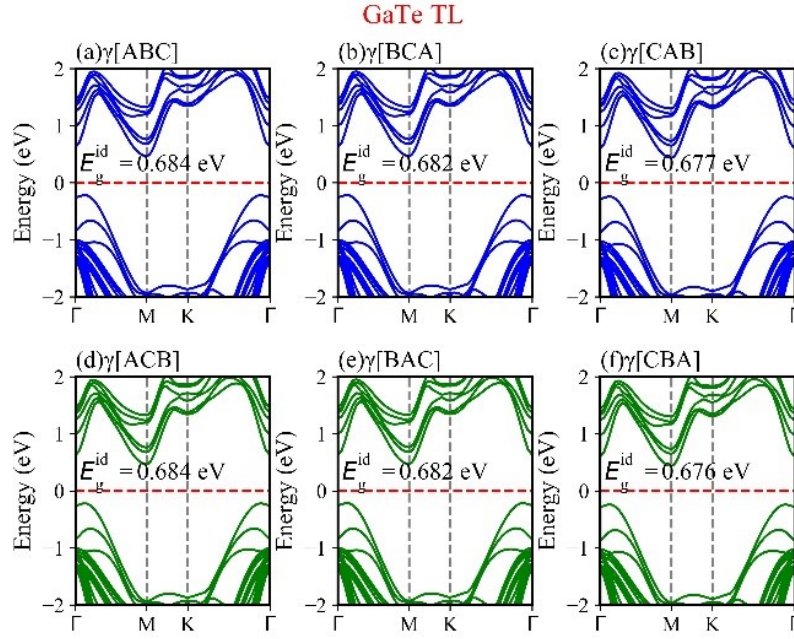
**Fig. S2** Band structures of GaTe bulk with different stacking orders (PBE method).



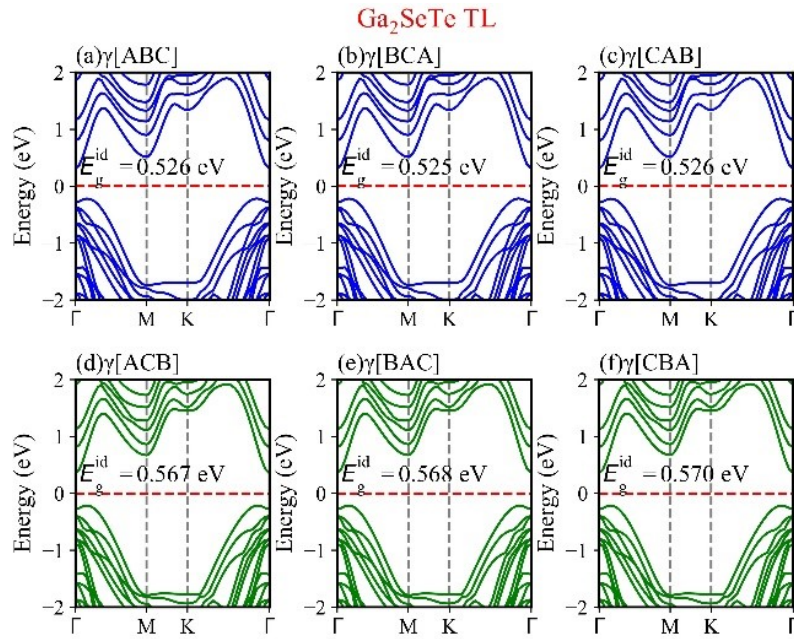
**Fig. S3** Band structures of Ga<sub>2</sub>SeTe bulk with different stacking orders (PBE method).



**Fig. S4** Band structures of GaSe TLs with different stacking orders (PBE method).



**Fig. S5** Band structures of GaTe TLs with different stacking orders (PBE method).



**Fig. S6** Band structures of Ga<sub>2</sub>SeTe TLs with different stacking orders (PBE method).