

Supplementary Material for “ $\alpha_n h_m$ -GeSe: A Multifunctional Semiconductor Combining Auxeticity and Piezoelectricity ”

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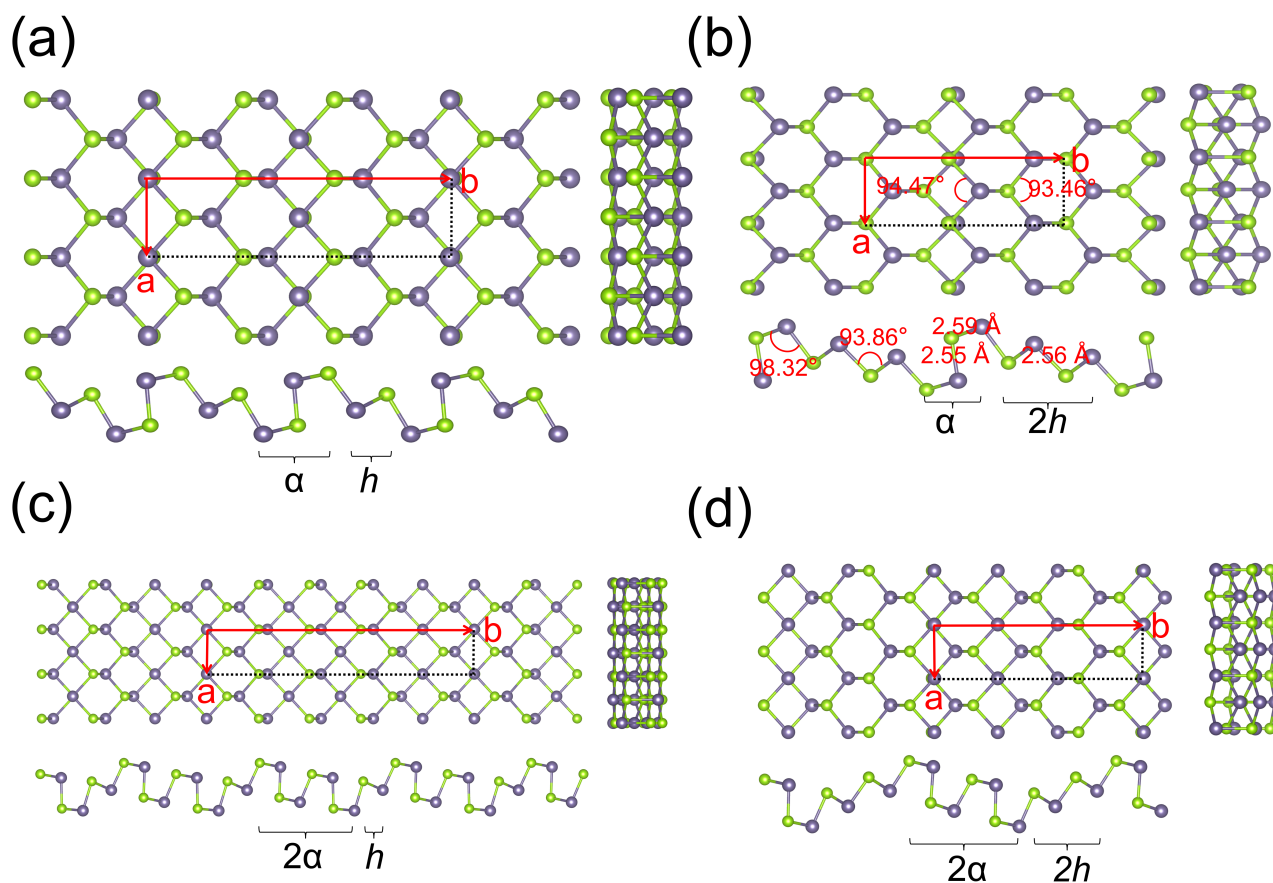


FIG. S1: (a-d) Top and side view of $\alpha_1 h_1$ -GeSe, $\alpha_1 h_2$ -GeSe, $\alpha_2 h_1$ -GeSe and $\alpha_2 h_2$ -GeSe .

Fig.S1(a) exhibits the conventional cell of $\alpha_1 h_1$ -GeSe, which has twelve atoms in a cell. The primitive cell of $\alpha_1 h_1$ -GeSe has six atoms and space group Cm (No.8). Fig.S1(c) exhibits

the conventional cell of α_2h_1 -GeSe, which has twenty atoms in a cell. The primitive cell of α_1h_1 -GeSe has ten atoms and space group Cm (No.8). Fig.S1(d) exhibits the primitive cell of α_2h_2 -GeSe has eight atoms and space group Pm (No.6).

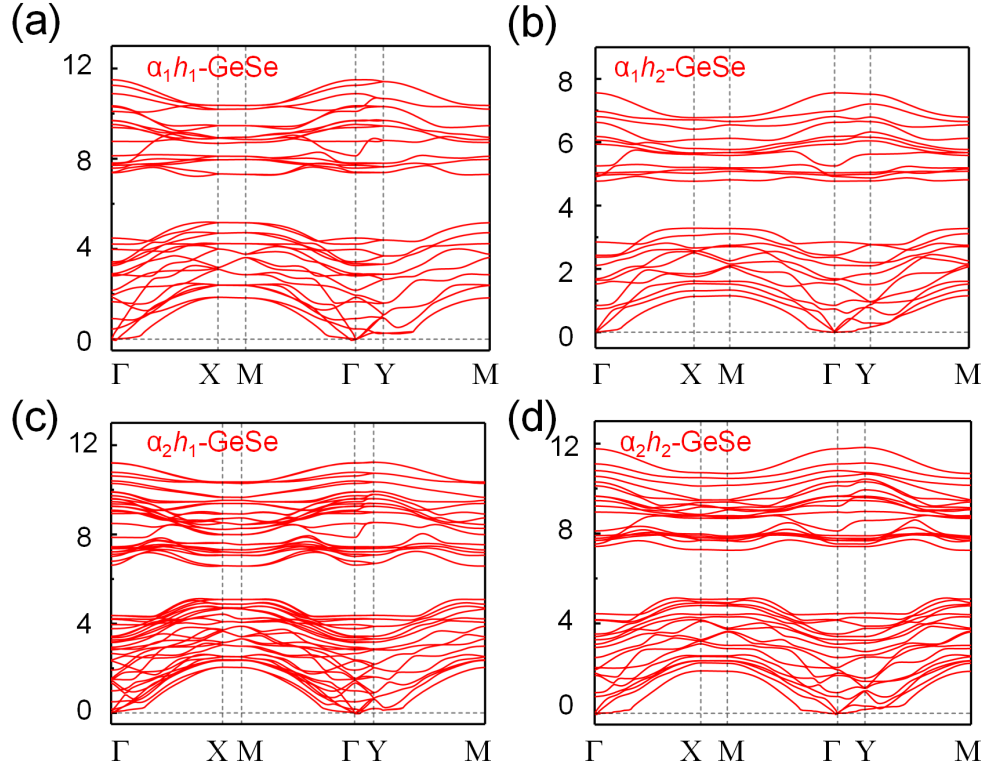


FIG. S2: (a-d) The phonon dispersion of α_1h_1 -GeSe, α_1h_2 -GeSe, α_2h_1 -GeSe and α_2h_2 -GeSe.

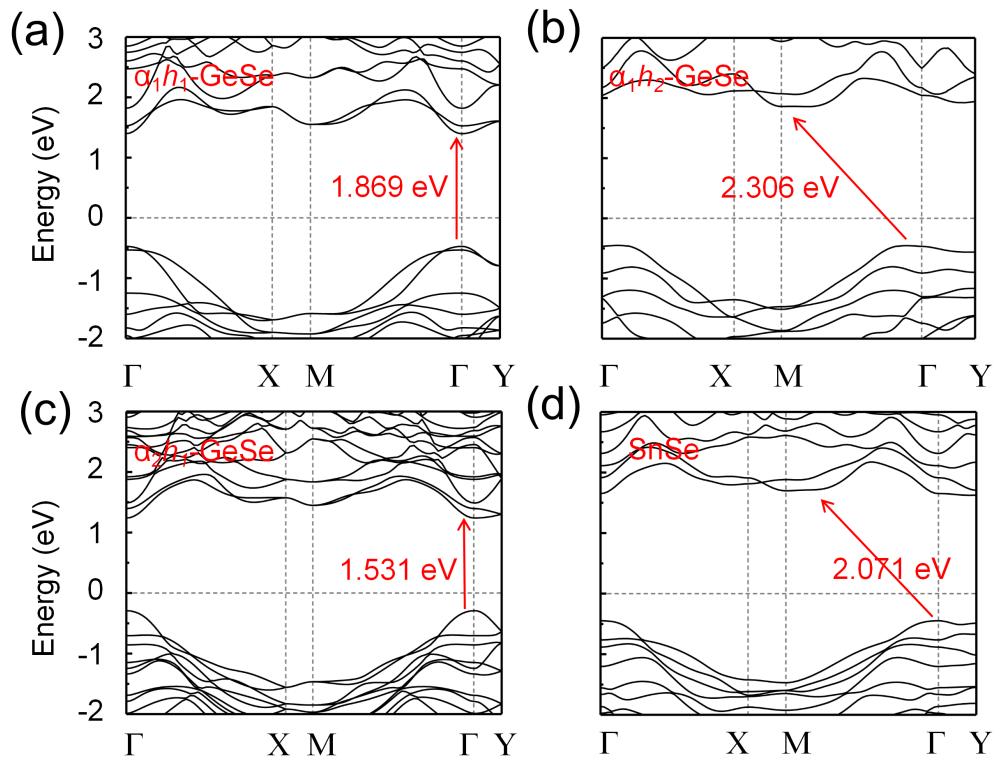


FIG. S3: (a-d) The band structure of $\alpha_1 h_1$ -GeSe, $\alpha_1 h_2$ -GeSe, $\alpha_2 h_1$ -GeSe and $\alpha_2 h_2$ -GeSe at the HSE06 level.

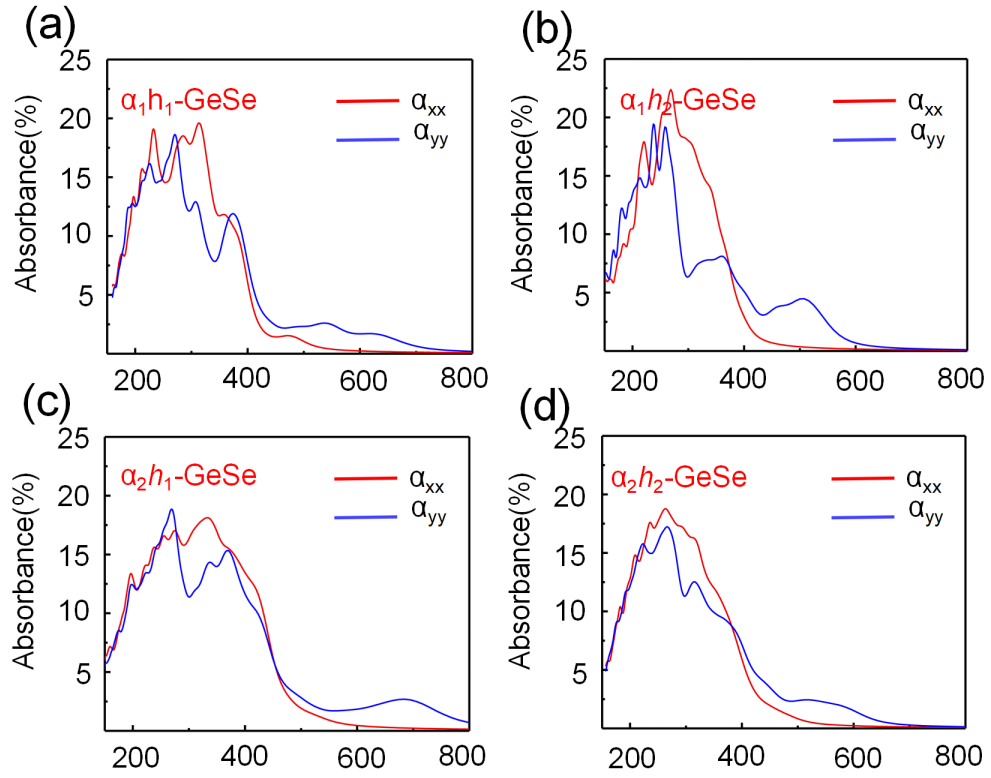


FIG. S4: (a-d) The light absorption of $\alpha_1 h_1$ -GeSe, $\alpha_1 h_2$ -GeSe, $\alpha_2 h_1$ -GeSe and $\alpha_2 h_2$ -GeSe at the HSE06 level.

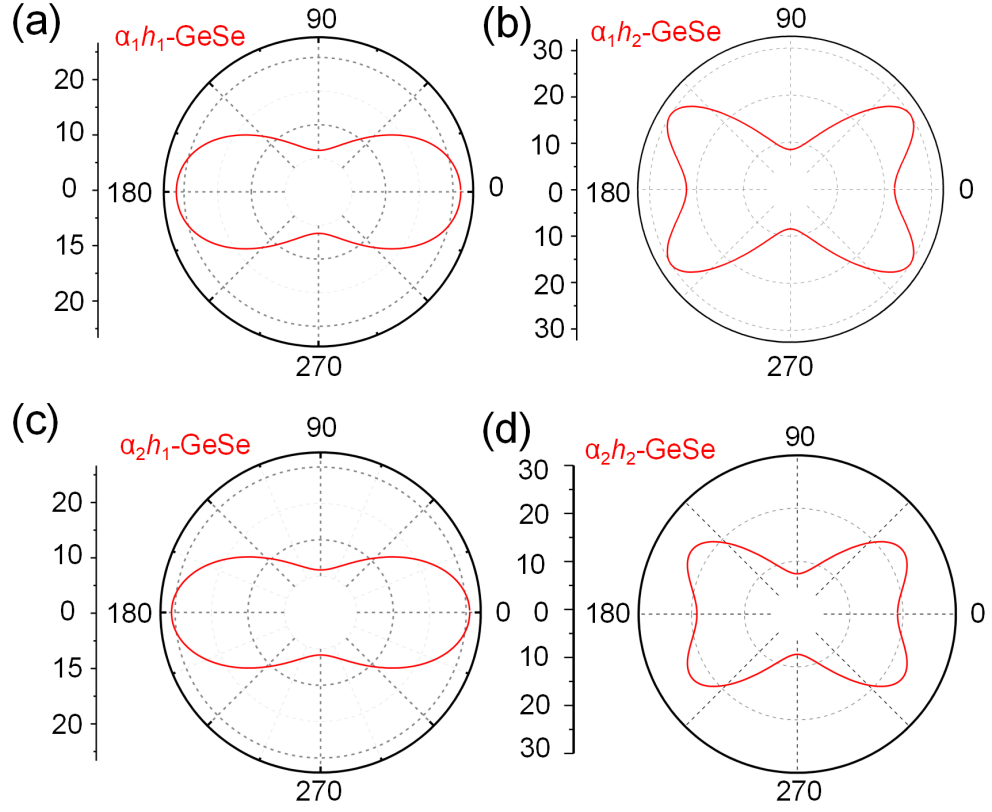


FIG. S5: (a-d) The Young's modulus $Y(\theta)$ of $\alpha_1 h_1$ -GeSe, $\alpha_1 h_2$ -GeSe, $\alpha_2 h_1$ -GeSe and $\alpha_2 h_2$ -GeSe.

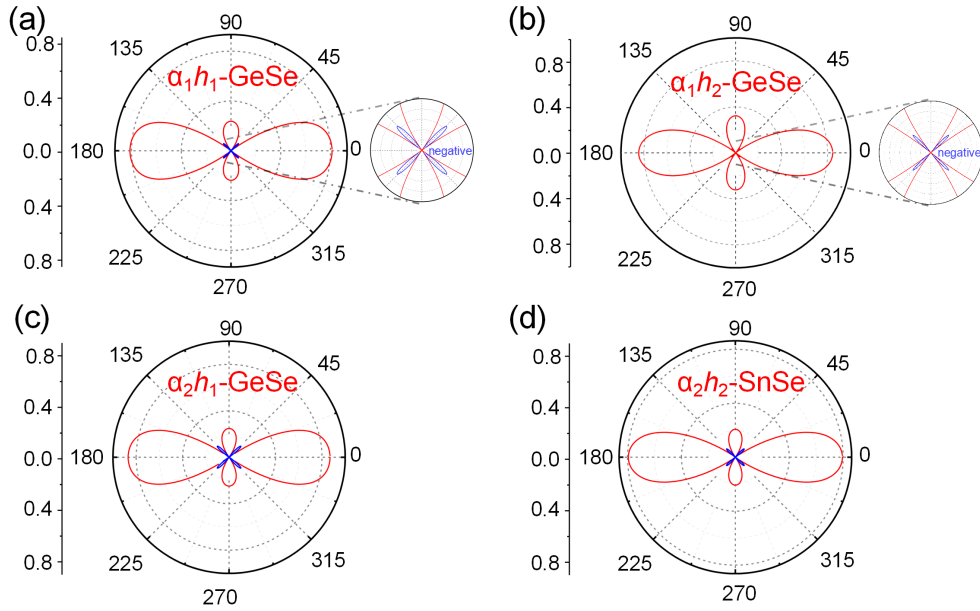


FIG. S6: (a-d) The Poisson's ratio $\nu(\theta)$ of $\alpha_1 h_1$ -GeSe, $\alpha_1 h_2$ -GeSe, $\alpha_2 h_1$ -GeSe and $\alpha_2 h_2$ -GeSe.

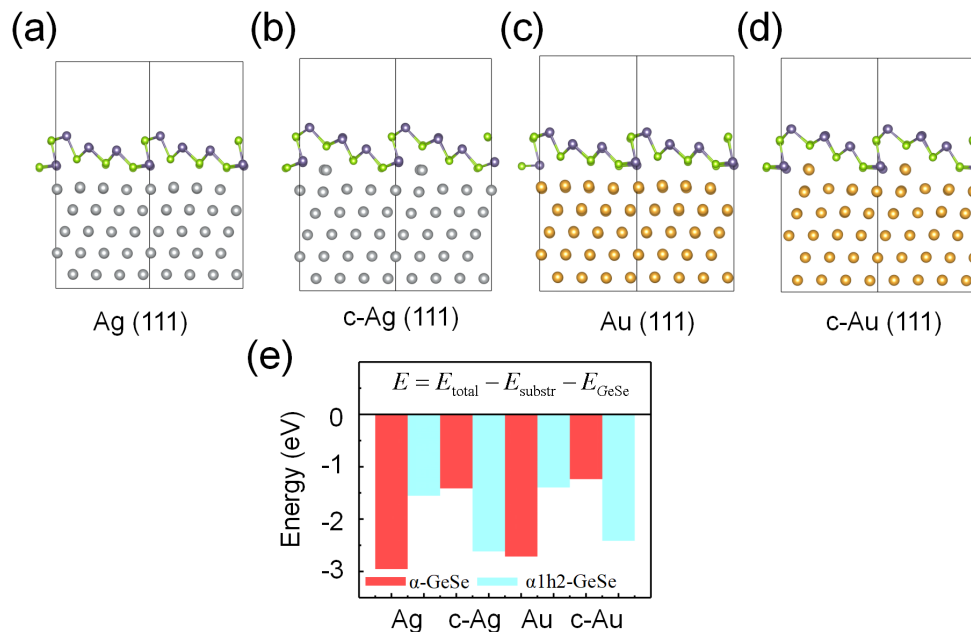


FIG. S7: (a-d) Atomic structures of $\alpha_1 h_2$ -GeSe on smooth substrate and uneven substrates are drawn. (e) The formation energies of pure phases α -GeSe and mixed phases $\alpha_1 h_2$ -GeSe on different substrates.

Here, we will discuss the possibility of synthesizing mixed phase $\alpha_1 h_2$ -GeSe on a substrate. Researchers have synthesized two-dimensional monolayers of α -phase and h -phase black phosphorus on the smooth substrates of Au(111) and Ag(111) surfaces^{1,2}. The two substructures of mixed phase $\alpha_1 h_2$ -GeSe share the same structure as α -phase and h -phase black phosphorus, which motivates the exploration of synthesis on the Au(111) and Ag(111) surfaces. We place the pure phase α -GeSe and the mixed phase $\alpha_1 h_2$ -GeSe on smooth Au and Ag substrates to calculate the formation energy. We find that the formation energy of pure phase α -GeSe on the smooth substrate is significantly lower than the binding energy of the mixed phase, leading to a high probability that atomic deposition on the substrate will result in the formation of pure phase α -GeSe.

Based on previous research³, we found that substrates with uneven surfaces are more promising for synthesizing materials with significant fluctuations. Therefore, we removed the top layer of atoms from the Au and Ag substrates to create uneven substrates, referred to as c-Au and c-Ag. We further calculated the formation energy on the uneven substrates and discovered that the formation energy of the mixed phase is lower on the uneven substrate compared to the pure phase, indicating that the uneven substrate is more favorable for

material synthesis.

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