Supplementary Information

Enhancing Optoelectronic Properties of SnS via Mixed Phase Heterostructure Engineering

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1. Convergence of *k*-grid and lattice parameters

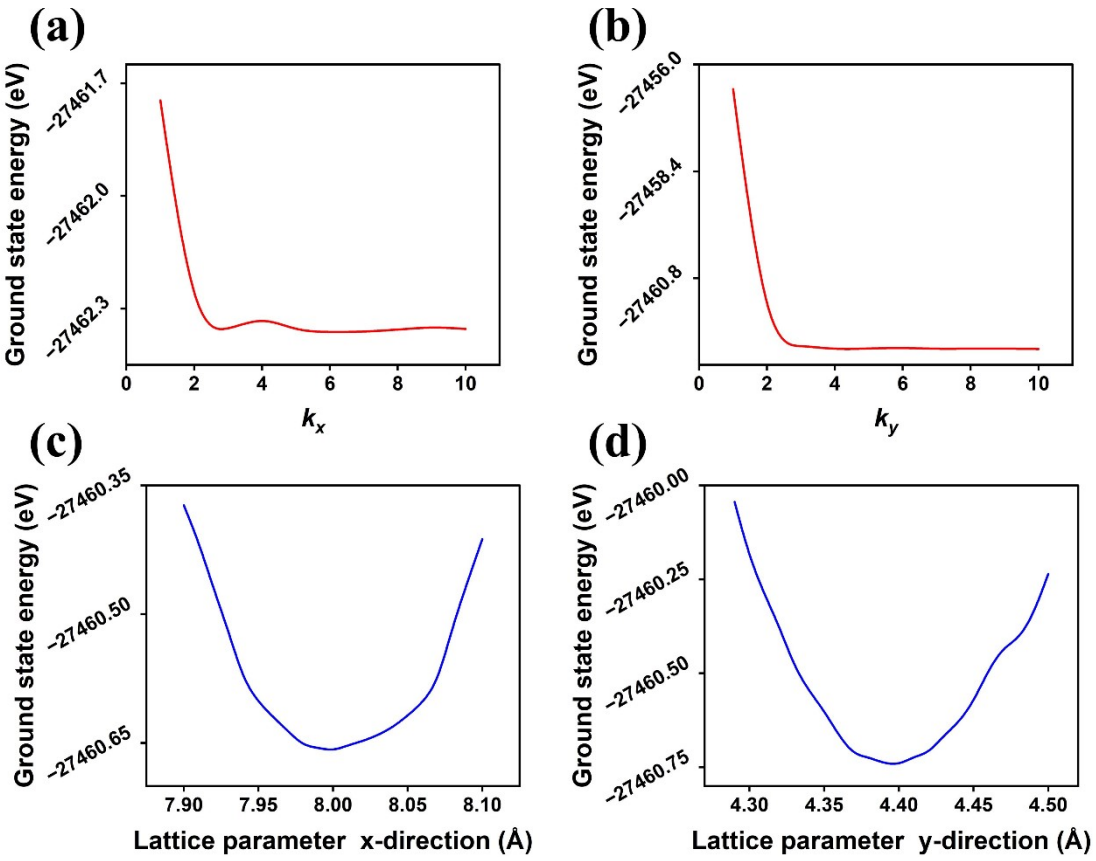

Figure S2: Convergence test of **(a,b)** lattice parameters vs ground state energy, and **(c,d)** *k*-grid vs ground state energy.

2. Schematic illustrating the modelling of the mixed phase SnS/*h*-BN heterostructure

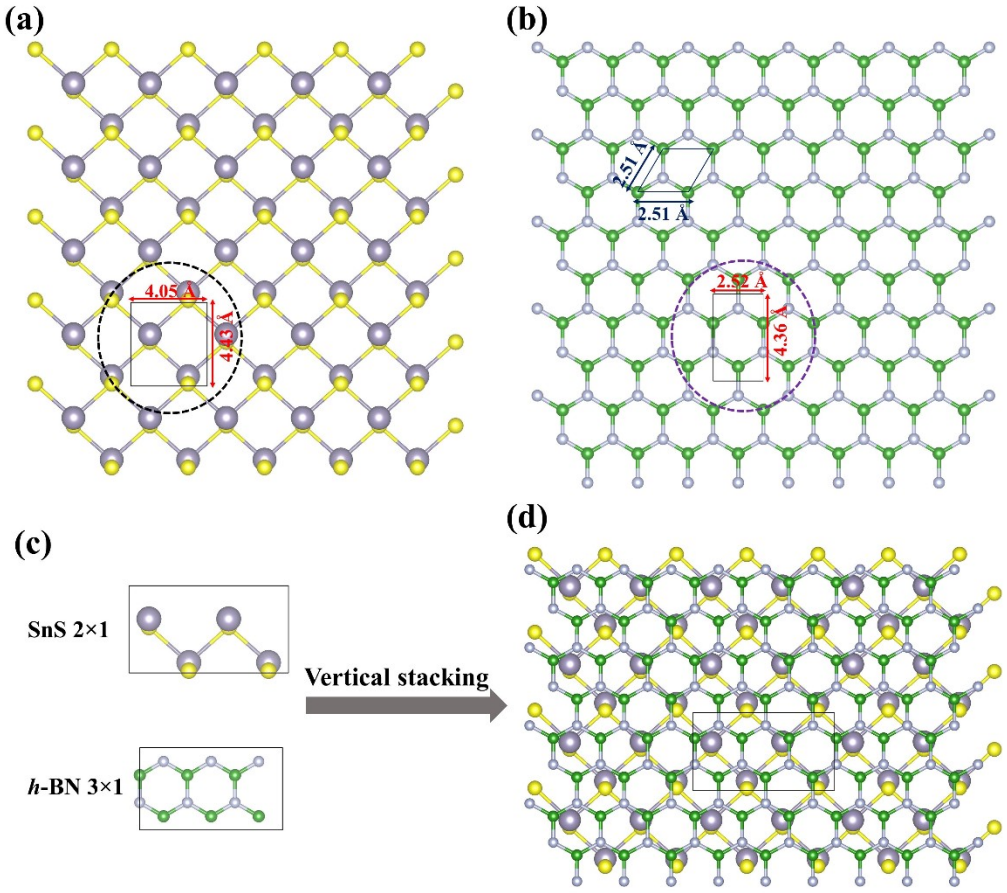

Figure S2: Schematic representing the modelling process of the SnS/*h*-BN heterostructure.

The following transformation matrix was used to model the orthorhombic unit cell of *h*-BN.

$$\begin{pmatrix} 1 & -1 & 0 \\ 1 & 1 & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

3. Table S3: Lattice parameters of the monolayer supercells and the strain experienced by them within the heterostructure

		SnS		*h*-BN		Heterostructure
Cell size		**1×1**	**2×1**	**1×1**	**3×1**	**1×1**
Lattice parameters	**a**	4.05 Å	8.1 Å	2.52 Å	7.56 Å	8.00 Å
	b	4.43 Å	4.43 Å	4.36 Å	4.36 Å	4.40 Å
Strain %	**a**		-1.23%		5.82%	
	b		-0.67%		0.92%	

Lattice mismatch:

6.67% along x-direction (a), and 1.58% along y-direction (b).

Lattice mismatch along a particular direction is calculated by,

$$\frac{L_{SnS} - L_{h\text{-}BN}}{L_{SnS}} \times 100$$

Here, L_{SnS} and $L_{h\text{-}BN}$ are the lattice parameters of SnS and *h*-BN supercell along a particular direction.

The strain % on each layer along a particular direction is calculated by,

$$\frac{L_{hetero} - L_{SnS/h\text{-}BN}}{L_{SnS/h\text{-}BN}} \times 100$$

Here, L_{hetero} is the lattice parameter of the heterostructure along a particular direction; $L_{SnS/h\text{-}BN}$ is the lattice parameter of SnS or *h*-BN supercell along the same direction.

4. AIMD simulation

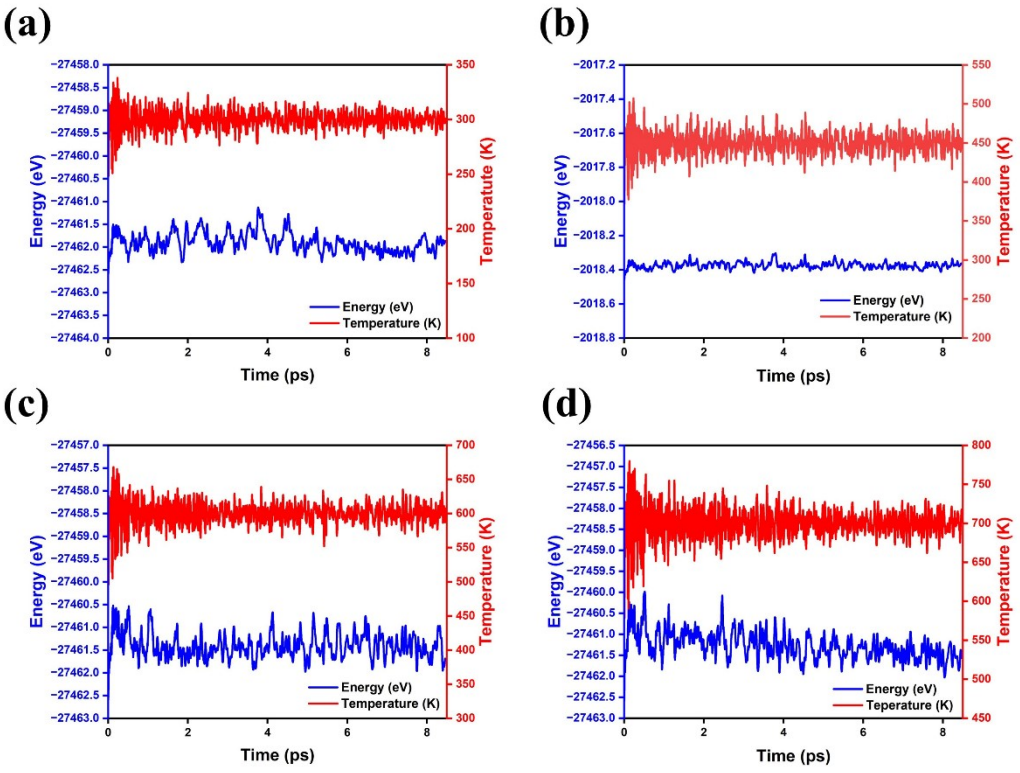

Figure S4: AIMD simulation at **(a)** 300 K, **(b)** 450 K, **(c)** 600 K, and **(d)** 700 K (the axis on the left side of the figures represents the energy of the heterostructure and the axis on the right side of the figures represents the temperatures scale).

Figure S4 illustrates the thermal stability of the SnS/h-BN heterostructure, calculated via ab-initio molecular dynamics (AIMD) simulations at 300 K (room temperature) and higher

temperatures of 450 K, 600 K, and 700 K. The maximum energy fluctuations in the total energy of the heterostructure over 8.5 ps at these temperatures were compared to the binding energy (Eb = 1.91 eV) of the heterostructure. At room temperature and 450 K, the maximum energy fluctuations are 0.27 eV and 0.16 eV, respectively. This suggests that the ideal temperature for experimentally synthesizing the mixed-phase SnS/h-BN heterostructure is around 450 K. At 600 K, the energy fluctuation increases to 1.87 eV, nearly equal to the binding energy of the heterostructure. At 700 K, the energy fluctuation (~2.61 eV) due to thermal agitation surpasses the binding energy, potentially causing the heterostructure layers to separate. Thus, we conclude that the mixed-phase SnS/h-BN heterostructure is thermally stable up to 600 K.

5. Band structure with 0% strain of SnS surface

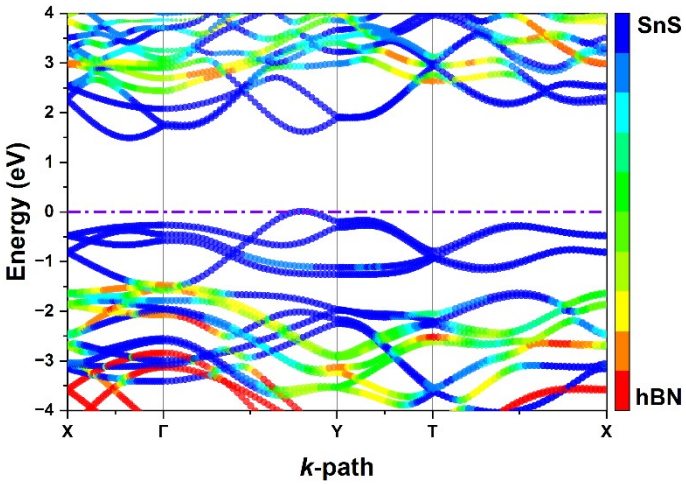

Figure S5: Layer projected electronic band structure of SnS/*h*-BN heterostructure with no strain on SnS layer, plotted along the X-Γ-Y-T-X high symmetric path (blue and red colour indicates the contribution from the SnS and *h*-BN monolayers, respectively);

6. Effective mass

Table S6: Calculated charge carrier effective mass of SnS/*h*-BN heterostructure and SnS monolayer.

Band valley		SnS	SnS/*h*-BN
	Carrier	$m^*(m_0)$	$m^*(m_0)$
Γ-X	electron	0.08	0.04
	hole	0.24	0.38
Γ-Y	electron	0.06	0.06
	hole	0.07	0.08