Supplementary Information (SI)

Strain-engineered bright excitons and nearly flat band in SnNBr monolayer for high-speed LED applications

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Dynamical Stability

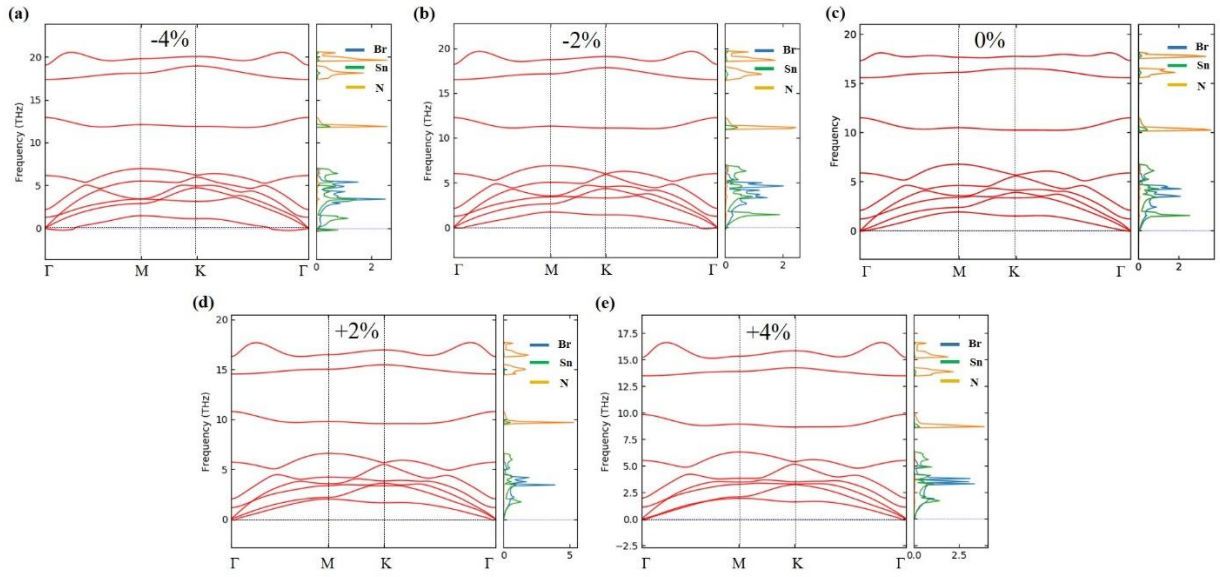

Figure S1. Phonon dispersion of the strained SnNBr monolayer under the (a) compressive biaxial strain 4%, (b) compressive biaxial strain 2%, (c) unstrained condition or 0%, (d) tensile biaxial strain 2%, and (e) tensile biaxial strain 4%.

A small pocket of imaginary frequency is observed around the Γ point in the acoustic flexural (ZA) mode of 4% compressive strained SnNBr, which is a mere artifact commonly observed in 2D materials and is extremely sensitive to the details of the calculations and in some cases, it goes off altogether with a bigger supercell and/or a denser k-mesh. It simply shows the difficulty in reaching numerical convergence for the acoustic flexural phonon mode, which happens to be a common issue in ab initio calculations on 2D materials. The small pockets of negative frequency (<0.3 THz or 10 cm^{-1}) around the Γ point arising from the flexural acoustic (ZA) modes have also been observed in graphene, silicone, molybdenum disulfide, indium and gallium chalcogenides[1,2]. This region of instability is particularly dependent on simulation parameters, such as supercell size and k-point sampling, and can be reduced by using a bigger supercell, denser k-mesh or including non-analytical correction (NAC) term.

Band dispersion in hexagonal and orthorhombic unit cell under biaxial strain

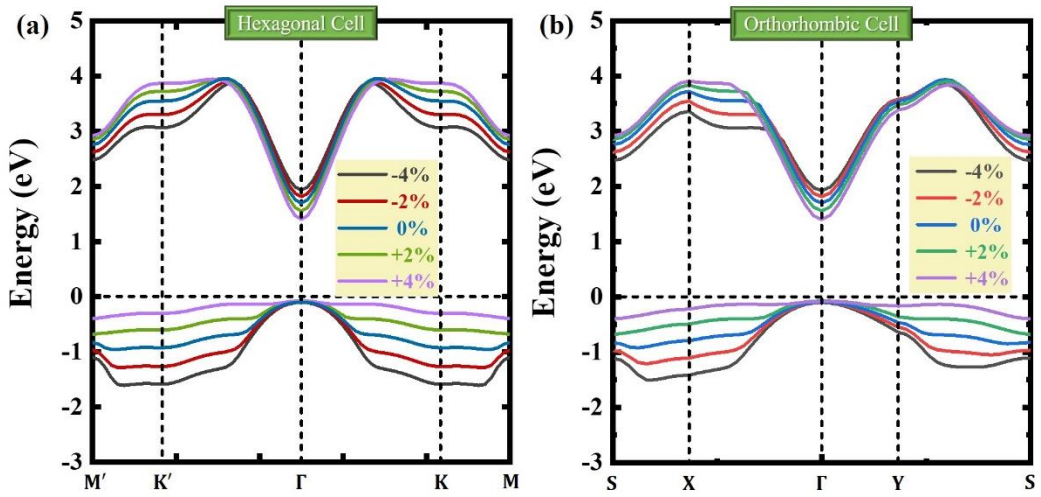

Figure S2. Effect of strain on lowest conduction band (LCB) and highest valence band (HVB) in (a) hexagonal unit cell and (b) orthorhombic unit cell

Figure S2 indicates that the band gap remains identical regardless of the unit cell type under strain. Additionally, the band edges consistently remain at Γ across all strain levels, irrespective of the unit cell structure. For both unit cells, the change in band dispersion follows an identical trend: tensile strain introduces a similar degree of flatness/narrowness to the HVB and LCB across both structures.

Convergence of parameters for GW+BSE calculations:

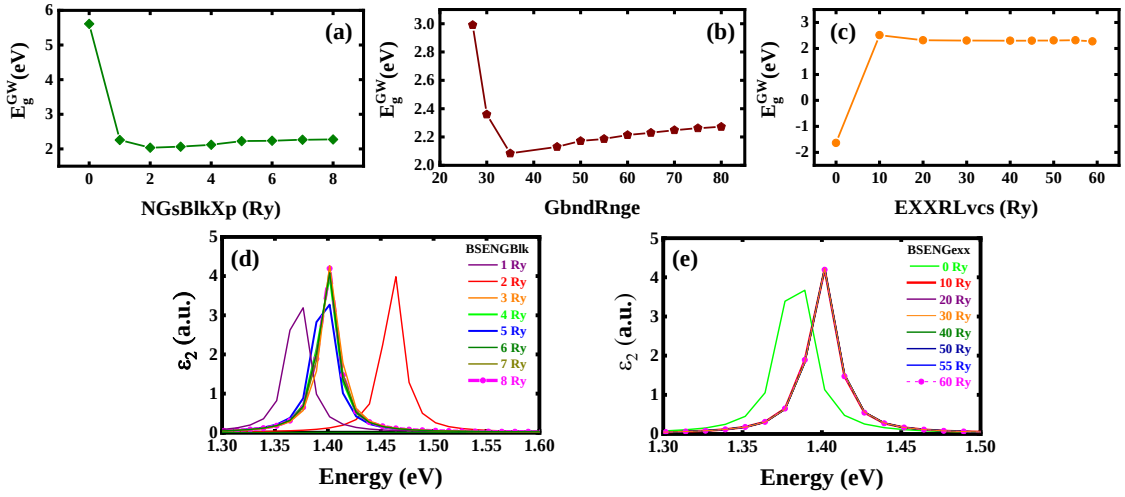

Figure S3. Convergence of (a,b,c) GW parameters and (d,e) BSE parameters for pristine SnNBr

Details of exciton lifetime calculations

- **Wannier approach with Fermi's golden rule**

Fermi's golden rule, based on first-order perturbation theory, describes the transition rate in a quantum system as a function of the transition matrix element and the density of states, which is given as follows:

$$\gamma_i^f = \frac{2\pi}{\hbar}|\langle i|H'|f\rangle|^2\delta\left(E_f - E_i - \hbar\omega\right) \quad (1)$$

This transition is driven by a perturbation

$$H' = -\frac{e}{mc}\mathbf{A}\cdot\mathbf{p}\,;\,\mathbf{A} = \mathbf{A}_0\hat{\mathbf{e}}(\exp\left[i(\mathbf{k}\cdot\mathbf{r} - \omega t)\right] + \exp\left[-i(\mathbf{k}\cdot\mathbf{r} - \omega t)\right]) \quad (2)$$

where **p** is momentum and **A** the vector potential in second quantized form, $|i\rangle$ represents the initial state, and $|f\rangle$ represents the final state. Fermi's golden rule is also known as the decay probability, which is related to the inverse of the lifetime.

The transition rate γ_i^f is therefore proportional to the dipole matrix element:

$$\gamma_i^f = \frac{2\pi}{\hbar}\frac{e^2}{m^2c^2}A_0^2\Sigma\left|\langle v\mathbf{k}|e^{(i\mathbf{k}\cdot\mathbf{r})}\hat{\mathbf{e}}\cdot\mathbf{p}|c\mathbf{k}\rangle\right|^2\delta\left(E_f - E_i - \hbar\omega\right) \quad (3)$$

$$\gamma_i^f \propto \sum_{\mathbf{k}}|\langle v\mathbf{k}|\hat{\mathbf{e}}\cdot\mathbf{r}|c\mathbf{k}\rangle|^2 \propto p_s^2 \quad (4)$$

Transition Dipole Matrix Element defined by k.p model for electron and hole by equations (5) and (6) respectively:

$$\left(\frac{m_0}{m_{e^-}^*}\right)_{ij} = \delta_{ij} + \frac{2}{m_0}\sum_k\frac{|\langle vk|p|ck\rangle|^2}{E_g} \quad (5)$$

$$\left(\frac{m_0}{m_{h^-}^*}\right)_{ij} = \delta_{ij} + \frac{2}{m_0}\sum_k\frac{|\langle vk|p|ck\rangle|^2}{E_g} \quad (6)$$

Where i & j are the band indices, $p = -i\frac{\partial}{\partial k}$ stands for the momentum operator and vk & ck are the valence and conduction band as operands. For the exciton pair ($i \neq j$), Kronecker delta, δ_{ij} turns to be zero and adding the above two equations (5) & (6) result as follows:

$$\Rightarrow \quad \frac{m_0}{m_{e^-}^*} + \frac{m_0}{m_{h^-}^*} = \frac{4}{m_0}\frac{|\langle vk|p|ck\rangle|^2}{E_g}$$

$$\Rightarrow \quad \frac{1}{m_{e^-}^*} + \frac{1}{m_{h^-}^*} = \frac{4}{m_0^2}\frac{|\langle vk|p|ck\rangle|^2}{E_g}$$

$$\Rightarrow \qquad \frac{1}{\mu_{ex}} = \frac{4|\langle vk|p|ck\rangle|^2}{m_0^2 E_g} = \frac{4p_s^2}{m_0^2 E_g} \tag{7}$$

- **Recombination rate and effective exciton lifetime**

The recombination rate of electron-hole pair at zero momentum, $\gamma_s(0)$ and therefore, the exciton lifetime (τ) can be obtained by using the following relations[3]:

$$\gamma_s(0) = \frac{e^2 p_s^2}{\varepsilon_0 m^2 c A E_s(0)} \tag{8}$$

$$\tau = \frac{1}{\gamma_s(0)} \tag{9}$$

where ε_0 is the absolute permittivity of free space, e is the charge of an electron, c is the speed of light, m is the reduced effective mass, *A* is the area of the unit cell and $E_s(0)$ is the excitation energy at zero momentum.

References:

1 S. Radescu, D. Machon and P. Mélinon, Origin of dynamical instabilities in some simulated two-dimensional materials: GaSe as a case study, *Phys. Rev. Mater.*, 2019, **3**, 074002.

2 V. Zólyomi, N. D. Drummond and V. I. Fal'ko, Electrons and phonons in single layers of hexagonal indium chalcogenides from ab initio calculations, *Phys. Rev. B*, 2014, **89**, 205416.

3 H. Y. Chen, M. Palummo, D. Sangalli and M. Bernardi, Theory and Ab Initio Computation of the Anisotropic Light Emission in Monolayer Transition Metal Dichalcogenides, *Nano Lett.*, 2018, **18**, 3839–3843.