Supplementary Information

Strongly Anisotropic Optoelectronic Properties and Long Exciton Lifetimes in Two-Dimensional $GaInS_3$-Type Monolayers

Nian Zhang [1,2], Wangping Xu [1,3], Li Xiang [1], Ziwei Zhao [1], Juexian Cao [1,3], and Xiaolin Wei [1,2*]*

[1] *Department of Physics & Hunan Institute of Advanced Sensing and Information Technology, Xiangtan University, Xiangtan 411105, People's Republic of China*

[2] *College of Physics and Electronics Engineering, Hengyang Normal University, Hengyang 421002, People's Republic of China*

[3] *Hunan Provincial Key Laboratory of Smart Carbon Materials and Advanced Sensing, Xiangtan University, Xiangtan 411105, People's Republic of China*

*E-mail: xuwp@xtu.edu.cn; xlw@xtu.edu.cn

Supplementary Figures

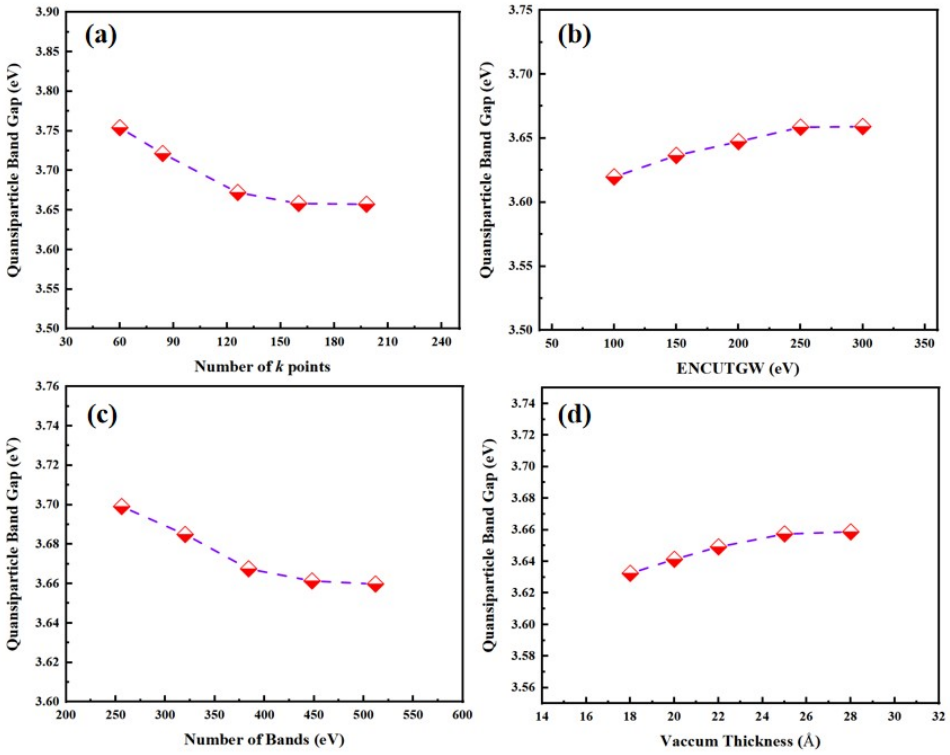

Figure S1. Convergence tests of the quasiparticle band gap of α-$GaInS_3$ monolayer with respect to (a) k-point numbers, (b) GW cutoff energy, (c) the number of bands, and (d) the vacuum thickness.

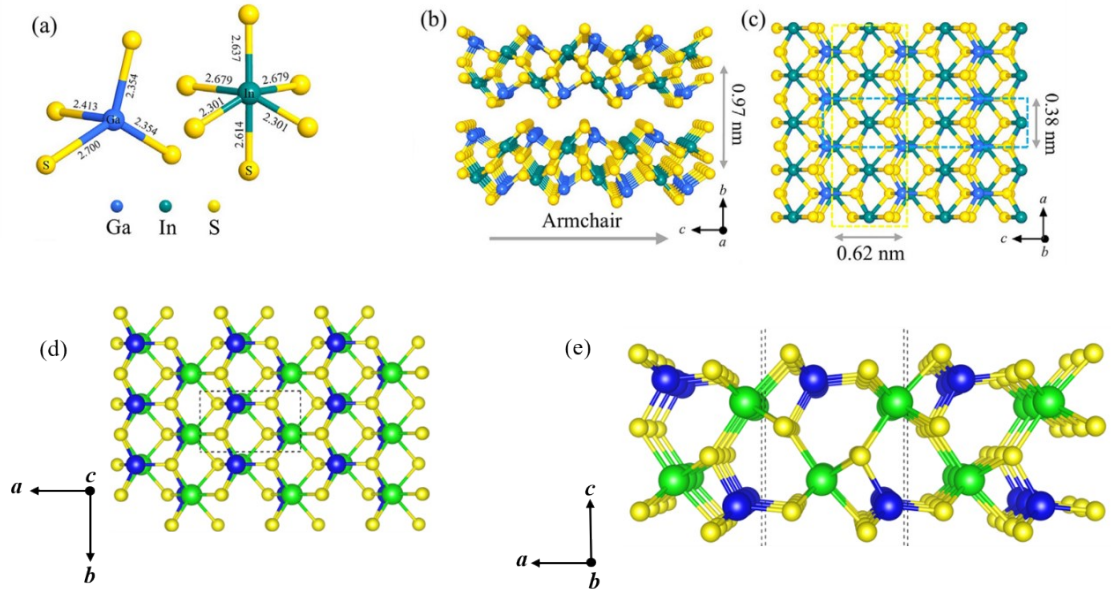

Figure S2. (a) The two structural units comprising GaS_4 tetrahedra and InS_6 octahedra within $GaInS_3$; (b) a side view of the bilayer $GaInS_3$ structure; and (c) a top view. (d) Top view of a single-

layer α-$GaInS_3$ structure and (e) side view.

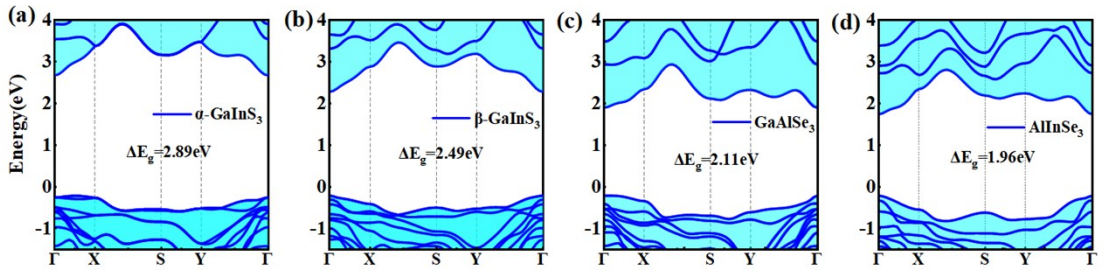

Figure S3. (a) Band structure diagram of α-$GaInS_3$ in the HSE06 model. (b-d) Band structure diagrams of three direct-bandgap variants of HSE06 in the ABM_3 model.

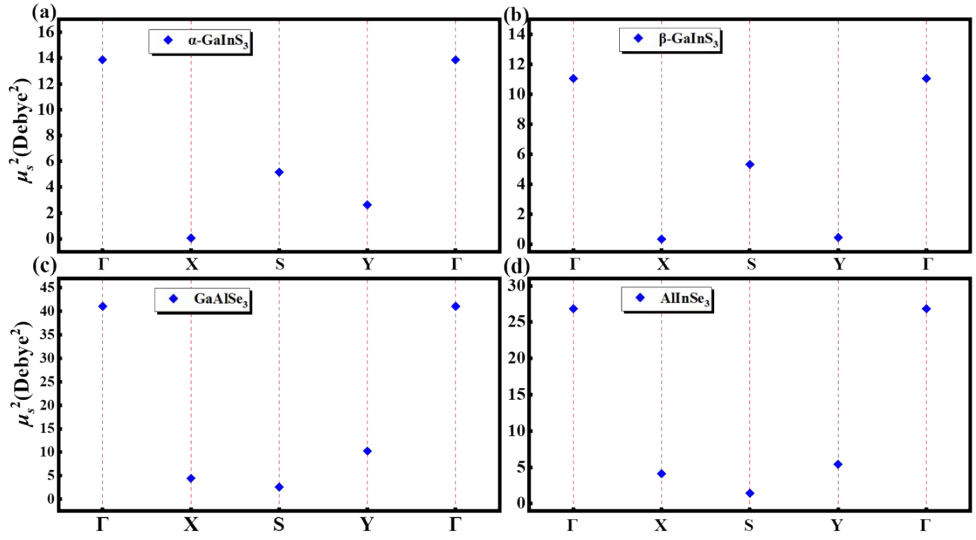

Figure S4. Calculated exciton dipole moments of $GaInS_3$-like rectangular monolayers (a) α-$GaInS_3$ (b) β-$GaInS_3$ (c) $GaAlSe_3$ and (d) $AlInSe_3$.

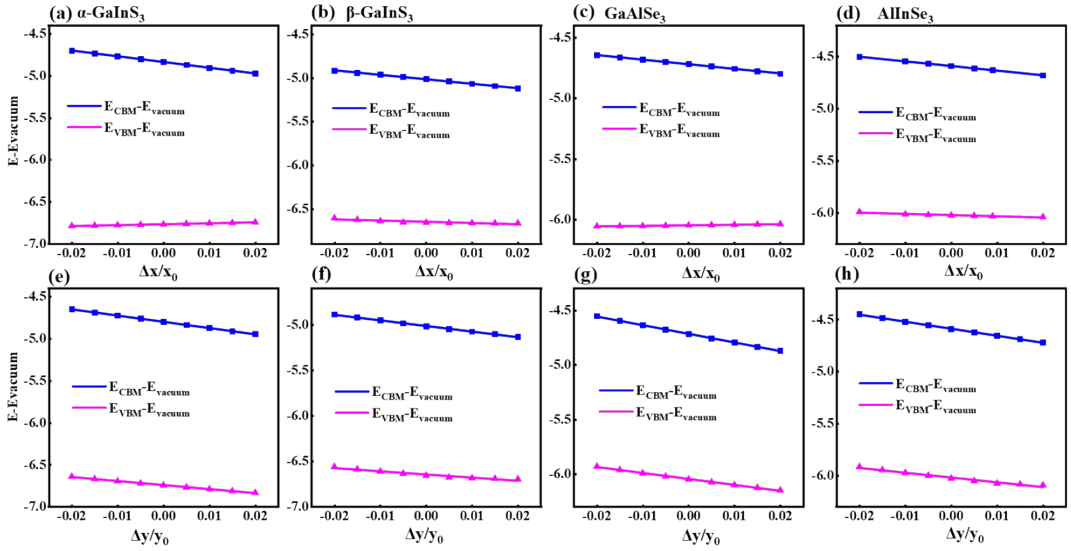

Figure S5. (a–d) Formation energies of the experimental α-$GaInS_3$ structure and ABM_3

structure along the x-direction. (e–h) Formation energies of the experimental α-$GaInS_3$ structure and ABM_3 structure along the y-direction.

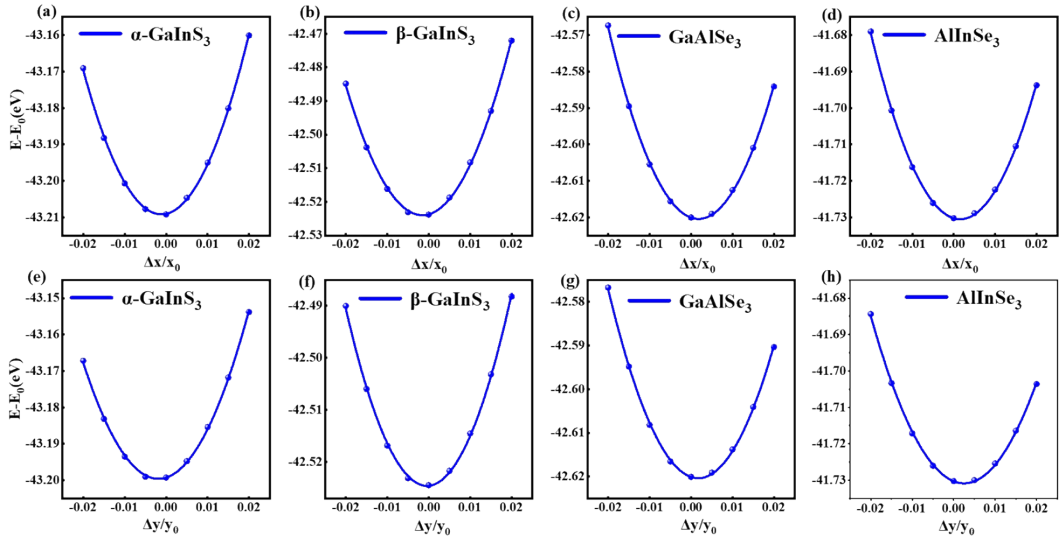

Figure S6. (a–d) Experimental α-$GaInS_3$ structures and ABM_3 structures: elastic modulus (C_{2D}) images along the x-direction. (e–h) Experimental α-$GaInS_3$ structures and ABM_3 structures: elastic modulus (C_{2D}) images along the y-direction.

Table S1. The calculated elastic constants of the three single-layer materials.

Elastic constant	β-$GaInS_3$	$GaAlSe_3$	$AlInSe_3$
C_{11}	45.52	48.08	41.09
C_{12}	10.34	9.45	9.15
C_{22}	34.16	36.72	31.95
C_{66}	8.21	9.97	8.85
$C_{11}C_{22}$-C_{12}^2	1448.25	1676.20	1229.10

Carrier Mobility calculation

To investigate the material electron and hole transport properties, here, we use the phonon-limited scattering model of the deformation potential theory proposed by Bardeen and Shockley to predict the carrier mobility of monolayer ABM_3 (A=Ga, In, Al; B=Ga, In, Al; M=S, Se, Te). For the 2D semiconductor system, we used the following equation to calculate the intrinsic carrier mobility of the monolayers[1,2]:

$$\mu_{2D} = \frac{e\hbar^3 C_{2D}}{K_B T m^* m_d E_l^2} \quad (1)$$

where T, m^*, C_{2D}, E_l, K_B are the temperature, the effective masses of electrons and holes, the elastic modulus of a uniformly deformed crystal, the variational constant along the transport direction, and the Boltzmann constant, respectively. m^* depends on the energy change of the wave vector *k* along different transport directions, defined as $m^* = \frac{\hbar^2}{(\partial^2 E/\partial^2 k)}$. m_d is the average effective mass, which is determined $m_d = \sqrt{m_x m_y}$. The valence band maximum (VBM) of electrons along the transport direction or the conduction band minimum (CBM) of the deformation situation constant E_l, determined by $E_x = \frac{\Delta E_{lx}}{(\Delta l_x / l_{x0})}$ and $E_y = \frac{\Delta E_{ly}}{(\Delta l_y / l_{y0})}$. The calculation of the deformation potential C_{2D} and the distortion potential E_l in the paper is based on the equilibrium lattice structure, respectively, along with the uniaxial axis of the in-plane cell or by applying different values of stretching and compression in the interval from -2% to 2% (taking a series of discrete values at 0.5% intervals) to achieve the calculation of the cell under different strains.

References:

[1] J. Bardeen, W. Shockley, Deformation potentials and mobilities in non-polar crystals, Phys. Rev., 80(1950) 72-80.

[2] J. Qiao, X. Kong, Z. Hu, F. Yang, W. Ji, High-mobility transport anisotropy and linear dichroism in few-layer black phosphorus, Nat. Commun., 5(2014) 4475.