Supporting Information

Electric Field and Strain Engineering Tuning of 2D Gr/α-Ga_2O_3 van der Waals Heterostructures

Xiangyu Wu[1#], Zhiyang Xie[2#], Yu Zhang[3,4], Xuefei Liu[1*], Jinshun Bi[1*], Wentao Wang[5], Zhaofu Zhang[6,7], Ruyue Cao[8,9*]

[1] School of physics and electronic science, Guizhou Normal University, Guiyang, 550025, China.
[2]Wuhan Institute of Shipbuilding Technology, Department of ship electrical and electronic technology, Qintai Road, Hanyang distrinct, Wuhan, 430050, China.
[3]Jincheng Research Institute of Opto-mechatronics Industry.
[4]Shanxi Key Laboratory of Advanced Semiconductor Optoelectronic Devices and Integrated Systems.
[5]Guizhou Provincial Key Laboratory of Computational Nano-Material Science, Guizhou Education University, Guiyang, 550025, China.
[6]The Institute of Technological Sciences, Wuhan University, Wuhan, Hubei, 430072, China
[7]Hubei Key Laboratory of Electronic Manufacturing and Packaging Integration, Wuhan University, Wuhan, 430072, China
[8]State Key Laboratory of Superlattices and Microstructures, Institute of Semiconductors, Chinese Academy of Sciences, Beijing, 100083, China.
[9]Engineering Dept, Cambridge University, Cambridge CB2 1PZ, UK

* Corresponding author:

E-mail: 201307129@gznu.edu.cn (X.L.);

E-mail: bijinshun@gznu.edu (J.B.);

E-mail: rc921@cam.ac.uk.

[#] Contributing equally to this work

Table S1. Elastic constants C_{ij}, layer modulus (γ2D), Young's modulus (Y), Poisson's ratio (ν), intrinsic strength (σ_{int}), and bending modulus (D) of monolayers and vdWHs.

System	C_{11} (N/m)	C_{12} (N/m)	C_{66} (N/m)	γ2D (N/m)	Y (N/m)	ν	G^{2D}	σ_{int} (N/m)	D (eV)
Gr	358.55	61.02	144.83	209.785	348.16	0.17	144.83	38.68	1.93
α-Ga_2O_3	174.24	95.97	38.41	135.10	120.8	0.55	38.41	13.42	68.47
Gr/α-Ga_2O_3↑	474.02	155.45	169.41	314.73	440.5	0.32	169.41	48.94	526.21
Gr/α-Ga_2O_3↓	494.26	133.20	180.56	313.73	458.41	0.27	180.56	50.93	530.93

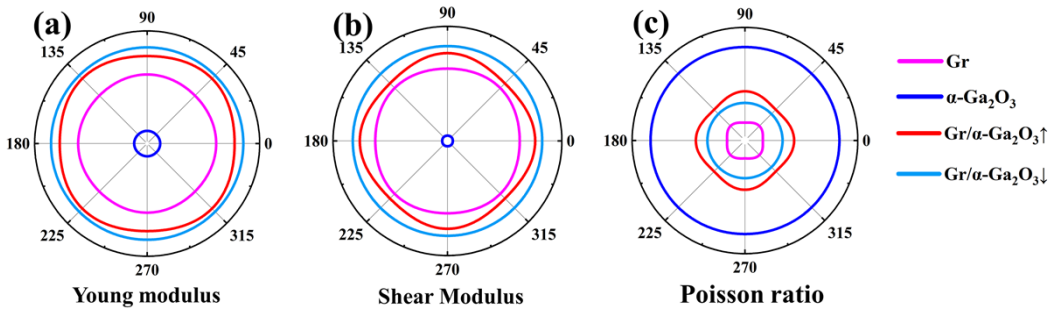

Figure S1. Angle-dependent (a) Young modulus, (b) Shear Modulus, and (c) Poisson rat

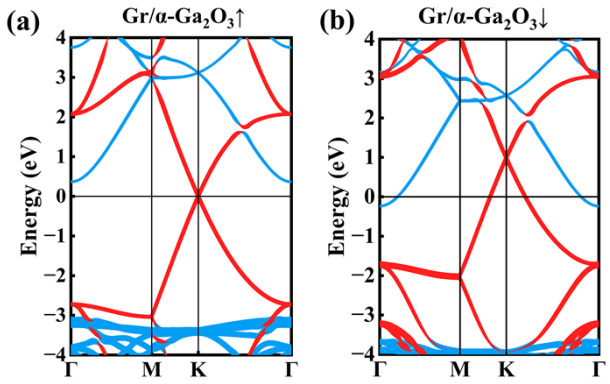

Figure S2. The band structure for (a) Gr/α-Ga_2O_3↑ and (b) Gr/α-Ga_2O_3↓ vdWHs at HSE06 level.

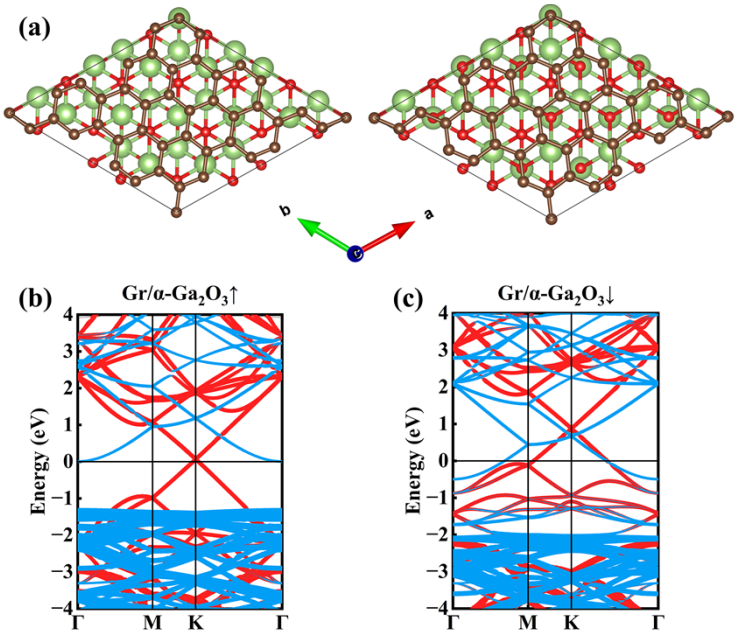

Figure S3. (a) The relaxed atomic structures of Gr/α-Ga_2O_3 vdWHs with low strain contain 98 atoms. (Gr is squeezed by around 0.5%, whereas α-Ga_2O_3 vdWHs has no tensile or compressive strain.) The band structure for (b) Gr/α-Ga_2O_3↑ and (c) Gr/α-Ga_2O_3↓ vdWHs with low strain.

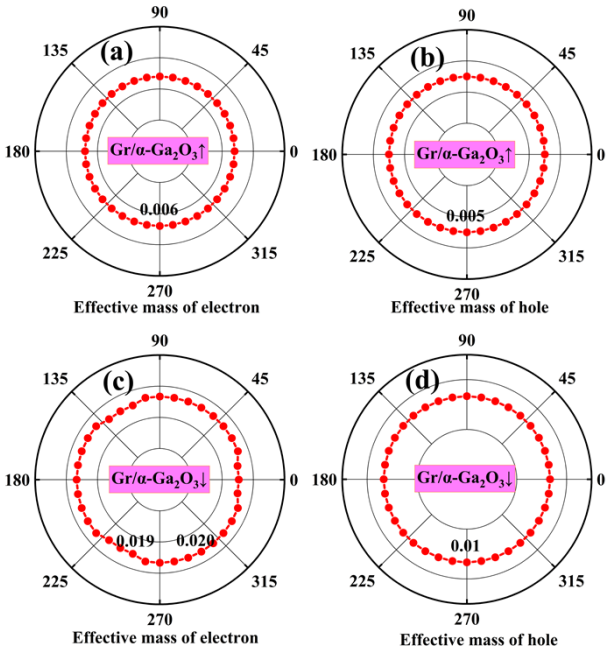

Figure S4. Angular dependence of effective masses of (a, b) electrons and (b, d) holes of for Gr/α-Ga_2O_3↑ and Gr/α-Ga_2O_3↓ vdWHs

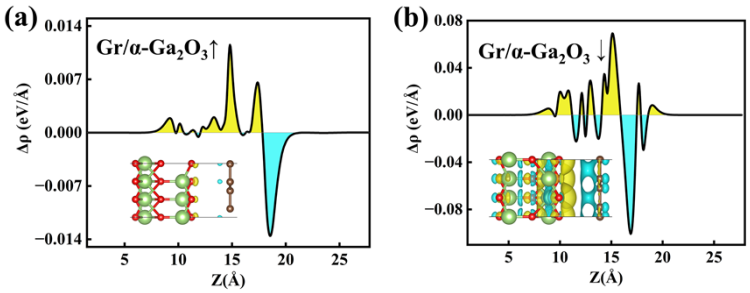

Figure S5. The plane-average charge density difference along the z-direction: (a) Gr/α-Ga_2O_3↑ vdWHs and (b) Gr/α-Ga_2O_3↓ vdWHs, where yellow/cyan-blue indicates the accumulation/depletion of electrons. two Illustrations are the Charge density difference for Gr/α-Ga_2O_3↑ and Gr/α-Ga_2O_3↓ vdWHs and the isosurface value is 0.000318 electrons/$Bohr^3$.

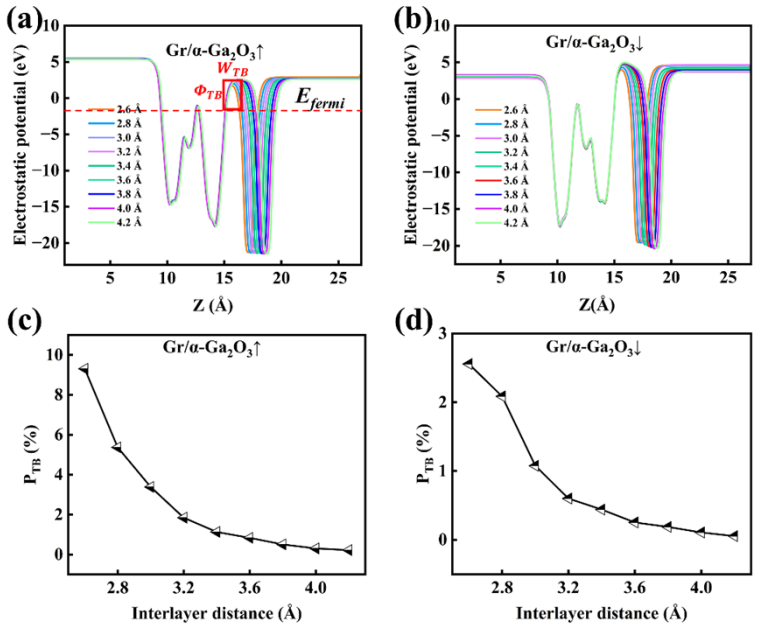

Figure S6. Electrostatic potential distribution along the z-axis under interlayer distance: (a) Gr/α-Ga_2O_3↑ and (b) Gr/α-Ga_2O_3↓ vdWHs. P_{TB} of Gr/Ga_2O_3↑ (c) and Gr/α-Ga_2O_3↓ (d) vdWHs varies with interlayer spacings.

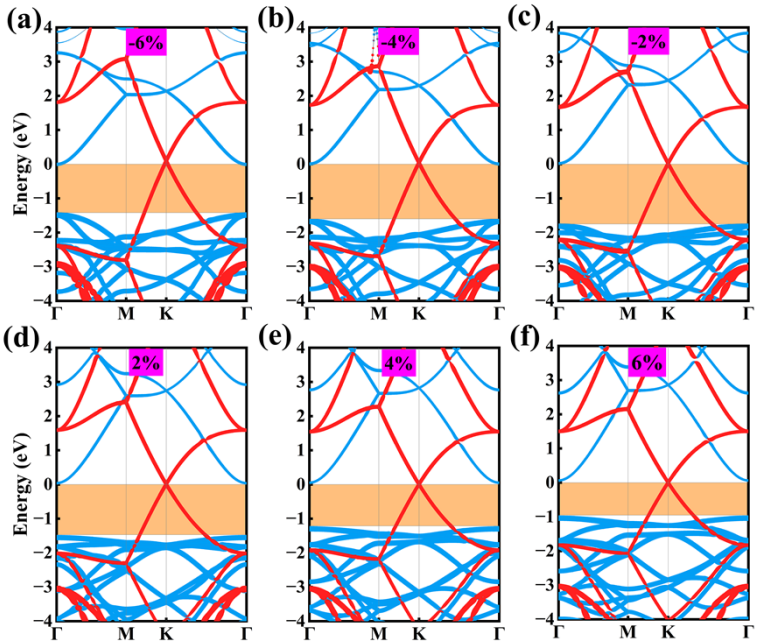

Figure S7. The band structure changes of Gr/α-Ga_2O_3↑ vdWHs for biaxial strain at -6% to 6%.

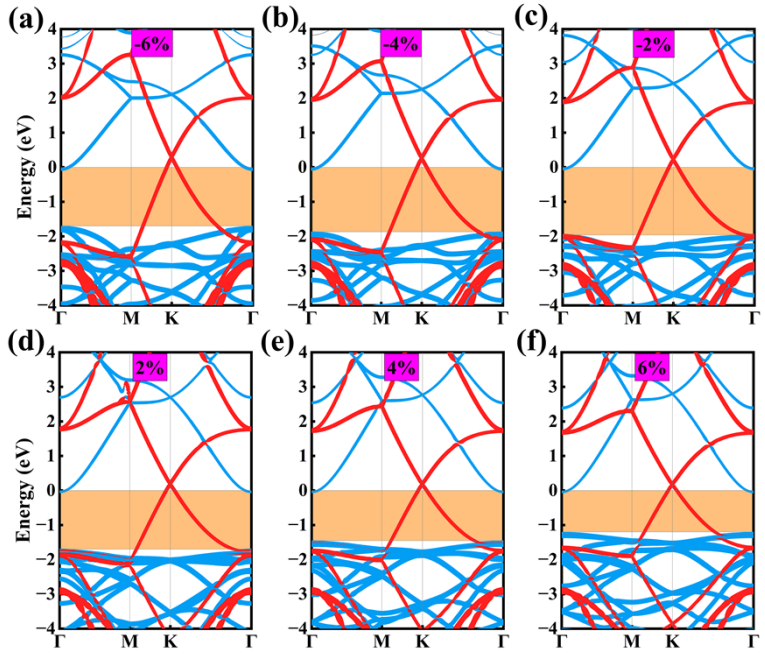

Figure S8. The band structure changes of Gr/α-Ga_2O_3↓ vdWHs for biaxial strain at -6% to 6%.

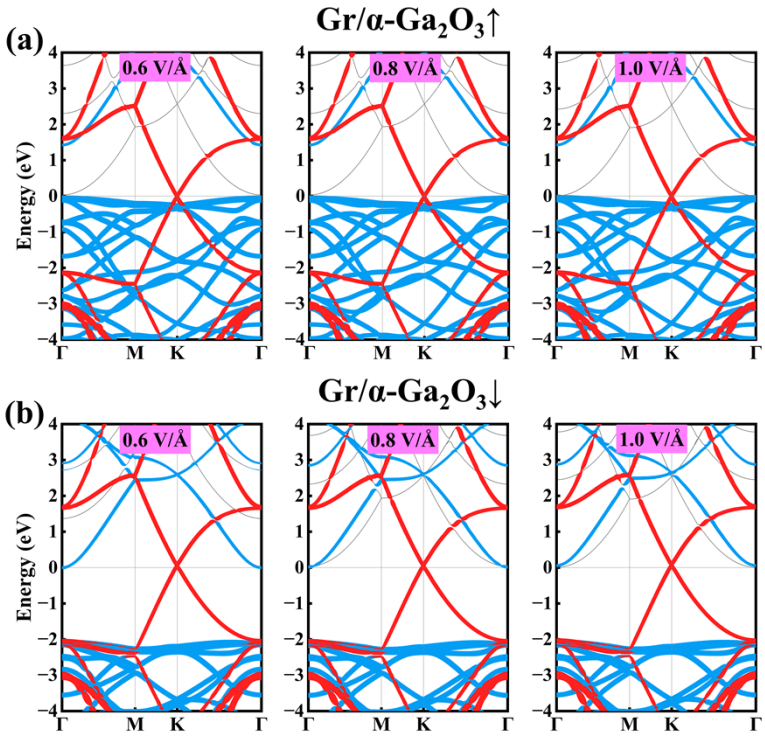

Figure S9. The band structures of Gr/α-Ga_2O_3↑ (a) and Gr/α-Ga_2O_3↓ (b) vdWHs at 0.6, 0.8, and 1.0 Å, respectively.

Note 1:

The complex dielectric function $\varepsilon(\omega) = \varepsilon_1(\omega) + i\varepsilon_2(\omega)$ can be used to calculate all optical characteristics. The momentum matrix elements between occupied and unoccupied states can be leveraged to identify the imaginary part $\varepsilon_2(\omega)$, whereas the real part $\varepsilon_1(\omega)$ is realizable from $\varepsilon_2(\omega)$ using the Kramer Kronig expressions[16]:

$$\varepsilon_2(\omega) = \frac{2e^2\pi}{\Omega\epsilon_0}\sum_{k,v,c}\left|\left\langle\psi_k^c\middle|\hat{u}\times\hat{r}\middle|\psi_k^v\right\rangle\right|^2\delta\left(E_k^c - E_k^v - E\right) \tag{1}$$

$$\varepsilon_1(\omega) = 1 + \left(\frac{2}{\pi}\right)\int_0^\infty d\omega' \frac{\omega'^2\varepsilon_2(\omega')}{\omega'^2 - \omega^2} \tag{2}$$

In the preceding equations, û, e, ψ_k^c, and ψ_k^v are vectors that characterize the polarization of the incident electric field, electronic charge, and the wave functions of the conduction band and valence band, respectively. Using the formulas below, the function of ε(ω) can be used to compute various optical constants such as absorption coefficient (α(ω)), reflectivity (R(ω)), energy loss function (L(ω)), and real portion of optical conductivity (σ(ω)):

$$\alpha(\omega) = \sqrt{2}\omega\left[\sqrt{\varepsilon_1^2(\omega) + \varepsilon_2^2(\omega)} - \varepsilon_1(\omega)\right]^{\frac{1}{2}} \tag{3}$$

$$R(\omega) = \left|\frac{\sqrt{\varepsilon_1(\omega) + j\varepsilon_2(\omega)} - 1}{\sqrt{\varepsilon_1(\omega) + j\varepsilon_2(\omega)} + 1}\right|^2 \tag{4}$$

$$L(\omega) = Im\left(\frac{-1}{\varepsilon(\omega)}\right) = \frac{\varepsilon_2(\omega)}{\varepsilon_1^2(\omega) + \varepsilon_2^2(\omega)} \tag{5}$$

$$Re[\sigma(\omega)] = \frac{\omega}{4\pi}\varepsilon_2(\omega) \tag{6}$$

References

(66) Liu, Q.-J.; Liu, Z.-T.; Feng, L.-P. Elasticity, Electronic Structure, Chemical Bonding and Optical Properties of Monoclinic ZrO_2 from First-Principles. *Phys. B: Condens. Matter* **2011**, *406* (3), 345–350. https://doi.org/10.1016/j.physb.2010.10.057.