Supporting Information

Self-Doped p-n Junction with High Carrier Concentration in 2D GaN/MoSSe Heterostructures: A First-principles Study

Dawei Deng[1,3], Rutong Si[2], Bo Wen[4], Nicola Seriani[2], Xiao-Lin Wei[5], Wen-Jin Yin[1,2,3], and Ralph Gebauer[2*]*

[1]School of Physics and Electronic Science, Hunan University of Science and Technology, Xiangtan 411201, China

[2]The Abdus Salam International Centre for Theoretical Physics (ICTP), Strada Costiera 11, I-34151 Trieste, Italy

[3]Key Laboratory of Intelligent Sensors and Advanced Sensing Materials of Hunan Province, Hunan University of Science and Technology, Xiangtan 411201, China

[4]School of Physics and Electronics, Henan University, Kaifeng 475001, P. R. China

[5]Department of Physics and Laboratory for Quantum Engineering and Micro-Nano Energy Technology, Xiangtan University, Xiangtan 411105, Hunan, China

[*]Corresponding Emails: wyin@ictp.it, rgebauer@ictp.it

S1: Projected band structure of N-S-AB-2 calculated by HSE06 method

Table S1. *The band edge positions of CBM, VBM, and the type of band gap and band alignment of N-S-AB-2 heterostructures in different component thickness simulated by HSE06 method, together with PBE result for comparison. The Fermi level is set at zero eV.*

Heterostructure	Functional	CBM	VBM	Band gap Type	Band Alignment
N(1L)-S(1L)-AB-2	PBE	0.10	-0.110	Indirect	type-II
	HSE06	0.75	-0.218	Indirect	type-II
N(2L)-S(1L)-AB-2	PBE	-0.07	0.040	Indirect	type-III
	HSE06	-0.14	0.007	Indirect	type-III
N(3L)-S(1L)-AB-2	PBE	-0.07	0.050	Indirect	type-III
	HSE06	-0.18	0.003	Indirect	type-III

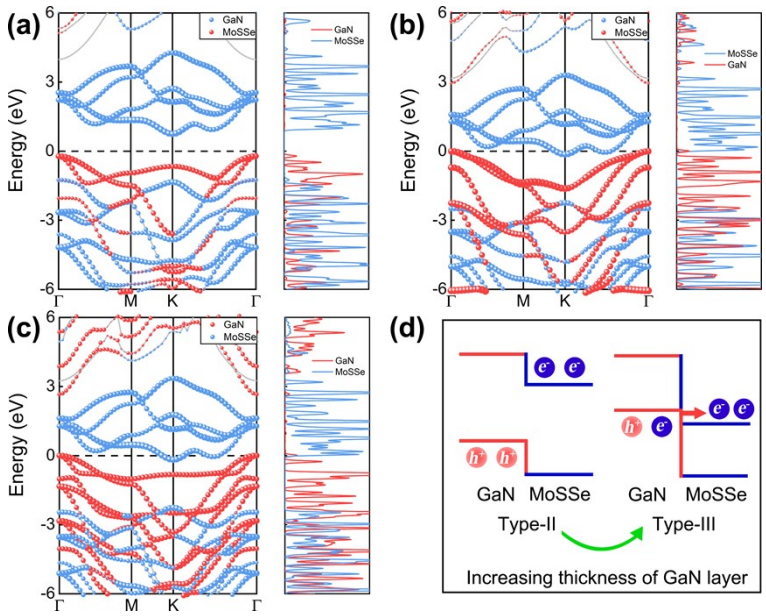

Figure S1. *(Color online) Projected band structure (left panel) of GaN/MoSSe heterostructures in parallel polarization case for N-S combinations with different GaN thickness (a) 1L, (b) 2L, and (c) 3L, together with their projected density of states in right panel. The blue and red dotted lines denote the contribution of GaN and MoSSe, respectively. (d) A schematic of band alignment transitions with increasing GaN thickness.*

S2: Band alignment of N-S-AB-2 tuned by changing polarization strength through varying MoSSe thickness

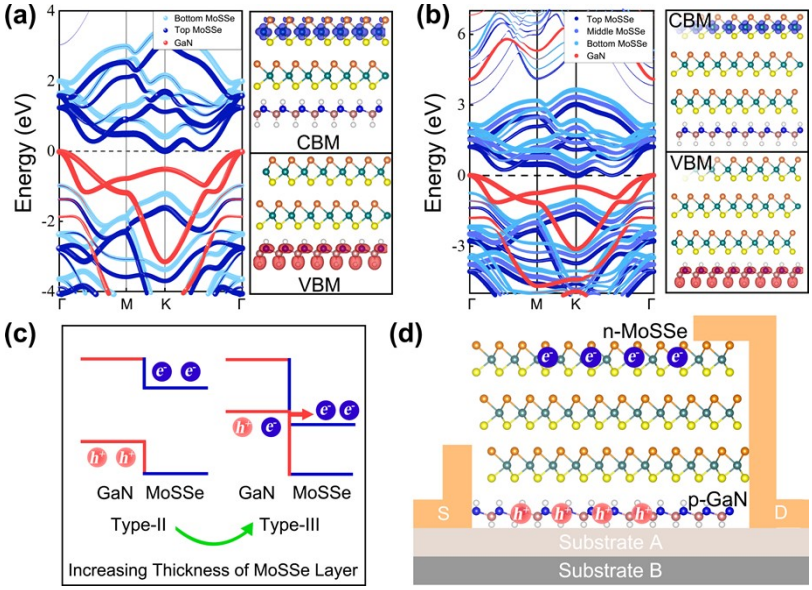

Figure S2. *(Color online) Projected band structure (left panel) of GaN/MoSSe heterostructures with N-S combination in different MoSSe thickness with (a) 2L and (b) 3L, together with their CBM and VBM charge density isosurfaces (right panel). The blue and red dotted lines denote the contribution of GaN and MoSSe components, respectively. (d) The band alignment transition of the heterostructure with GaN thickness changing from 2L to 3L. (e) Schematic of carrier distribution of p-GaN/n-MoSSe junction. The Fermi level is set at zero eV.*

S3: Electronic structures of Ga-Se-AA-2 and N-Se-AB-2

Table S2. *The structural parameters as lattice mismatch ratio (ε), interlayer distance (d), interfacial binding energies (γ_{int}), and the electronic properties as vertical dipole moments (D), and electrostatic potential difference (ΔV) for N-Se-AB-2 and Ga-Se-AA-2 heterostructures in different component thickness. The thickness of GaN and MoSSe component ranges from 1L to 3L.*

Combination	***GaN Thickness***	***ε (%)***	***d (Å)***	***γ_{int} (J/m²)***	***D (Deby)***	***ΔV (eV)***
N-Se	**1L**	0.65	2.35	-0.37	0.89	3.80
	2L	0.84	2.25	-0.43	0.92	3.90
	3L	0.53	2.29	-0.59	0.93	3.91
Ga-Se	**1L**	0.65	2.32	-0.33	0.95	3.86
	2L	0.84	2.24	-0.39	0.99	4.35

3L	0.53	2.16	-0.47	1.02	4.30

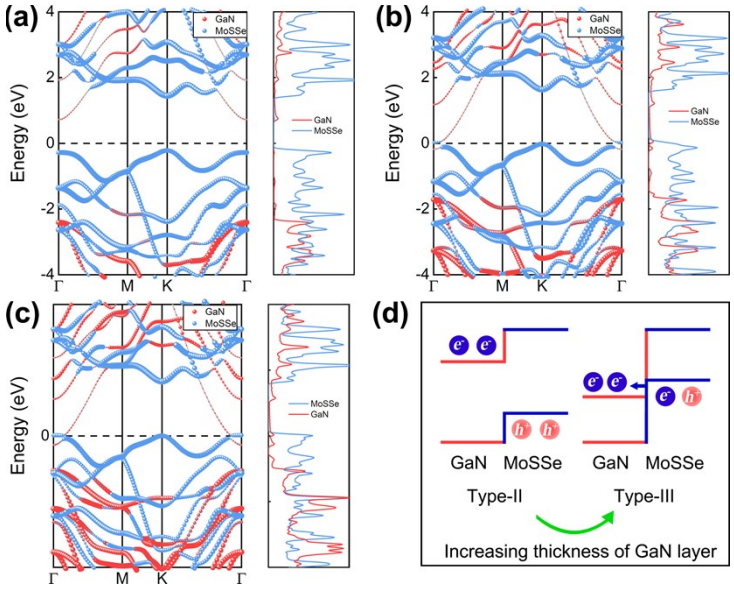

Figure S3. *(Color online) Projected band structure (left panel) of GaN/MoSSe heterostructures in parallel polarization case for Ga-Se combinations with different GaN thickness (a) 1L, (b) 2L, and (c) 3L, together with their projected density of states (right panel). The blue and red dotted lines denote the contribution of GaN and MoSSe component, respectively. (d) A schematic of band alignment transitions with increasing GaN thickness.*

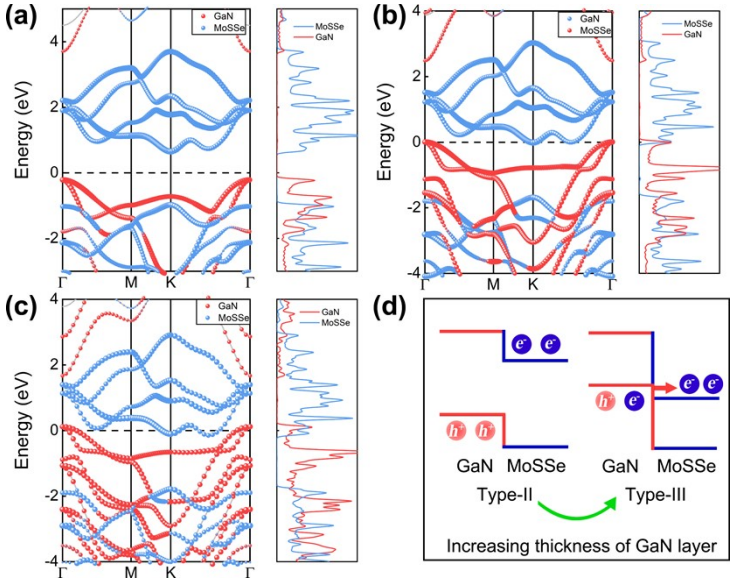

Figure S4. *(Color online) Projected band structure (left panel) of GaN/MoSSe heterostructures in inverse polarization case for N-Se combination with different GaN thickness (a) 1L, (b) 2L, and (c) 3L, together with their projected density of states (right panel). The blue and red dotted lines denote the contribution of GaN and MoSSe component, respectively. (d) A schematic of band alignment transitions with increasing GaN thickness.*

S4: Electronic structure of mix-dimensional heterostructures in N-S-AB-2 pattern with GaN films in five layers (5L)

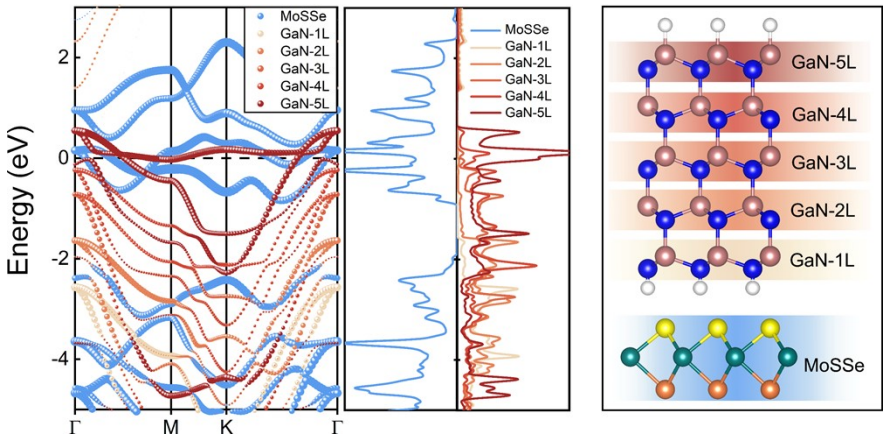

Figure S5*. (Color online) Projected band structure (left panel) of GaN/MoSSe heterostructures in parallel polarization case for N-S configurations with different GaN thickness in 5L, together with their projected density of states (middle panel). The blue and red dotted lines denote the contribution of GaN and MoSSe component, respectively. It should be mentioned that dots in red from light to dark refers denoted different GaN layers from bottom to top.*

S5: Schematic of the possible band alignment in GaN/MoSSe heterostructures

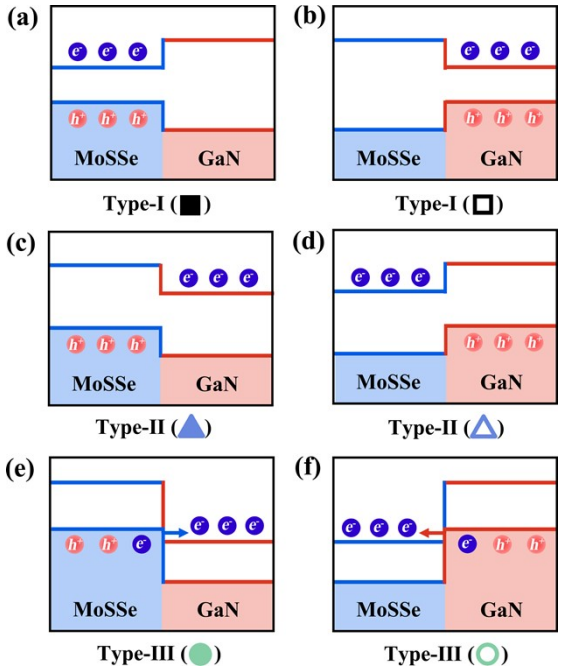

Figure S6. *(Color online) Possible band alignment of GaN/MoSSe heterostructures corresponding to combinations in* ***Fig. 8(a)****. Type-I alignment with both CBM and VBM contributed by (a) MoSSe or (b) GaN component. Type-II alignment with CBM and VBM contributed by (c) MoSSe and GaN component, respectively, with(d) in the opposite situation. Type-III alignment with CBM and VBM contributed by (e) MoSSe and GaN component, respectively, with (f) in the opposite situation.*

S6: Calculated I-V curve of N-S-AB-2 with GaN in different thickness

The current-volt (I-V) curve is simulated based on self-consistent equilibrium Green function, which can be expressed as,[1]

$$I = \frac{e}{h}\int_{-\infty}^{+\infty} T(E;V_b)\left[f_L(E;E_F^L - V_b/2) - f_R(E;E_F^R + V_b/2)\right]dE$$

where $f_{L/R}(E;E_F^{L/R} - V_b/2)$, $E_F^{L/R}$ are the Fermi distribution function and Fermi level of the left/right electrode, respectively. V_b is the applied bias volt. Since Fermi distribution quickly approaches 0 and 1 at $-\infty$ and $+\infty$, like step shape at Fermi level, the range of actual integration can be determined as $\left(E_F^L - V_b/2 - nk_BT, E_F^R + V_b/2 + nk_BT\right)$. The factor nk_BT is added to consider the width of the Fermi-Dirac distribution, where k_B is the Boltzmann constant and T is room temperature. $T\left(E;\ V_b\right)$ is the electron transmission probability function spectrum. Calculated result of N-S-AB-2 in representation is shown in **Fig. S7**. Clearly, the current can be enhanced as the thickness of GaN increases, corresponding a higher carrier concentration in type-III case.

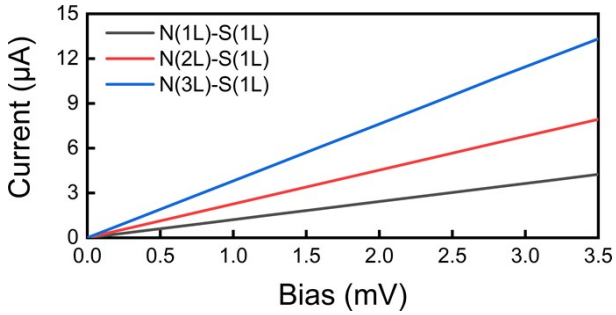

Figure S7. *(Color online) The calculated I–V curve for the N-S-AB-2 heterostructures in different thickness of GaN (black) 1L, (red) 2L, and (blue) 3L.*

S7: Calculated electrostatic potential energy of N-S-AB-2 in different component thickness, together with isolated GaN layers for comparison

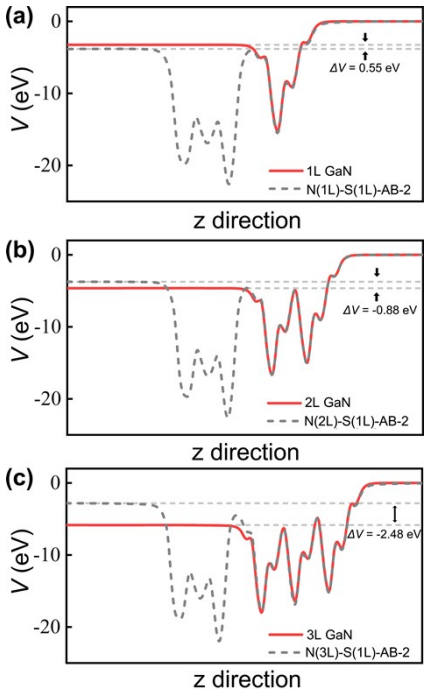

Figure S8. *(Color online) Calculated electrostatic potential energy distribution of N-S-AB-2 heterostructures in different thickness of GaN (a) 1L, (b) 2L, and (c) 3L, together with GaN layers for comparison. ΔV denotes difference between electrostatic potential of heterostructure and isolated GaN.*

S8: Projected band structure of N(2L)-S(1L)-AB-2

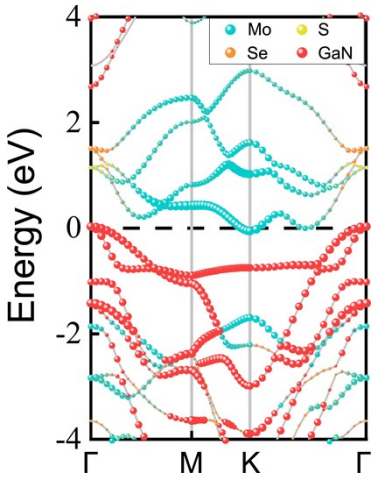

Figure S9. *(Color online) Projected band structure of N(2L)-S(1L)-AB-2. The green, yellow, orange, and red dotted lines denote the contribution of Mo, S, Se atoms, and GaN layer.*

References

(1) M. Büttiker, Y. Imry, R. Landauer, S. Pinhas, Generalized many-channel conductance formula with application to small rings, Physical Review B 31 (1985) 6207-6215.