Electronic Supplementary Material (ESI) for Nanoscale.
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Supplementary information

Ambipolar and n/p-type conduction enhancement of two-dimensional materials by surface charge transfer doping

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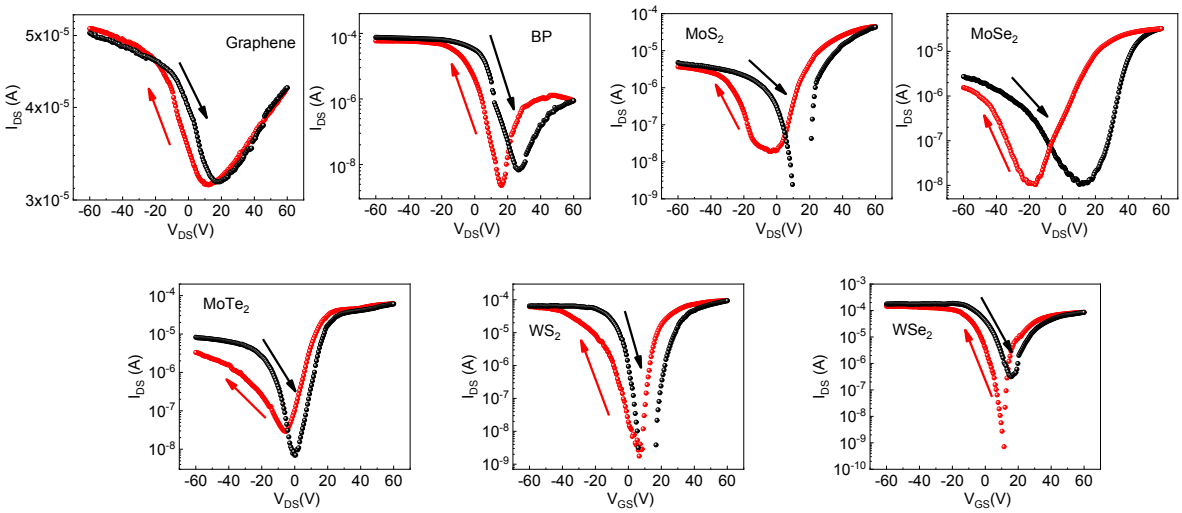

Figure S1. I_{DS} as a function of both increasing (black line) and decreasing (red line) values of V_{GS}.

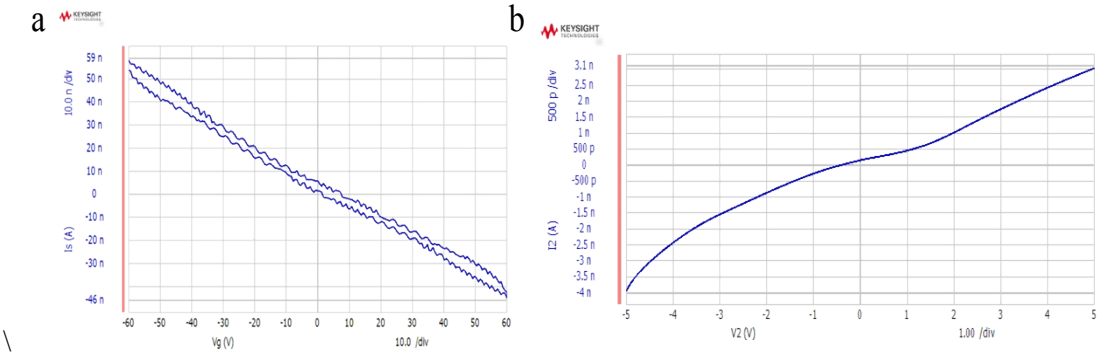

Figure S2. (a) I_{DS}-V_G curve of 10 nm TPB film. (b) I_{DS}-V_{DS} curve of 10 nm TPB film.

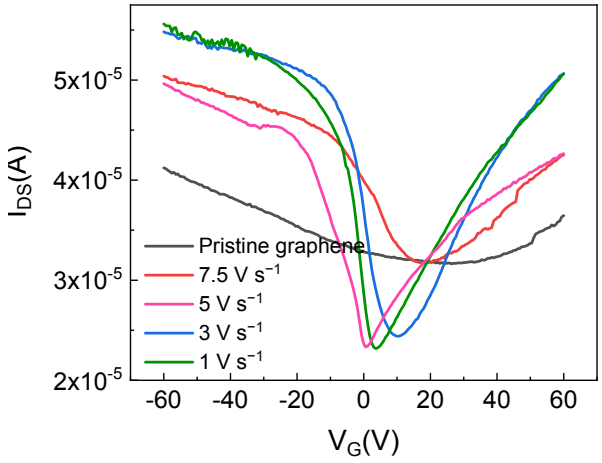

Figure S3.The I_{DS}-V_G curves of the TPB-treated graphene FETs were measured at different V_G sweep rates.

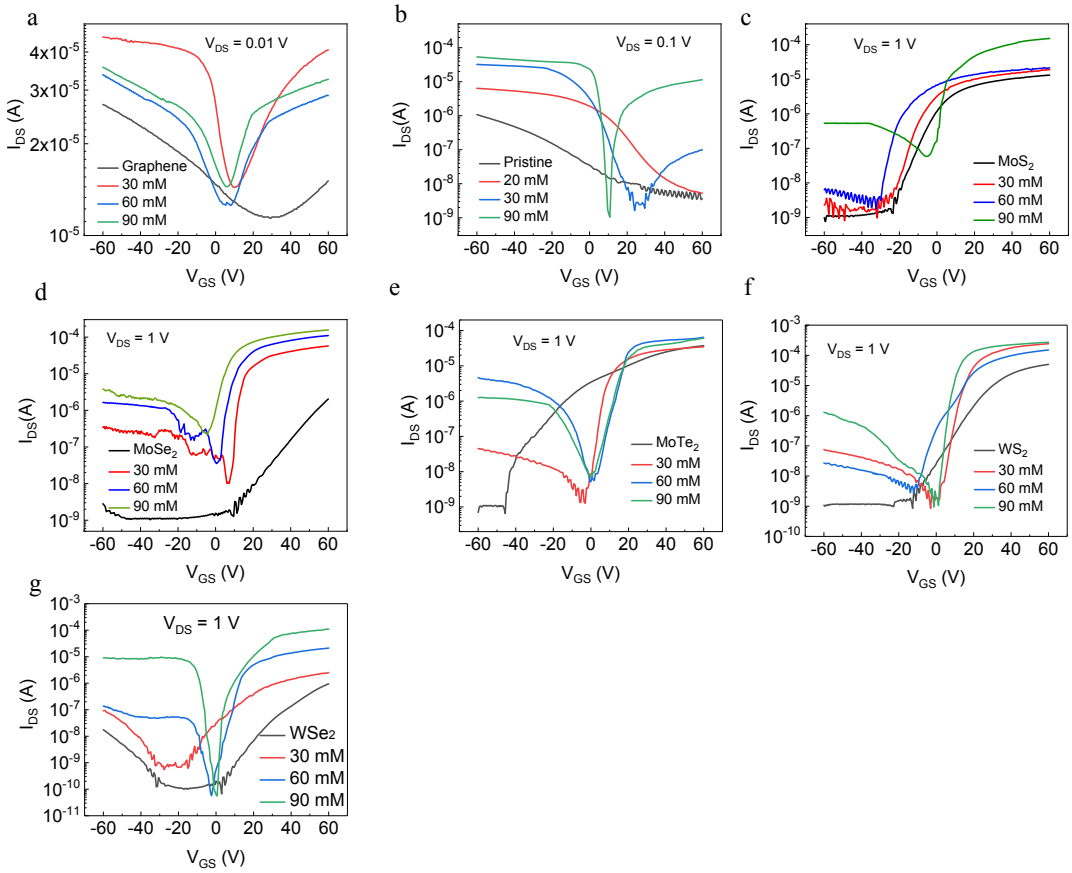

Figure S4. (a-g) Ambipolar improvement of graphene, BP, MoS_2. $MoSe_2$, $MoTe_2$, WS_2, and WSe_2 coated by TPB with three different concentrations.

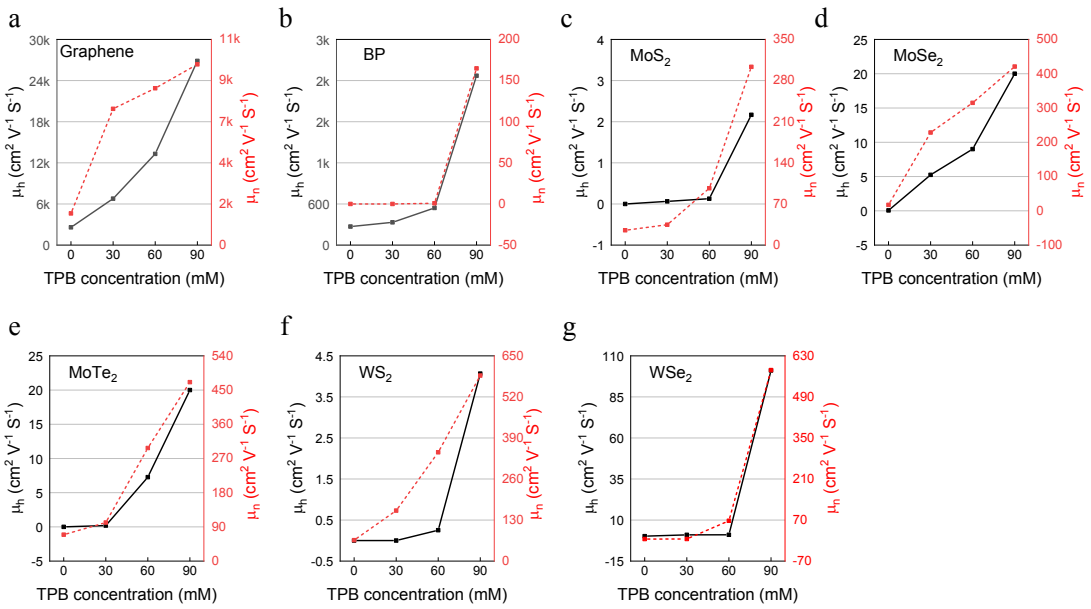

Figure S5. The extracted hole and electron mobility of graphene, BP, MoS_2, $MoSe_2$, $MoTe_2$, WS_2 and WSe_2 transistors. The inherent carrier mobilities μ_h (graphene) increased 10.4-fold, μ_e (graphene) increased 5.7-fold, μ_h (BP) 9.1-fold, μ_e (MoS_2) 12.1-fold, μ_e ($MoSe_2$) 24.7-fold, μ_e ($MoTe_2$) 6.8-fold, μ_e (WS_2) 8.9-fold, and μ_e (WSe_2) 116-fold when the TPB concentration increased to 90 mM. It is clear that WSe_2 is more easily doped than other 2D materials. And the non-inherent mobilities also increased several to hundreds of times, but still lower than the inherent carrier mobility, which demonstrates that TPB does not change the dominant carrier type. Additionally, as shown in Figure S2, μ_h and μ_e values increased faster at higher concentration than at lower concentration, possibly as a result of more charge transferred from TPB to FETs.

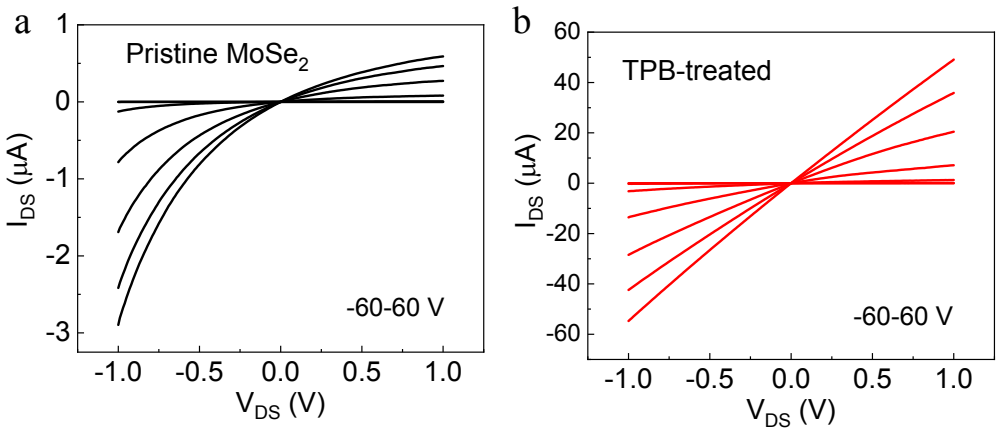

Figure S6. (a, b) I_{DS}-V_{DS} characteristics of the same $MoSe_2$ FET device before (a) and after (b) TPB-treated.

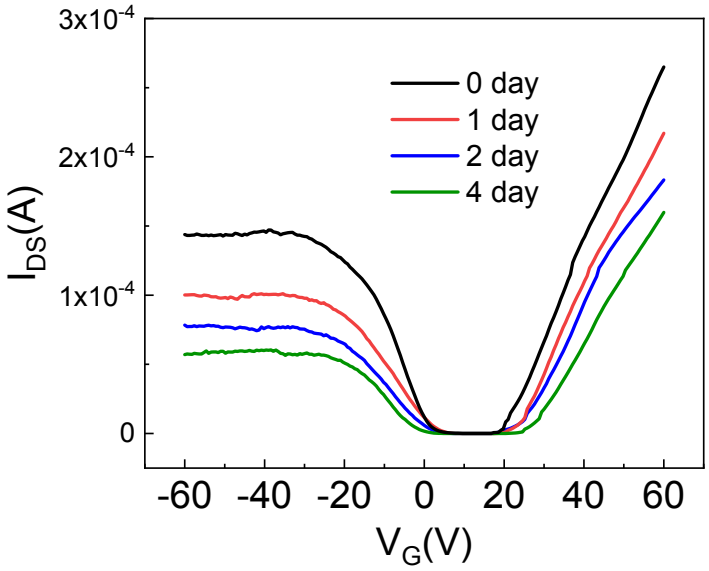

Figure S7. Transfer characteristic curves of the TPB-treated $MoSe_2$ FET after exposure to air for 0, 1, 2, 4 days.

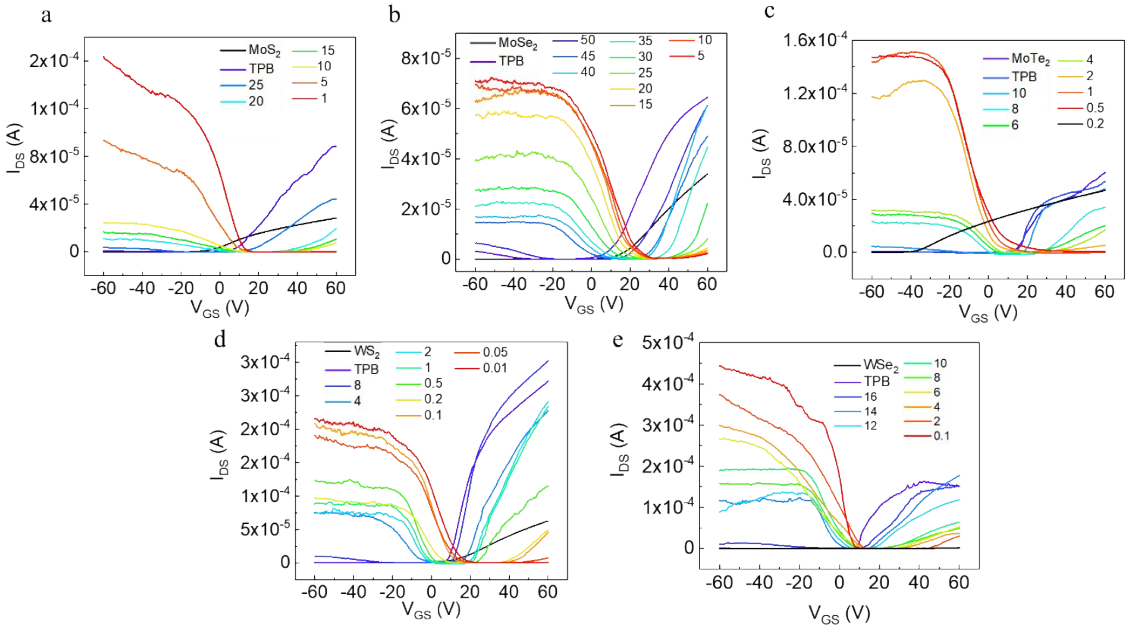

Figure S8. (a-e) The transfer characteristics of the MoS_2, $MoSe_2$, $MoTe_2$, WS_2, and WSe_2 FETs in linear scale with decreasing ratio of TPB/BCF (V_{DS} = 0.1 V).

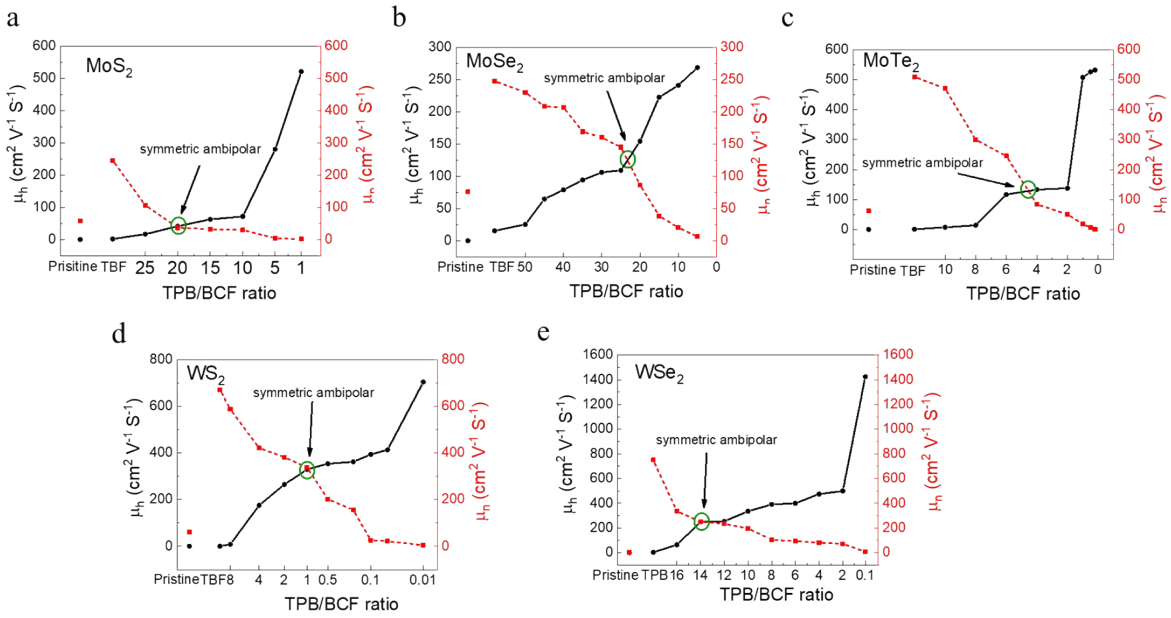

Figure S9. (a-e) The extracted hole and electronic mobility of MoS_2, $MoSe_2$, $MoTe_2$, WS_2, and WSe_2 as function of TPB/BCF ratio. The green circle represents the TPB/BCF ratio at which symmetric ambipolar doping can be reached.

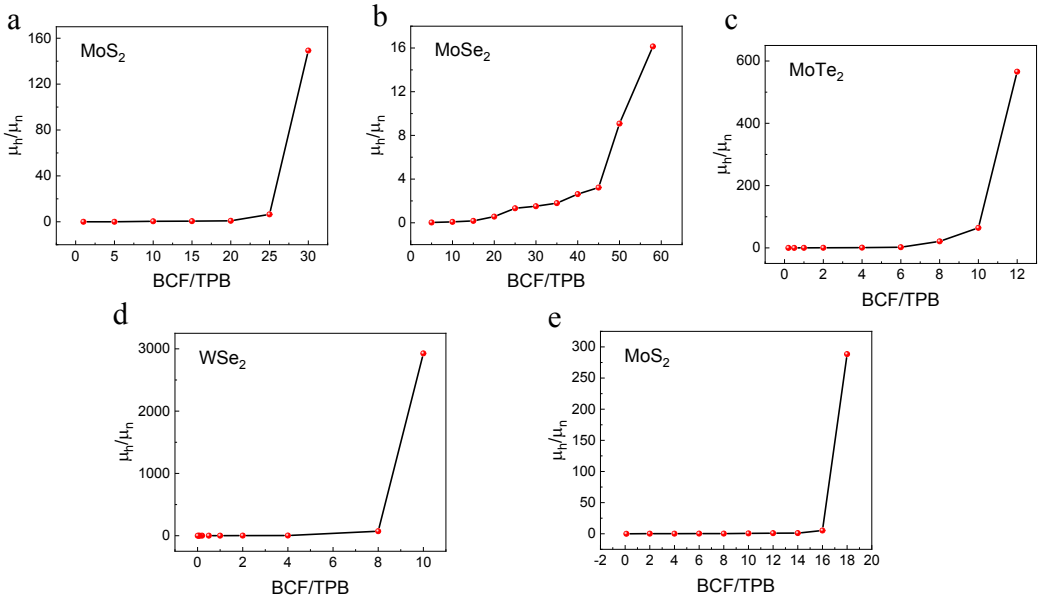

Figure S10. (a-e) The μ_h/μ_n ratio of MoS_2, $MoSe_2$, $MoTe_2$, WS_2, and WSe_2 as a function of BCF/TPB.

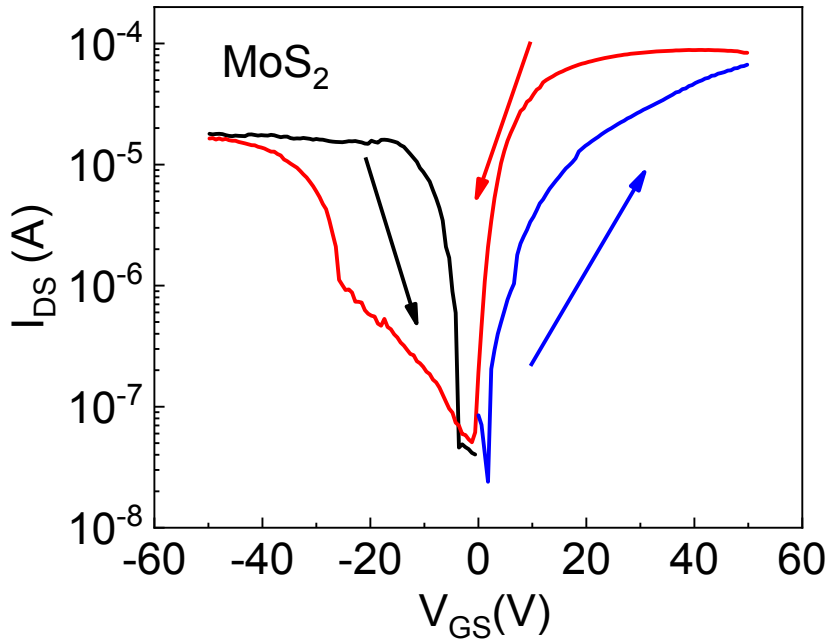

Figure S11. Transfer characteristic of TPB modulated MoS_2. (the V_{GS} sweep from 0V to 50 V (blue line), then 50V to -50 V (red line) and go back to 0 V (black line). The I_{DS} value decreases to the minimum with the increase of VGS and then increases when VGS scans from 0V to 50V, which means p-doping of MoS_2 at V_{GS}=0 V.

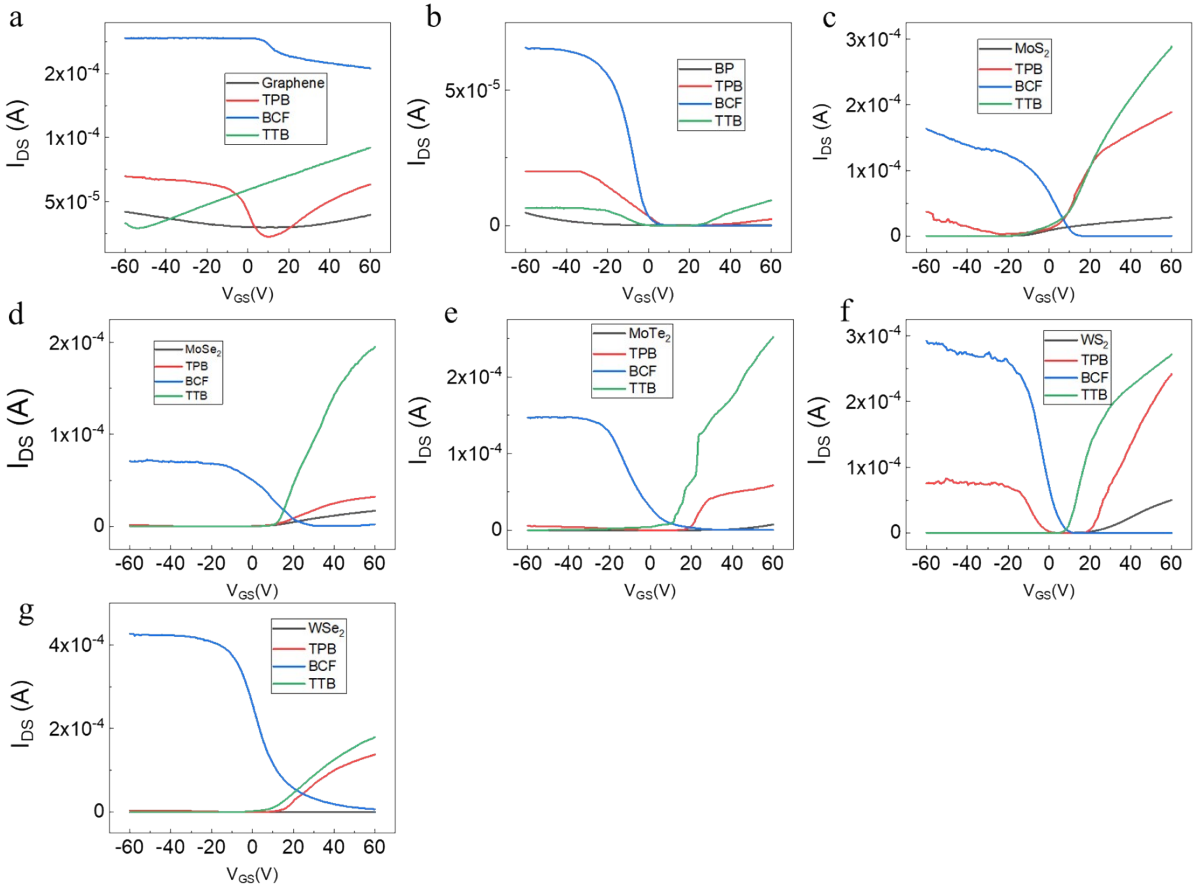

Figure S12. Transfer characteristics for pristine (black line), ambipolar (red line), p-type (blue line), and n-type modulated (green line) graphene **(a)**, BP **(b)**, MoS_2 **(c)**, $MoSe_2$ **(d)**, $MoTe_2$ **(e)**, WS_2 **(f)**, and WSe_2 **(g)** FETs

Table S1. Hole and electron carrier concentration of pristine, TPB, BCF, and TTB modulated graphene, BP, MoS_2, $MoSe_2$, $MoTe_2$, WS_2, and WSe_2 FETs.

State / 2d materials	Pristine		TPB		BCF	TTB
	n_h (10^{12} cm^{-2})	n_e (10^{12} cm^{-2})	n_h (10^{12} cm^{-2})	n_e (10^{12} cm^{-2})	n_h (10^{12} cm^{-2})	n_e (10^{12} cm^{-2})
Graphene	7.2	6.1	19.7	23.4	28.3	16.7
BP	6.7	-	21.6	15.3	30.1	21.9
MoS_2	-	5.3	26.3	31.7	29.4	36.5
$MoSe_2$	-	4.6	7.8	26.1	15.2	30.9
$MoTe_2$	-	5.1	8.1	16.2	18.9	28.6
WS_2	-	4.3	11.3	15.2	18.4	29.1
WSe_2	1.6	1.6	21.5	26.7	26.1	38.9

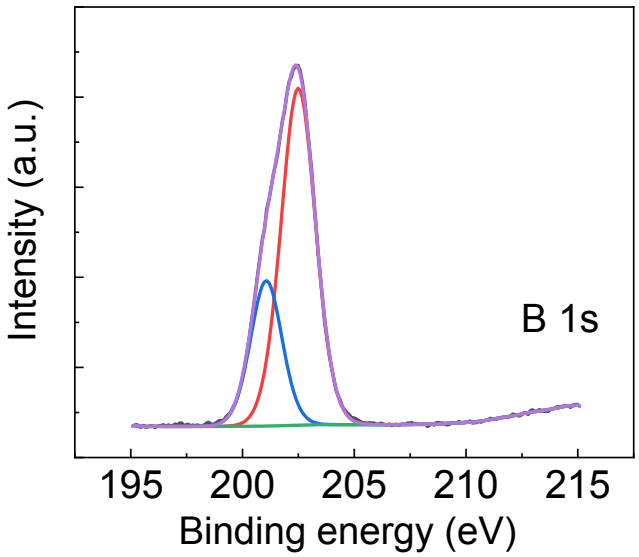

Figure S13. XPS core-level B 1s of TPB doped 2D materials.

Table S2. The Raman shift and doping concentration of TPB coted 2D materials.

2D materials	Graphene	BP	MoS_2	$MoSe_2$	$MoTe_2$	WS_2	WSe_2
Raman shift (cm^{-1})	1.1	1.2	1.2	1.3	1.7	0.7	1.4
Concentration (10^{13})	1.9	2.1	2.6	2.6	8.1	1.1	2.7

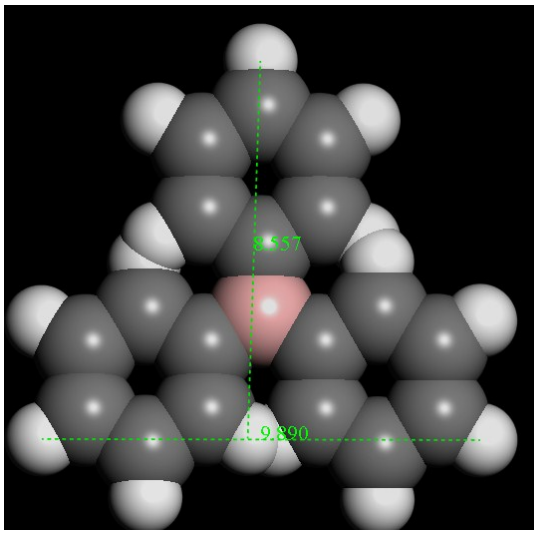

Figure S14. Top view of 3D model of BCF molecular. Consider the van der Waals radius of non-bonded H is 1.2 Å and C is 1.7 Å, the unit-cell area of BCF estimated to 71.1 $Å^2$.

Table S3. Estimates of TPB coverage (%) and the carrier concentration on the surface of the 2D materials.

2D materials	Graphene	BP	MoS_2	$MoSe_2$	$MoTe_2$	WS_2	WSe_2
Atomic Ratio	0.056±0.008	0.069±0.007	0.053±0.010	0.068±0.009	0.072±0.008	0.061±0.011	0.081±0.013
Coverage (%)	45 ± 8	37 ± 5	34 ± 6	38 ± 7	40 ± 8	42 ± 4	48 ± 5
Concentration (10^{13})	7.4±1.5	6.2±0.7	5.6±0.9	6.3±1.2	6.6±1.4	6.9±0.7	7.9±0.8

The coverage is calculated based on the ratio of dopant to 2D material relative to the theoretical value, which is estimated based on how many dopant monomers are closely packed on the surface of the 2D material.[1] Base 2D materials unit-cell area (≈0.0524 nm^2 for single-layer graphene (a= 0.246 nm), 0.1507 nm^2 for BP (a=0.453 nm, b=0.336 nm), 0.0843 nm^2 for single-layer MoS_2, WS_2; 0.0932 nm^2 for $MoSe_2$, WSe_2 and 0.1091 nm^2 for $MoTe_2$ (a=0.355 nm)),[2] and TPB unit-cell area of 71.1 $Å^2$ (Figure S14), together with dopant:2D materials ratio from XPS spectra, the coverage of a molecular monolayer on 2D materials surface and the doping concentration can be estimated and summarized in Table S2. The type of doping depends on the surface potential of the 2D material and TPB.

References

1 A. Tarasov, S. Zhang, M. Y. Tsai, P. M. Campbell, S. Graham, S. Barlow, S. R. Marder and E. M. Vogel, *Advanced Materials*, 2015, **27**, 1175–1181.

2 S. Zhang, H. M. Hill, K. Moudgil, C. A. Richter, A. R. Hight Walker, S. Barlow, S. R. Marder, C. A. Hacker and S. J. Pookpanratana, *Advanced Materials*, 2018, **30**, 1802991.