Supporting Information

Electronic Property Modulation in Two-dimensional Lateral Superlattices of Monolayer Transition Metal Dichalcogenides

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Table S1. Calculated lattice parameters, bond length, bandgap, and effective masses of electron and hole of MX_2 monolayers.

	a (Å)		*d* (Å)	E_g (eV)		*Effective mass* (m_0)		
	Cal.	Exp.	Cal.	Cal.	Exp.	y-direction	x-direction	m*
MoS_2	3.18	3.16[3]	2.417	1.68	2.4[6]	e: 0.48	e: 0.48	e:0.37[10]/0.44[11]/0.65[12]
	3.18[1]		2.41[2]	1.68[5]		h: 0.60	h: 0.59	h:0.44[10]/0.44[11]/0.69[12]
WS_2	3.18	3.153[4]	2.416	1.82	2.73[7]	e: 0.32	e: 0.32	0.40[11]/0.75[12]
	3.18[1]		2.41[2]	1.81[1]		h: 0.43	h: 0.43	0.43[11]/0.57[12]
$MoSe_2$	3.3	3.288[3]	2.546	1.45	2.18[8]	e: 0.56	e: 0.56	0.48[11]/0.74[12]
	3.32[1]		2.54[2]	1.44[1]		h: 0.68	h: 0.66	0.51[11]/0.77[12]
WSe_2	3.32	3.28[3]	2.546	1.55	2.12[9]	e: 0.35	e: 0.35	0.36[11]/0.48[12]
	3.32[2]		2.55[2]	1.54[5]		h: 0.47	h: 0.46	0.39[11]/0.62[12]

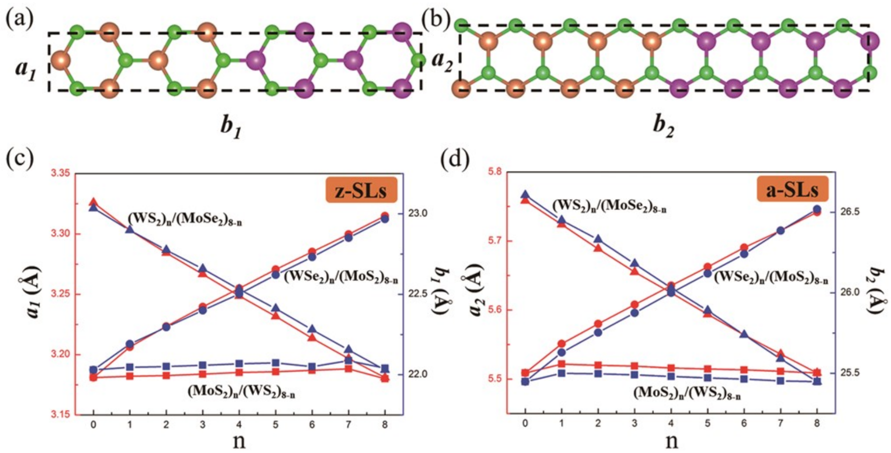

Fig. S1 (a,b) Schematic diagram of z- and a-SLs, respectively. (c,d) Lattice parameters evolution with compositions in 2D lateral TMD-HSs. The a_1, b_1, a_2 and b_2 represent the corresponding lattice parameters in (a,b).

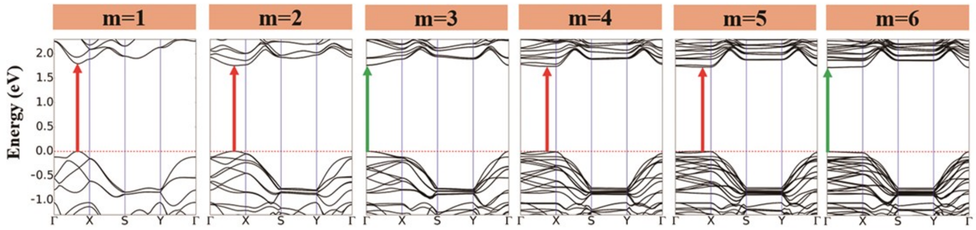

Fig. S2 Calculated band structures of lateral a-$(MoS_2)_m/(WS_2)_m$ SLs.

Table S2. Detailed Bandgaps of all studied 2D lateral TMD-SLs and alloys.

Heterostructures	Compositions	Bandgap (eV)			Effective mass (m_0)		
		zigzag	armchair	alloy	zigzag	armchair	alloy
MoS_2-WS_2	$(MoS_2)_1/(WS_2)_7$	1.7791	1.7580	1.7809	x-m_e/x-m_h 0.46/0.52 y-m_e/y-m_h 0.36/0.45	x-m_e/x-m_h 0.65/0.44 y-m_e/y-m_h 0.41/0.44	x-m_e/x-m_h 0.35/0.43 y-m_e/y-m_h 0.35/0.43
	$(MoS_2)_2/(WS_2)_6$	1.7460	1.7220	1.7538	0.51/0.54 0.40/0.47	0.77/0.47 0.46/0.46	0.37/0.46 0.37/0.46
	$(MoS_2)_3/(WS_2)_5$	1.7185	1.7060	1.7349	0.53/0.56 0.43/0.51	0.51/0.48 0.46/0.48	0.39/0.47 0.39/0.47
	$(MoS_2)_4/(WS_2)_4$	1.7015	1.6822	1.7147	0.54/0.57 0.45/0.53	0.61/0.53 0.47/0.50	0.41/0.49 0.42/0.50
	$(MoS_2)_5/(WS_2)_3$	1.6910	1.6790	1.6992	0.54/0.59 0.46/0.54	0.51/0.55 0.48/0.52	0.42/0.51 0.42/0.51
	$(MoS_2)_6/(WS_2)_2$	1.6841	1.6750	1.6873	0.54/0.62 0.45/0.56	0.55/0.54 0.48/0.54	0.44/0.53 0.44/0.53
	$(MoS_2)_7/(WS_2)_1$	1.6794	1.6730	1.6770	0.54/0.64 0.47/0.58	0.61/0.57 0.48/0.57	0.45/0.55 0.45/0.56
	$(MoS_2)_1/(WS_2)_1$	1.7136	1.7254		0.45/0.54 0.42/0.52	0.43/0.52 0.41/0.50	
	$(MoS_2)_2/(WS_2)_2$	1.7137	1.7		0.42/0.49 0.42/0.50	0.46/0.50 0.43/0.50	
	$(MoS_2)_3/(WS_2)_3$	1.7094	1.6973		0.43/0.49 0.41/0.52	0.41/0.47 0.43/0.49	
	$(MoS_2)_5/(WS_2)_5$	1.6895	1.6724		0.51/0.54 0.42/0.55	0.70/0.50 0.48/0.48	
	$(MoS_2)_6/(WS_2)_6$	1.6797	1.6631		0.60/0.62 0.58/0.62	0.68/0.68 0.46/0.47	
MoS_2-$MoSe_2$	$(MoS_2)_1/(MoSe_2)_7$	1.4538	1.4560	1.4578	0.58/0.64 0.60/0.60	0.49/0.67 0.55/0.64	0.53/0.63 0.57/0.60
	$(MoS_2)_2/(MoSe_2)_6$	1.4695	1.4720	1.4829	0.59/0.64 0.61/0.72	0.57/0.83 0.54/0.65	0.53/0.64 0.52/0.69
	$(MoS_2)_3/(MoSe_2)_5$	1.4839	1.4828	1.5088	0.58/0.65	0.68/0.68	0.52/0.61

					0.62/0.72	0.52/0.64	0.56/0.59
	$(MoS_2)_4/(MoSe_2)_4$	1.5069	1.5029	1.5434	0.56/0.65 0.57/0.73	0.53/0.69 0.50/0.65	0.50/0.62 0.57/0.59
	$(MoS_2)_5/(MoSe_2)_3$	1.5342	1.5330	1.5706	0.54/0.64 0.57/0.83	0.51/0.80 0.50/0.64	0.50/0.62 0.48/0.60
	$(MoS_2)_6/(MoSe_2)_2$	1.5789	1.5740	1.6018	0.52/0.63 0.47/0.84	0.54/0.86 0.49/0.63	0.50/0.60 0.51/0.58
	$(MoS_2)_7/(MoSe_2)_1$	1.6220	1.6270	1.6400	0.50/0.60 0.51/0.64	0.46/0.71 0.48/0.60	0.47/0.59 0.47/0.57
	$(MoS_2)_1/(MoSe_2)_1$	1.5149	1.5531		0.56/0.64 0.53/0.62	0.53/0.67 0.52/0.62	
	$(MoS_2)_2/(MoSe_2)_2$	1.5340	1.5258		0.56/0.63 0.53/0.67	0.49/0.63 0.50/0.60	
	$(MoS_2)_3/(MoSe_2)_3$	1.5156	1.5121		0.55/0.64 0.53/0.74	0.55/0.62 0.51/0.62	
	$(MoS_2)_5/(MoSe_2)_5$	1.4833	1.4889		0.57/0.66 0.66/0.79	0.64/0.70 0.51/0.67	
	$(MoS_2)_6/(MoSe_2)_6$	1.4613	1.4676		0.57/0.67 0.74/0.92	0.75/0.75 0.50/0.65	
MoS_2-WSe_2	$(MoS_2)_1/(WSe_2)_7$	1.5298	1.5020	1.5400	0.46/0.56 0.45/0.49	0.82/0.49 0.43/0.49	0.39/0.47 0.39/0.47
	$(MoS_2)_2/(WSe_2)_6$	1.4955	1.4640	1.5556	0.51/0.58 0.45/0.55	0.83/0.57 0.48/0.51	0.41/0.49 0.47/0.50
	$(MoS_2)_3/(WSe_2)_5$	1.4847	1.4700	1.5599	0.52/0.60 0.46/0.61	0.64/0.68 0.49/0.52	0.43/0.51 0.43/0.51
	$(MoS_2)_4/(WSe_2)_4$	1.4953	1.4699	1.5757	0.51/0.63 0.53/0.68	0.69/0.70 0.47/0.55	0.47/0.52 0.44/0.54
	$(MoS_2)_5/(WSe_2)_3$	1.5176	1.4960	1.6030	0.50/0.66 0.50/0.72	0.63/0.74 0.48/0.57	0.48/0.54 0.44/0.55
	$(MoS_2)_6/(WSe_2)_2$	1.5628	1.5430	1.6208	0.49/0.69 0.51/0.70	0.54/0.76 0.48/0.61	0.46/0.56 0.54/0.56
	$(MoS_2)_7/(WSe_2)_1$	1.6326	1.6190	1.6460	0.48/0.69 0.48/0.63	0.52/0.68 0.47/0.64	0.46/0.57 0.49/0.58
	$(MoS_2)_1/(WSe_2)_1$	1.6116	1.6173		0.52/0.54 0.43/0.53	0.45/0.56 0.44/0.54	
	$(MoS_2)_2/(WSe_2)_2$	1.5477	1.5219		0.49/0.54 0.44/0.53	0.46/0.53 0.45/0.50	
	$(MoS_2)_3/(WSe_2)_3$	1.5222	1.5009		0.49/0.53 0.43/0.53	0.45/0.62 0.47/0.52	
	$(MoS_2)_5/(WSe_2)_5$	1.4557	1.4304		0.53/0.52 0.68/0.86	0.72/0.71 0.55/0.56	
	$(MoS_2)_6/(WSe_2)_6$	1.4193	1.3997		0.53/0.51 0.82/1.10	0.83/0.83 0.50/0.49	

$MoSe_2$-WS_2	$(MoSe_2)_1/(WS_2)_7$	1.7033	1.7200	1.7493	0.32/0.43 0.32/0.43	0.44/0.45 0.43/0.43	0.34/0.43 0.35/0.48
	$(MoSe_2)_2/(WS_2)_6$	1.6773	1.6610	1.6848	0.44/0.45 0.43/0.43	0.43/0.49 0.47/0.58	0.39/0.47 0.39/0.46
	$(MoSe_2)_3/(WS_2)_5$	1.6044	1.6190	1.6279	0.42/0.53 0.47/0.50	0.42/0.53 0.47/0.50	0.42/0.50 0.43/0.50
	$(MoSe_2)_4/(WS_2)_4$	1.5808	1.5728	1.5750	0.51/0.58 0.50/0.62	0.51/0.58 0.50/0.62	0.46/0.51 0.46/0.59
	$(MoSe_2)_5/(WS_2)_3$	1.5357	1.5400	1.5456	0.46/0.58 0.46/0.62	0.46/0.58 0.46/0.62	0.47/0.56 0.50/0.55
	$(MoSe_2)_6/(WS_2)_2$	1.5102	1.5000	1.4925	0.52/0.59 0.49/0.72	0.52/0.59 0.49/0.72	0.49/0.59 0.47/0.66
	$(MoSe_2)_7/(WS_2)_1$	1.4686	1.4690	1.4636	0.54/0.61 0.60/0.60	0.54/0.61 0.60/0.60	0.51/0.62 0.52/0.68
	$(MoSe_2)_1/(WS_2)_1$	1.4804	1.5903		0.46/0.54 0.45/0.53	0.48/0.55 0.47/0.52	
	$(MoSe_2)_2/(WS_2)_2$	1.5882	1.5757		0.47/0.54 0.49/0.59	0.55/0.56 0.49/0.55	
	$(MoSe_2)_3/(WS_2)_3$	1.5683	1.5932		0.51/0.54 0.47/0.60	0.52/0.56 0.47/0.55	
	$(MoSe_2)_5/(WS_2)_5$	1.5651	1.566		0.56/0.55 0.53/0.66	0.88/0.68 0.52/0.56	
	$(MoSe_2)_6/(WS_2)_6$	1.5647	1.5659		0.61/0.57 0.60/0.69	0.93/0.72 0.50/0.58	
$MoSe_2$-WSe_2	$(MoSe_2)_1/(WSe_2)_7$	1.5162	1.4996	1.5121	0.41/0.47 0.44/0.48	0.81/0.43 0.42/0.46	0.38/0.46 0.40/0.48
	$(MoSe_2)_2/(WSe_2)_6$	1.4891	1.4690	1.4944	0.46/0.49 0.44/0.48	0.89/0.48 0.49/0.48	0.42/0.47 0.40/0.48
	$(MoSe_2)_3/(WSe_2)_5$	1.4682	1.4591	1.4705	0.49/0.50 0.48/0.54	0.51/0.53 0.49/0.50	0.45/0.52 0.46/0.60
	$(MoSe_2)_4/(WSe_2)_4$	1.4545	1.4436	1.4610	0.51/0.53 0.54/0.60	0.51/0.53 0.50/0.53	0.47/0.52 0.46/0.54
	$(MoSe_2)_5/(WSe_2)_3$	1.4504	1.4421	1.4465	0.50/0.55 0.54/0.60	0.56/0.60 0.52/0.54	0.47/0.55 0.51/0.60
	$(MoSe_2)_6/(WSe_2)_2$	1.4503	1.4402	1.4441	0.53/0.58 0.54/0.70	0.56/0.56 0.53/0.58	0.51/0.60 0.48/0.60
	$(MoSe_2)_7/(WSe_2)_1$	1.4462	1.4410	1.4357	0.53/0.61 0.54/0.60	0.72/0.56 0.54/0.61	0.51/0.59 0.48/0.54
	$(MoSe_2)_1/(WSe_2)_1$	1.4633	1.4837		0.45/0.54 0.48/0.57	0.48/0.55 0.47/0.53	
	$(MoSe_2)_2/(WSe_2)_2$	1.4605	1.4468		0.48/0.53 0.47/0.57	0.54/0.55 0.50/0.54	
	$(MoSe_2)_3/(WSe_2)_3$	1.4628	1.4492		0.49/0.53 0.45/0.57	0.50/0.50 0.49/0.53	

	$(MoSe_2)_5/(WSe_2)_5$	1.4431	1.4254		0.52/0.52 0.57/0.63	0.64/0.58 0.59/0.56	
	$(MoSe_2)_6/(WSe_2)_6$	1.4390	1.4182		0.54/0.52 0.65/0.69	0.81/0.76 0.53/0.51	
WS_2-WSe_2	$(WS_2)_1/(WSe_2)_7$	1.5719	1.5721	1.5743	0.37/0.45 0.35/0.48	0.34/0.45 0.34/0.44	0.34/0.42 0.34/0.45
	$(WS_2)_2/(WSe_2)_6$	1.5976	1.5948	1.6034	0.36/0.45 0.35/0.45	0.32/0.47 0.34/0.44	0.34/0.43 0.34/0.41
	$(WS_2)_3/(WSe_2)_5$	1.6155	1.6150	1.6318	0.37/0.45 0.39/0.56	0.38/0.48 0.34/0.45	0.35/0.43 0.34/0.46
	$(WS_2)_4/(WSe_2)_4$	1.6464	1.6415	1.6714	0.35/0.45 0.34/0.50	0.32/0.45 0.33/0.45	0.33/0.43 0.34/0.42
	$(WS_2)_5/(WSe_2)_3$	1.6776	1.6742	1.7118	0.36/0.45 0.39/0.56	0.35/0.54 0.33/0.45	0.32/0.44 0.32/0.44
	$(WS_2)_6/(WSe_2)_2$	1.7275	1.7197	1.7410	0.34/0.45 0.39/0.56	0.32/0.52 0.33/0.44	0.32/0.42 0.33/0.45
	$(WS_2)_7/(WSe_2)_1$	1.7727	1.7724	1.7801	0.34/0.43 0.35/0.40	0.30/0.44 0.32/0.43	0.31/0.41 0.30/0.40
	$(WS_2)_1/(WSe_2)_1$	1.6165	1.6804		0.39/0.45 0.36/0.43	0.34/0.45 0.33/0.44	
	$(WS_2)_2/(WSe_2)_2$	1.67	1.6536		0.35/0.45 0.35/0.46	0.32/0.43 0.33/0.43	
	$(WS_2)_3/(WSe_2)_3$	1.6489	1.6454		0.37/0.45 0.35/0.47	0.32/0.45 0.33/0.45	
	$(WS_2)_5/(WSe_2)_5$	1.6286	1.6262		0.36/0.45 0.36/0.47	0.41/0.60 0.33/0.46	
	$(WS_2)_6/(WSe_2)_6$	1.6144	1.6038		0.36/0.46 0.43/0.68	0.42/0.59 0.33/0.45	

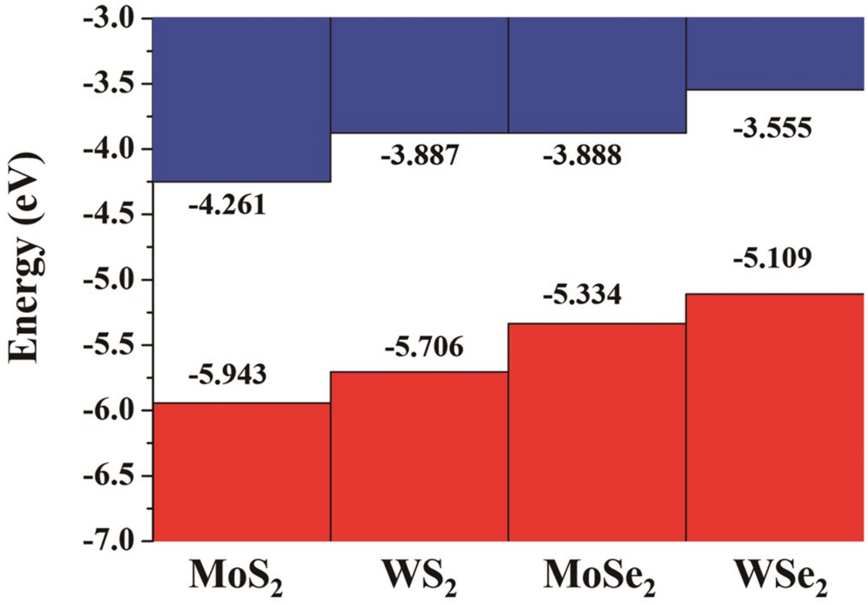

Fig. S3 Calculated band alignment of TMDs with PBE functional.

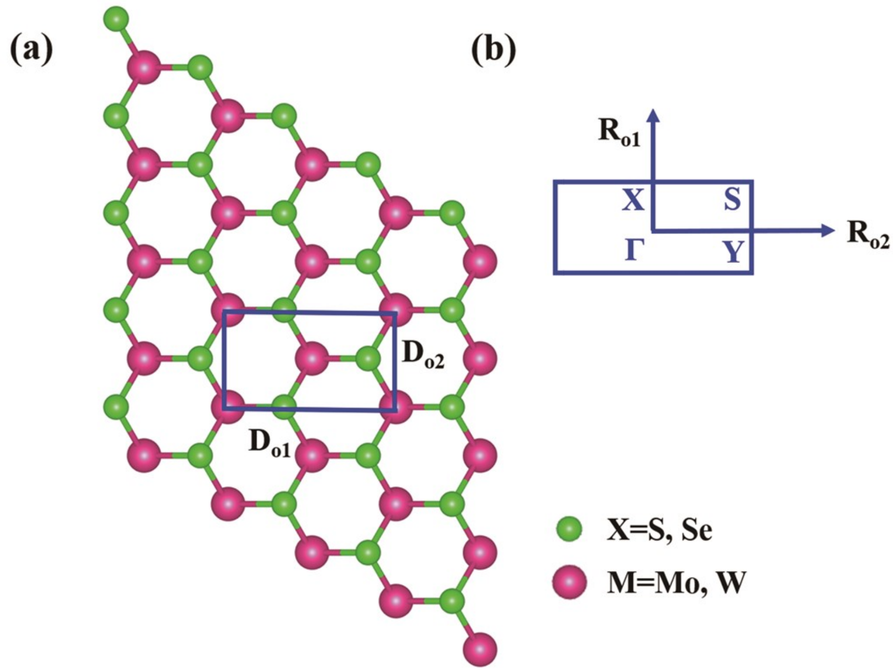

Fig. S4 (a) Atomic structure of monolayer MX_2(M=Mo, W; X=S, Se), where the orthogonal supercell (defined by D_{o1} and D_{o2}) is enclosed with blue frames. (b) The corresponding first Brillouin zones of the orthogonal supercell. R_{o1}, R_{o2} represent the orthogonal axes of the orthorhombic cell.

Table S3. Calculated band-edge energy levels of lateral $(MoS_2)_4/(WSe_2)_4$ SLs. The energy levels with strain-free from the band alignment calculations, without considering the lattice mismatch. Moreover, the bandgap is all listed. The red numbers represent the band-edge position of these SLs.

		MoS_2	WSe_2	Bandgap (eV)
Strain-free	CBM (eV)	-4.261	-3.555	0.848
	VBM (eV)	-5.943	-5.109	
Zigzag	CBM (eV)	-4.410	-3.458	0.667
	VBM (eV)	-5.901	-5.077	
Armchair	CBM (eV)	-4.409	-3.432	0.662
	VBM (eV)	-5.902	-5.071	

References

1. W. Wei, Y. Dai, Q. Sun, N. Yin, S. Han, B. Huang and T. Jacob, *Physical Chemistry Chemical Physics*, 2015, **17**, 29380–29386.
2. J. Kang, S. Tongay, J. Zhou, J. Li and J. Wu, *Appl. Phys. Lett.*, 2013, **102**, 012111.
3. R. Coehoorn, C. Haas, J. Dijkstra, C. J. F. Flipse, R. A. de Groot and A. Wold, *Phys. Rev. B*, 1987, **35**, 6195–6202.
4. W. J. Schutte, J. L. De Boer and F. Jellinek, *Journal of Solid State Chemistry,* 1987, **70**, 207–209..
5. H. Wang, W. Wei, F. Li, B. Huang and Y. Dai, *Phys. Chem. Chem. Phys*., 2018, **20**, 25000–25008.
6. Y. L. Huang, Y. Chen, W. Zhang, S. Y. Quek, C.-H. Chen, L.-J. Li, W.-T. Hsu, W.-H. Chang, Y. J. Zheng, W. Chen and A. T. S. Wee, *Nature Communications*, 2015, **6**, 6298.
7. B. Zhu, X. Chen and X. Cui, *Scientific Reports,* 2015, **5**, 9218.
8. M. M. Ugeda, A. J. Bradley, S.-F. Shi, F. H. da Jornada, Y. Zhang, D. Y. Qiu, W. Ruan, S.-K. Mo,

Z. Hussain, Z.-X. Shen, F. Wang, S. G. Louie and M. F. Crommie, *Nature Materials*, 2014, **13**, 1091–1095.

9. C. Zhang, Y. Chen, A. Johnson, M.-Y. Li, L.-J. Li, P. C. Mende, R. M. Feenstra and C.-K. Shih, *Nano Lett*., 2015, **15**, 6494–6500.
10. H. Peelaers and C. G. Van de Walle, *Phys. Rev. B*, 2012, **86**, 241401.
11. X. Yuan, M. Yang, L. Wang and Y. Li, *Physical Chemistry Chemical Physics*, 2017, **19**, 13846–13854..
12. Y. Sun, X. Wang, X.-G. Zhao, Z. Shi and L. Zhang, *Journal of Semiconductors,* 2018, **39**, 072001.