Electronic Supplementary Information for

Twisted MoS_2/MX_2 heterobilayers: Effect of van der Waals interaction on the electronic structure

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Table S1. Computed lattice constant (in Å) and layer distance *d* (in Å) of the MX_2/MoS_2 heterobilayers.

	0°	**13.2°**	**21.8°**	**27.8°**	**32.2°**	**38.2°**	**46.8°**	**60°**
System	T	√19	√7	√13	√13	√7	√19	C7
$CrSe_2$	3.21	13.97	8.48	11.55	11.55	8.47	13.97	3.21
$CrSe_2$_*d*	3.14	3.41	3.41	3.44	3.44	3.44	3.41	3.16
$MoSe_2$	3.26	14.19	8.62	11.74	11.74	8.62	14.19	3.26
$MoSe_2$_*d*	3.11	3.41	3.41	3.40	3.41	3.41	3.42	3.12
WSe_2	3.26	14.20	8.62	11.75	11.75	8.62	14.20	3.26
WSe_2_*d*	3.11	3.41	3.41	3.41	3.41	3.41	3.41	3.11
WS_2	3.19	13.91	8.44	11.50	11.50	8.44	13.91	3.19
WS_2_*d*	3.10	3.34	3.38	3.35	3.37	3.37	3.37	3.10
CrS_2	3.13	13.65	8.28	11.28	11.28	8.28	13.64	3.13
CrS_2_*d*	3.10	3.35	3.36	3.35	3.36	3.36	3.36	3.18

Table S2. Computed direct bandgap E_{g1} (K-K, in eV) and indirect bandgap E_{g2} (Γ-K, in eV) of the MX_2/MoS_2 heterobilayers.

	0°	**13.2°**	**21.8°**	**27.8°**	**32.2°**	**38.2°**	**46.8°**	**60°**
$CrSe_2$_E_{g1}	0.76	0.76	0.76	0.77	0.77	0.77	0.76	0.76

$CrSe_2\_E_{g2}$	0.65	0.79	0.79	0.80	0.81	0.82	0.79	0.69
$CrSe_2\_E_{g1\_SOC}$	0.68	0.69	0.69	0.69	0.69	0.69	0.69	0.69
$CrSe_2\_E_{g2\_SOC}$	0.62	0.76	0.76	0.77	0.77	0.78	0.76	0.66
$MoSe_2\_E_{g1}$	0.80	0.74	0.73	0.74	0.74	0.73	0.74	0.75
$MoSe_2\_E_{g2}$	0.75	0.88	0.87	0.87	0.88	0.86	0.89	0.74
$MoSe_2\_E_{g1\_SOC}$	0.67	0.61	0.60	0.61	0.60	0.60	0.61	0.62
$MoSe_2\_E_{g2\_SOC}$	0.76	0.88	0.86	0.86	0.86	0.86	0.88	0.74
$WSe_2\_E_{g1}$	0.62	0.55	0.54	0.54	0.54	0.54	0.55	0.57
$WSe_2\_E_{g1}$	0.69	0.79	0.79	0.78	0.78	0.79	0.79	0.67
$WSe_2\_E_{g1\_SOC}$	0.34	0.27	0.26	0.26	0.26	0.26	0.27	0.30
$WSe_2\_E_{g2\_SOC}$	0.66	0.72	0.73	0.71	0.71	0.73	0.72	0.64
$WS_2\_E_{g1}$	1.53	1.46	1.47	1.47	1.47	1.47	1.46	1.47
$WS_2\_E_{g2}$	1.18	1.22	1.26	1.23	1.24	1.24	1.24	1.16
$WS_2\_E_{g1\_SOC}$	1.27	1.21	1.21	1.21	1.21	1.21	1.21	1.22
$WS_2\_E_{g2\_SOC}$	1.18	1.22	1.25	1.23	1.24	1.24	1.23	1.16
$CrS_2\_E_{g1}$	0.61	0.62	0.66	0.67	0.67	0.66	0.63	0.64
$CrS_2\_E_{g2}$	0.33	0.43	0.44	0.44	0.45	0.44	0.44	0.39
$CrS_2\_E_{g1\_SOC}$	0.44	0.49	0.47	0.50	0.50	0.49	0.50	0.50
$CrS_2\_E_{g2\_SOC}$	0.31	0.41	0.41	0.42	0.43	0.42	0.42	0.37

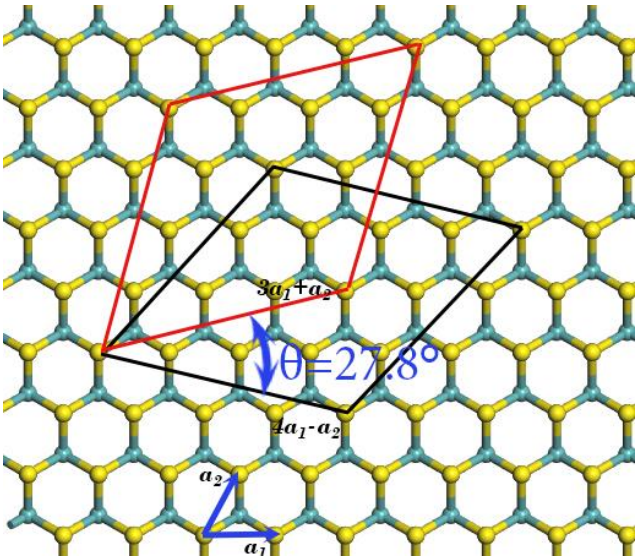

Figure S1. A schematic plot of the skewed supercell in a hexagonal lattice. The basis vector is (n$\mathbf{a}_1$+m$\mathbf{a}_2$). The rotation angle is θ.

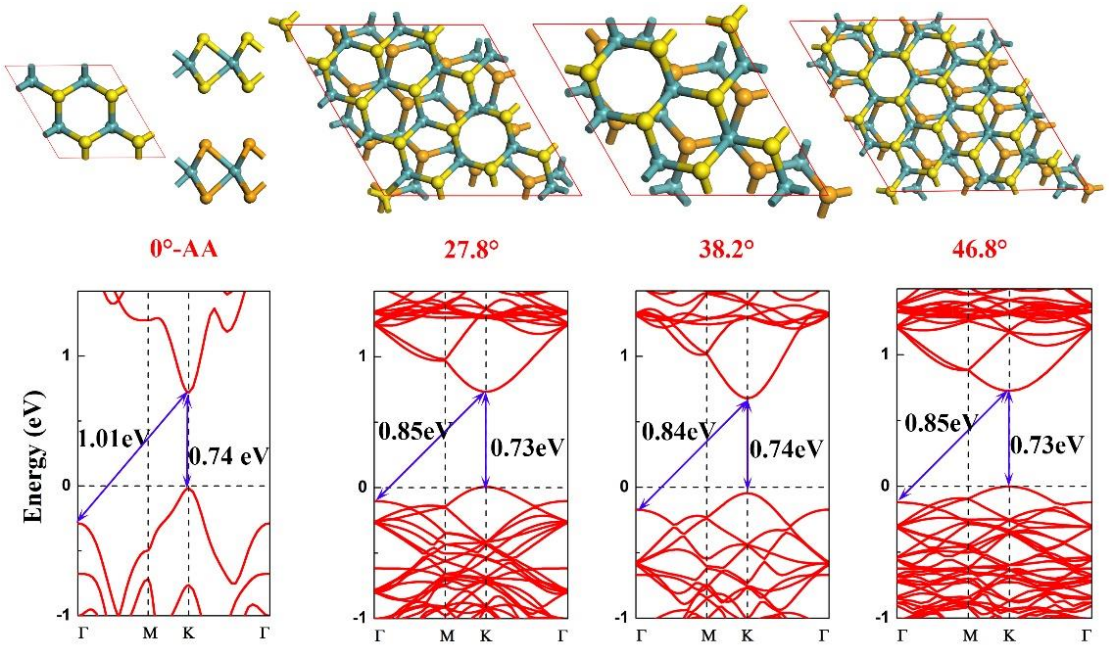

Figure S2. Atomic structures and band structures of the twisted heterobilayer $MoS_2/MoSe_2$ with the AA stacking order as the initial configuration. The electronic properties versus the rotation angle are almost identical to those of the corresponding $MoS_2/MoSe_2$ with the T-stacking order as the initial configuration.

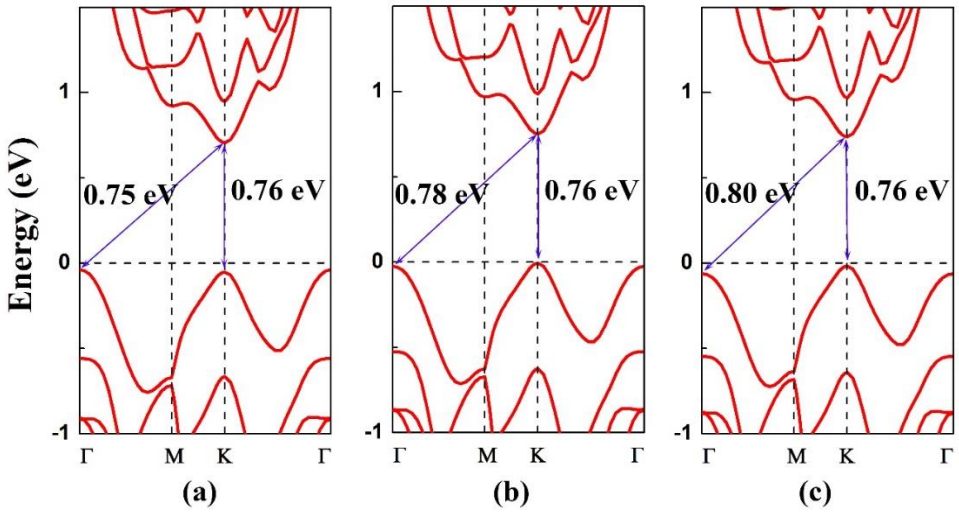

Figure S3. Computed band structures of the C7 stacking for $CrSe_2/MoS_2$ heterobilayer with enlarging the interlayer distance to (a) 3.3 Å, (b) 3.4 Å and (c) 3.5 Å.

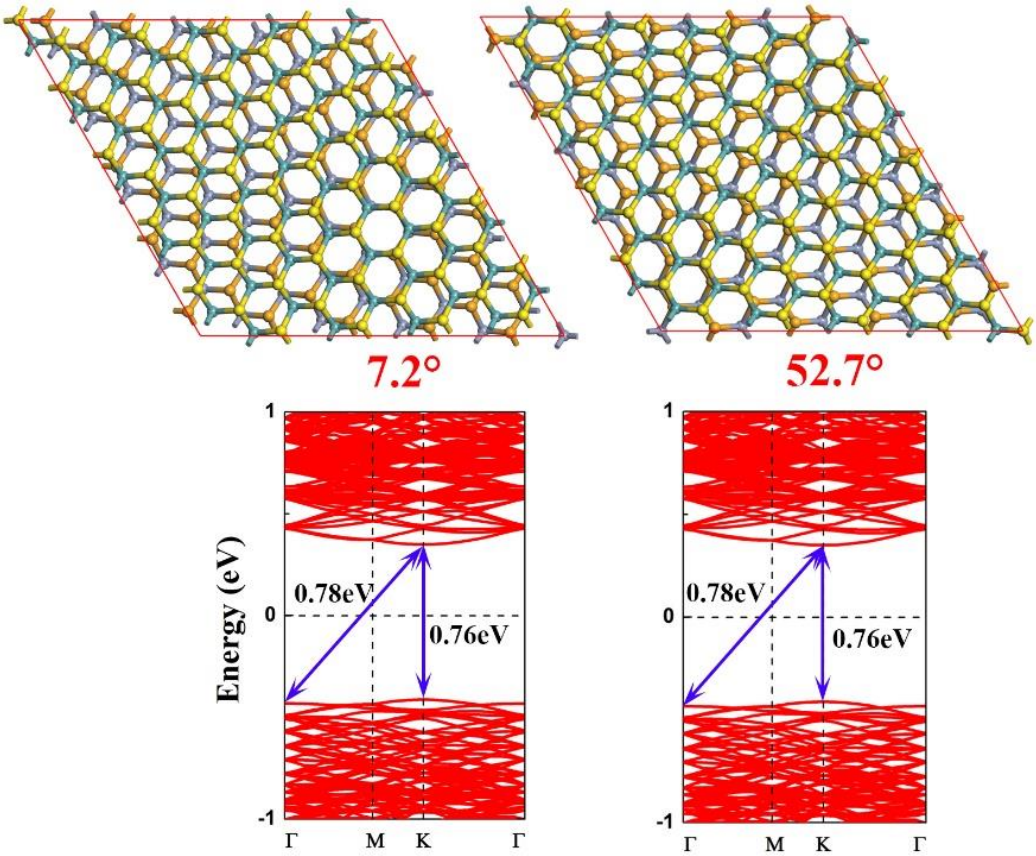

Figure S4. Computed band structures of $CrSe_2/MoS_2$ heterobilayer with rotation angle of 7.2 °and 52.7 °. The interlayer distance is 3.40 Å for both 7.2 °and 52.7 °.

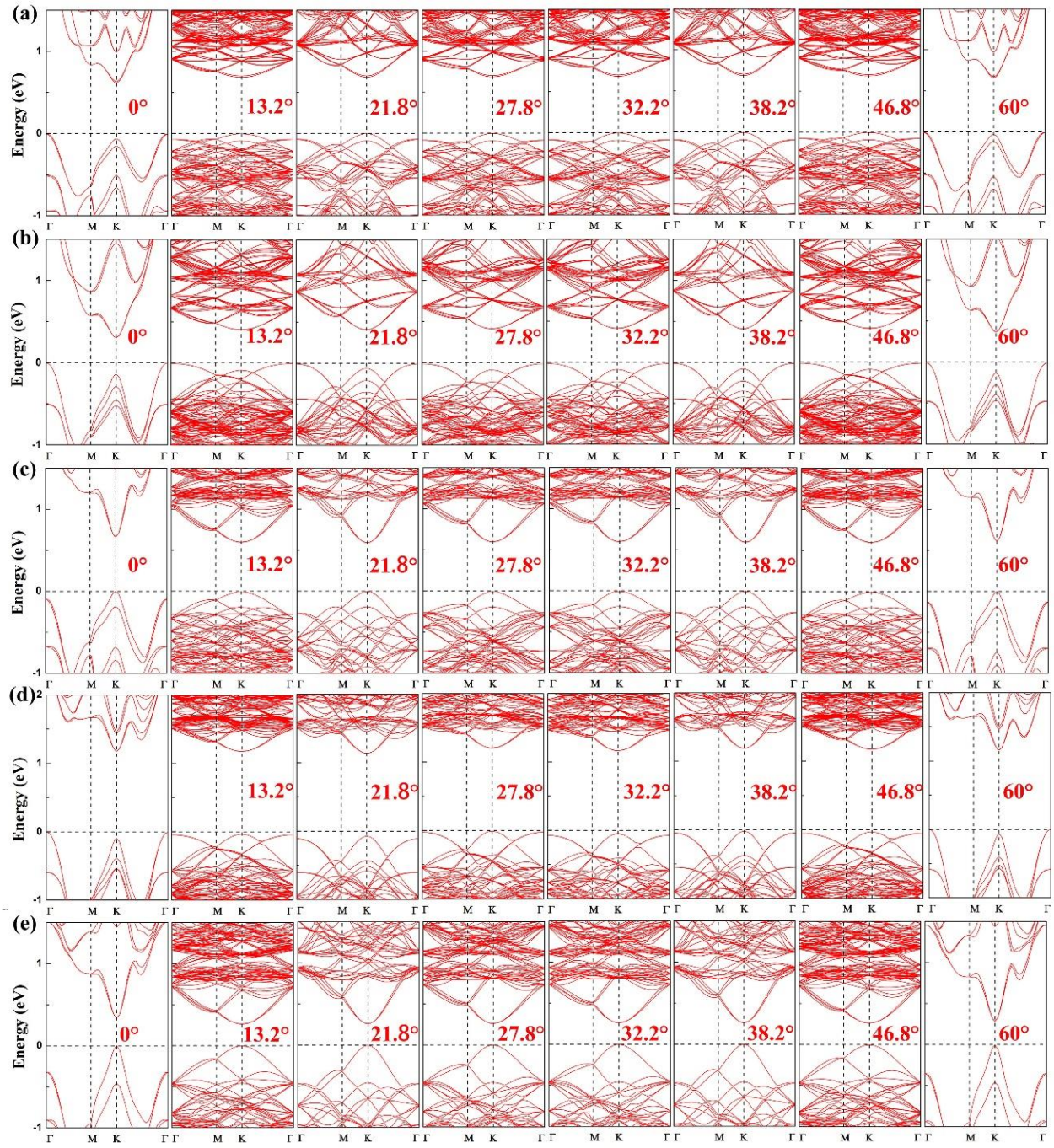

Figure S5. Computed band structures (PBE+SOC) of the heterobilayers of (a) $CrSe_2/MoS_2$, (b) CrS_2/MoS_2, (c) $MoSe_2/MoS_2$, (d) WS_2/MoS_2, and (e) WSe_2/MoS_2.