Rashba Splitting Modulated by Tuned Intrinsic Dipole Moment in MoXY/WXY Heterostructure.

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Open-Minded

1) 2D Janus Materials: An overview

2D Inorganic Janus transition-metal dichalcogenides MXY (M = Mo or W) with X=S, Se, and Te atoms at mirrori-image sites are promising for numerous applications, owing to their unique properties such as:

- Degenerate gaps at K and K' due to **lack of inversion symmetry:** Valley degree of freedom [1],
- Non-zero electric dipole moment [2] and nonlinear optical response due to **out-of-plane structural asymmetries**,
- Strong SOC splitting and locking of electron spin in split bands: Efficient Spin Field Effect Transistors,
- Type II band alignment (in heterostructure): large band offset, effective charge transfer (CT), and e and h localization.

2) Computational Methodology:

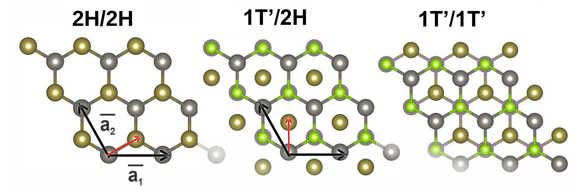
DFT modeling using FHI-aims code [3].

Supercell(1x1)'s of MoXY/WXY (6 atoms) with

- **all-electron** description using **atom-centered** basis set,
- **GGA** Exchange-Correlation parametrized as PBE functional,
- QM Many-Body Dispersion for dipole-dipole interactions.

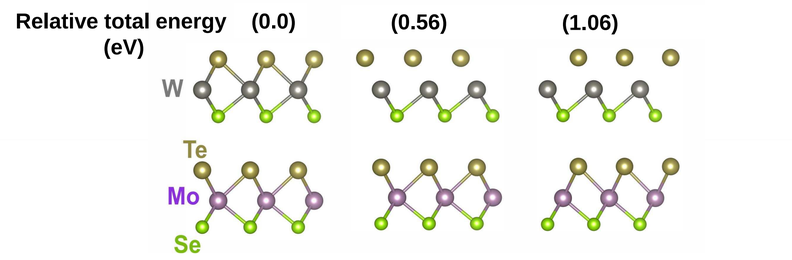
3) Structural Relaxations:

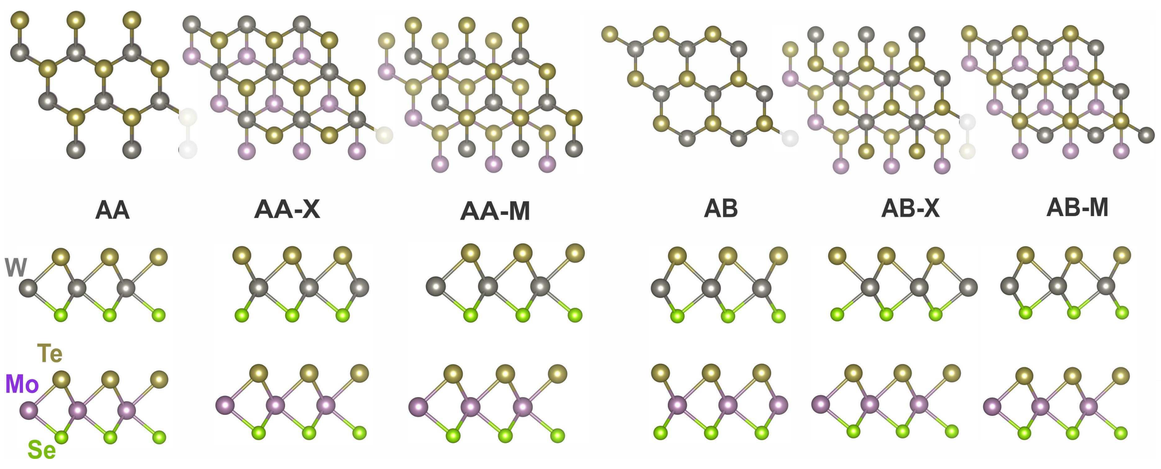
MoXY/WXY with different phases, chalcogenide atom sequences (CAS) and stackings:



Enthalpy of formation (meV)

Phases	2H	1T'
MoSeTe	160.7	236.7
WSeTe	400.0	19.3



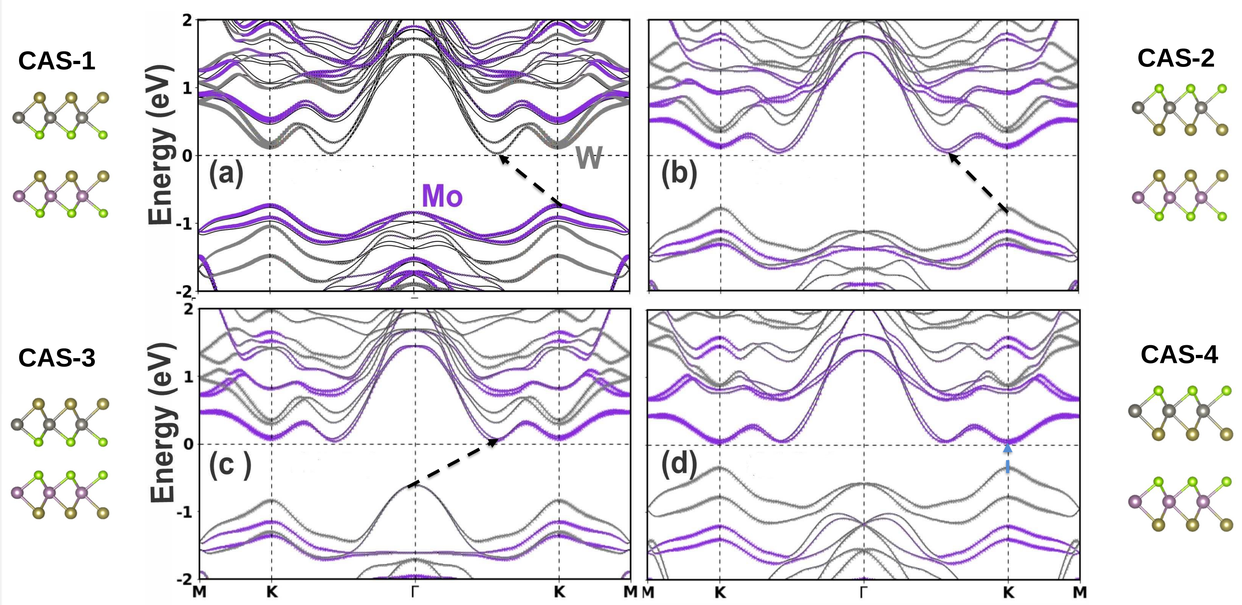


4) Electronic Structures:

- Band alignment, and band gap depends on chalcogenide atomic sequence (CAS) and stacking [4].
- Large band offset (BO) for CM at K/K′ for large net dipole moments (more effective CT).

Electric dipole moment (D_{tot}), band gap energy (E_{BG}), and band offset (BO) of MoSeTe/WSeTe.

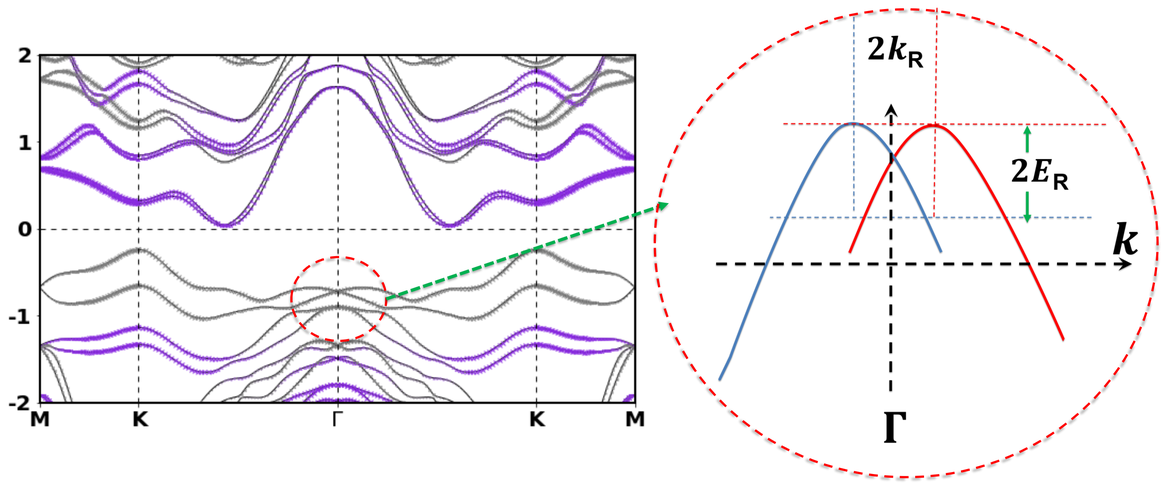
D_{tot} (e/Å)	E_{BG} (eV) (BO (meV))	AA	AA-X	AA-M	AB	AB-X	AB-M
-1.37	CAS1	0.80 (33.4)	0.74	0.85	0.78	0.83	0.79
-0.07	CAS2	0.87 (17.1)	0.88	0110	0.82	0.88	0.87
-0.02	CAS3	0.89 (19.1)	0.72	0.69	0.74	0.74	0.90
1.28	CAS4	0.25 (70.0)	0.41	0.31	0.39	0.37	0.30



5) Atomic Order-Tuned Rashba Splitting

- Atomic order change influences Rashba energy (E_R)

$$H_R = \frac{\hbar}{4m^2c^2}\nabla_\perp V.(p\times\sigma) \quad E_\pm(k) = \frac{\hbar^2k^2}{2m} \pm \alpha k = \frac{\hbar^2}{2m}(k\pm k_R)^2 - E_R \quad \alpha = \frac{2E_R}{k_R}$$



Rashba parameters for most stable bilayer stackings

			MoSeTe/WSeTe	MoSTe/WSTe	MoSSe/WSSe
MoSeTe/WSeTe	$2E_R$(meV)	k_R(Å^{-1})	α (meV Å)	α (meV Å)	α (meV Å)
CAS1	44.8	0.121	369.9	183.5	203.4
CAS2	68.0	0.117	577.8	414.6	80.0
CAS3	57.0	0.028	204.3	418.2	305.9
CAS4	51.0	0.097	531.5	473.0	271.2

6) Summary and Outlook:

- 2H/2H the most stable bilayer despite more stable 1T'-WSeTe,
- Band alignment, offset and gap depend on dipole moment direction,
- Indirect-to-direct bandgap transition by varying the atomic order,
- Rashba parameters tuned by atomic order and stacking,
- Beyond DFT methods: more accurate Rashba parameters

References:

[1] Nat. Phys., 10, 343, (2014).
[2] Phys. Rev. B, 106, 235414 (2022)
[3] Comp. Phys. Comm.180, 2175, (2009).
[4] J. Phys.: Cond. Matt. 31, 125003, (2019).

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