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$CrSBr$-MoS_2 heterostructures and the limits of VASP parallelization

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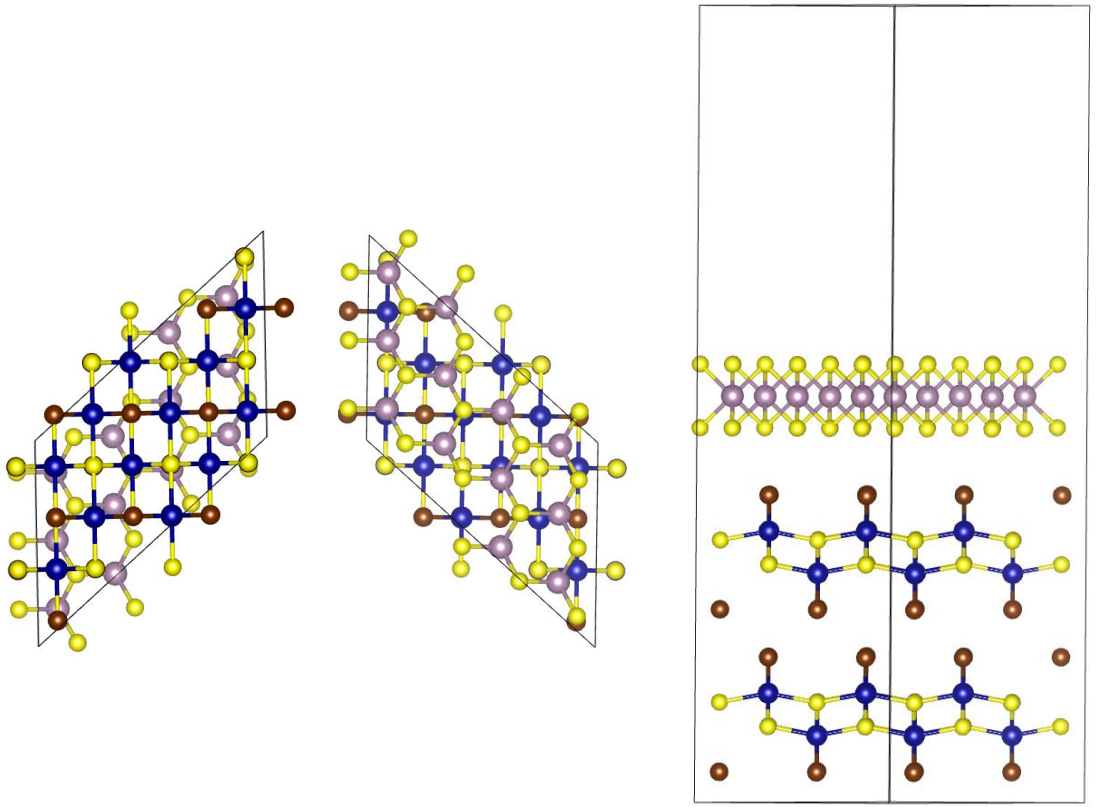
Part of AMULET project

Motivation-Key findings

- Controlling magnetic, optical, electronic properties in 2D materials via proximity effect → Spintronic, valleytronic applications
- CrSBr: layered semiconducting antiferromagnet. Interlayer coupling with MoS_2 → modifies lattice and valley dynamics
- Proximity-induced magnetization of MoS_2
- Band spin splitting due to proximity effect, enhanced by spin-orbit coupling
- Charge redistribution, bandgap renormalization
- Type-II heterostructure

Computational details



- Vienna Ab-Initio Simulation Package (VASP)
- Spin-polarized calculations (magnetic moment of Cr = 3 μ_B)
- GGA/PBE and HSE06 level of theory
- Hubbard correction for PBE (3.5eV for Cr)
- D3(BJ) dispersion correction (vdW)
- Optical absorption with Independent Particle Approximation (IPA)

Approach:

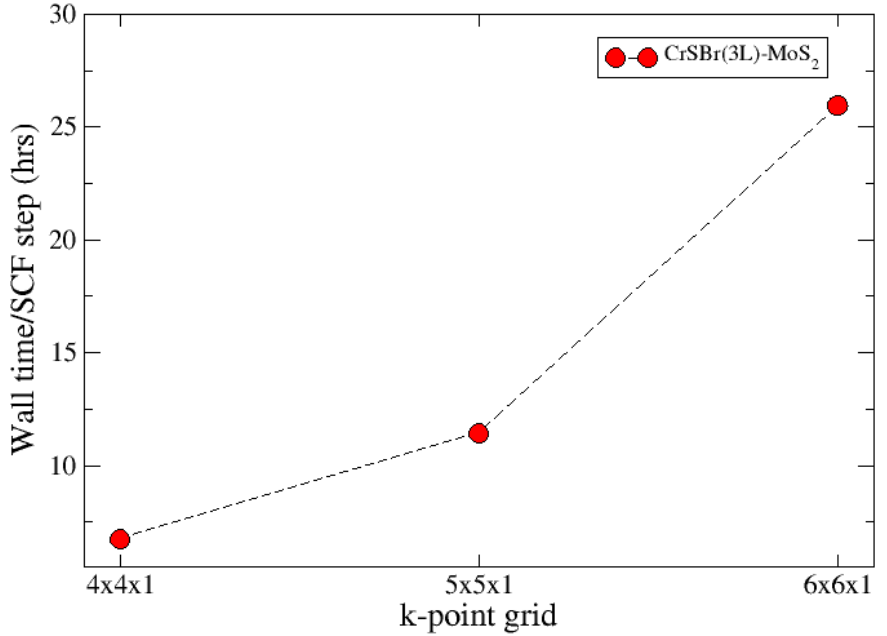
- Geometry optimization (PBE)
- Electronic structure, DOS, magnetization (PBE/HSE06)
- Band structure (PBE)
- Work function (PBE)
- IPA (PBE/HSE06)

Working with VASP in Karolina cluster

- OPEN-34-40 project “vdW heterostructures for electronic applications”
- 76 100 node-hours (Karolina: 47 700; Barbora NG: 28 400)
- CPU nodes of 128 cores each
- Karolina limit for a single job: 20 nodes
- VASP “limit”: 18 nodes (2,304 cores). Above that: bottleneck
- When are 16-18 nodes needed?
  - Band structure calculations
  - HSE06 calculations
  - Less: work function, optical properties

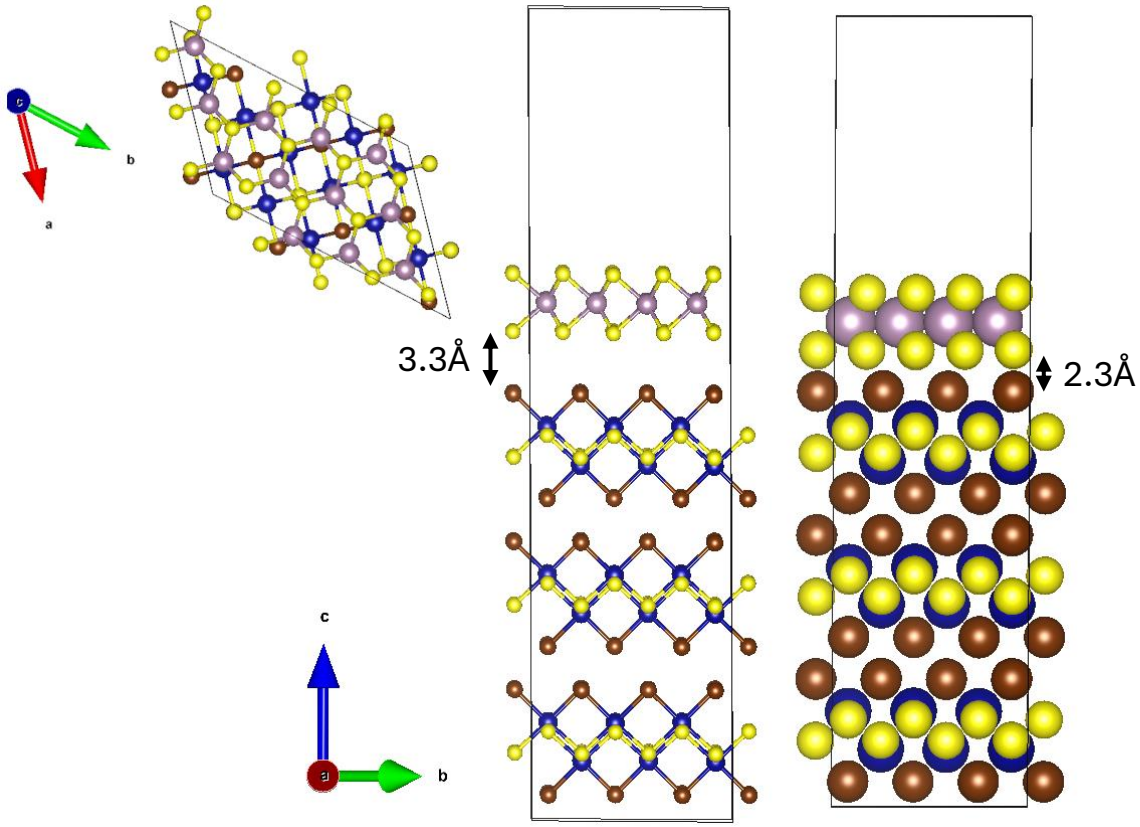
Working with VASP in Karolina cluster

- What about the time needed?
  - ➢ Mostly crucial for HSE06 SCF calculations
  - ➢ Highly dependent on k-points grid
- Maximum allowed time: 144 wall-time hours
- Graph: Time required for 1 SCF step with HSE06. An SCF cycle of converged geometry is 10-30 SCF steps → stop and restart



4x4x1: 20 SCF steps
5x5x1: 12 SCF steps
6x6x1: 5.5 SCF steps

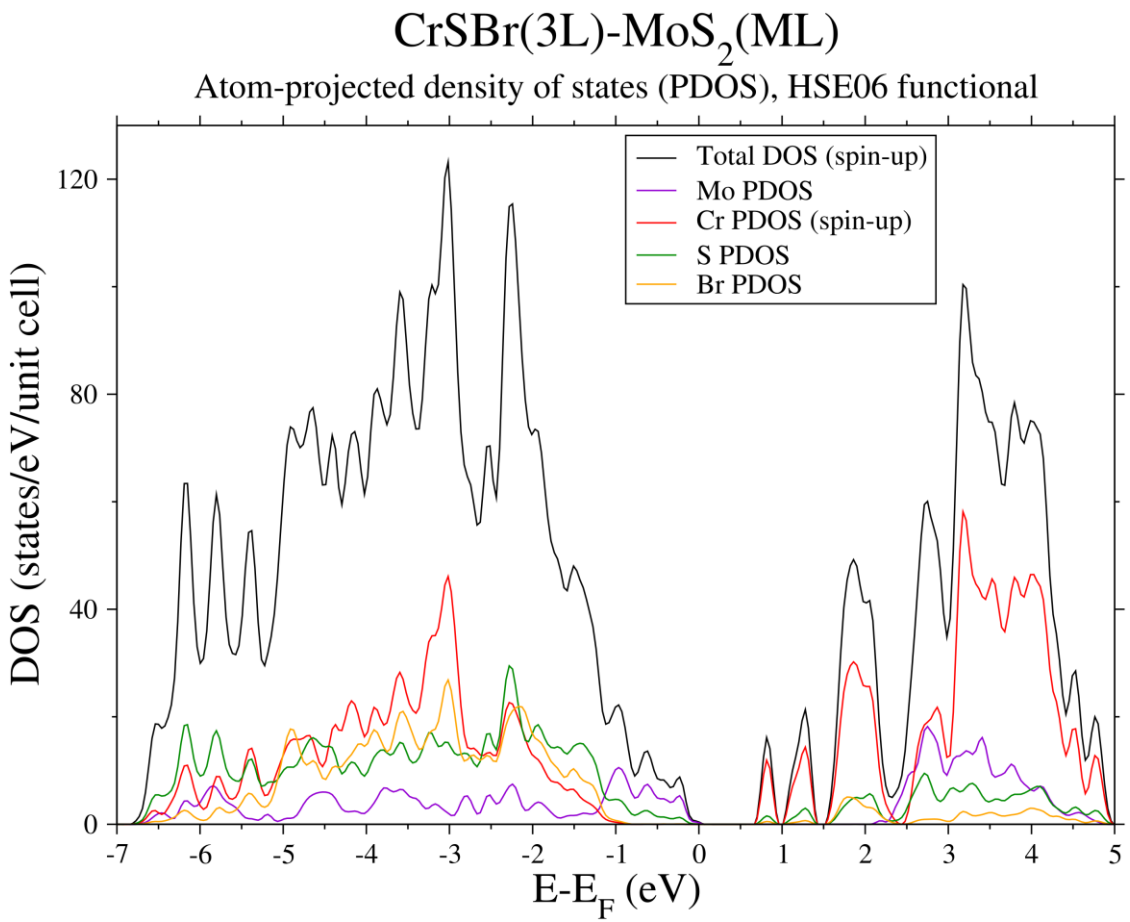
Constructing the heterostructure

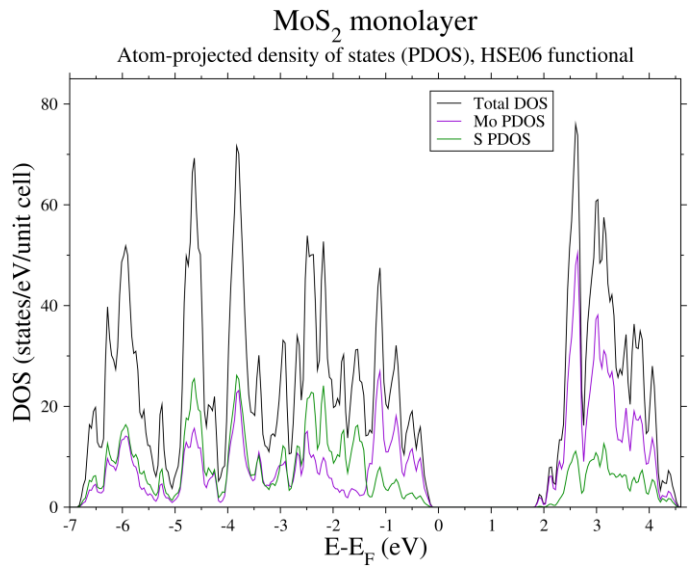


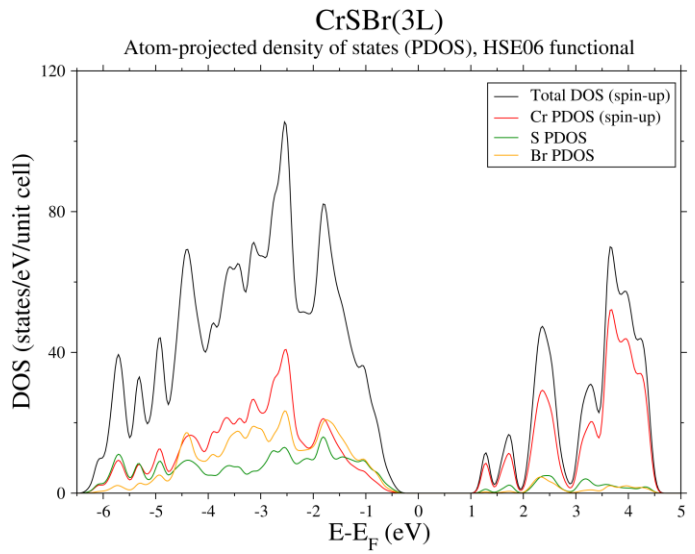
CrSBr (3L) with MoS_2 ML

- MoS_2: monolayer (non-magnetic, semiconducting)
- CrSBr: 1 to 4 layers in computations (~12 layers in experiment)
- Why the large supercell?
  - Small strain (1%) due to lattice mismatch
  - Larger with a smaller supercell (4-5%)
  - More expensive simulations

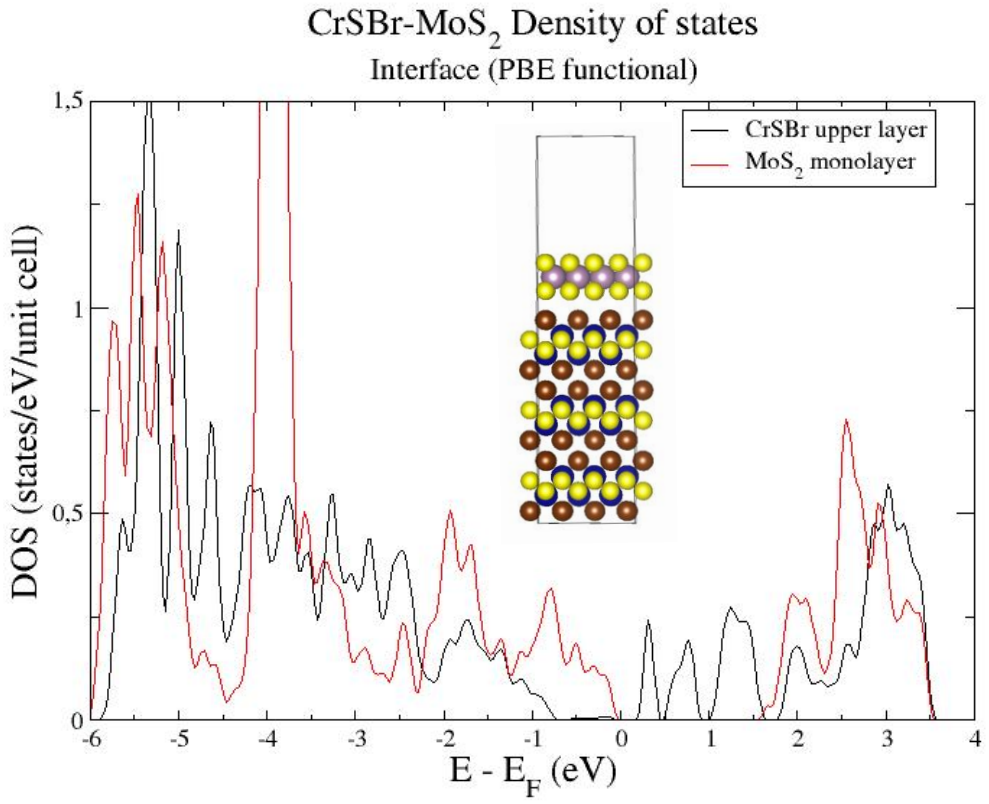
Electronic structure: DOS



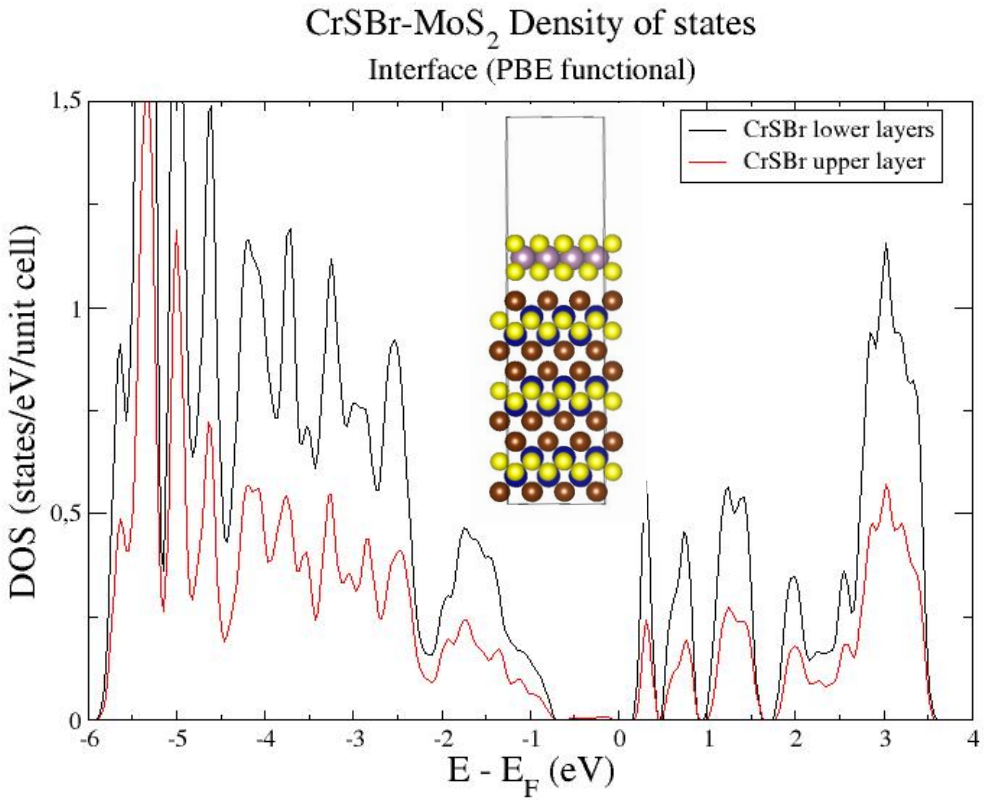




Electronic structure: DOS

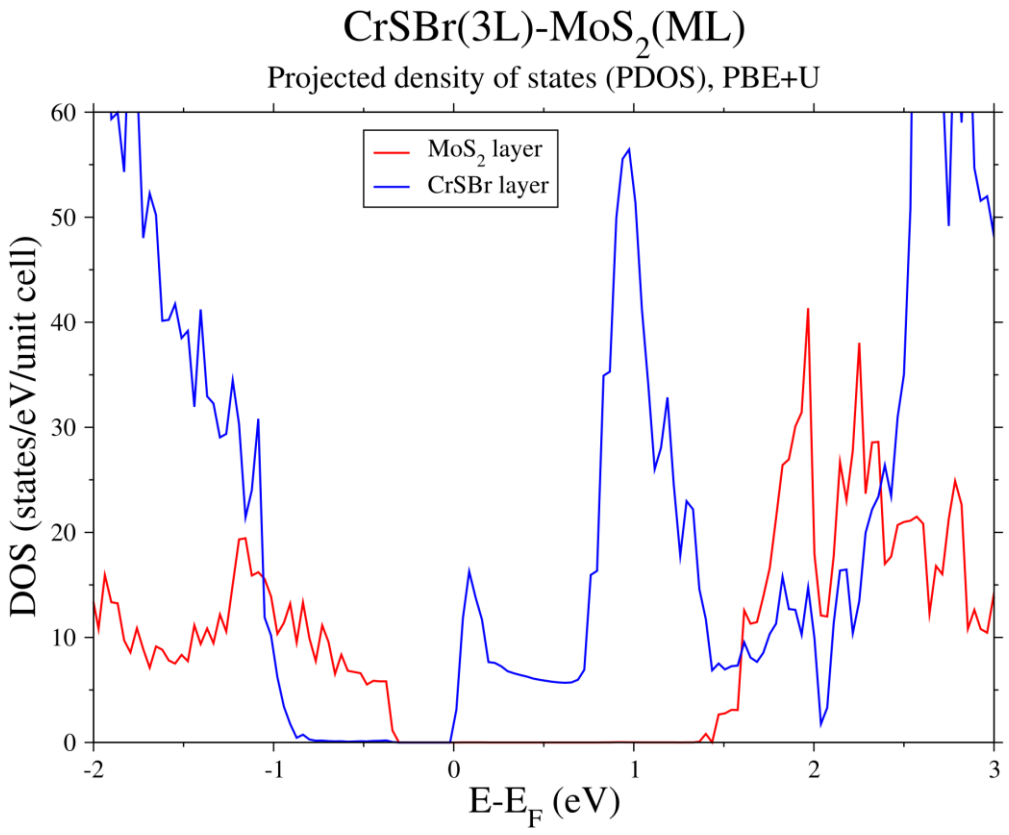


Narrowing of bandgap mainly due to MoS_2 layer in VBM (Mo(d) orbitals)

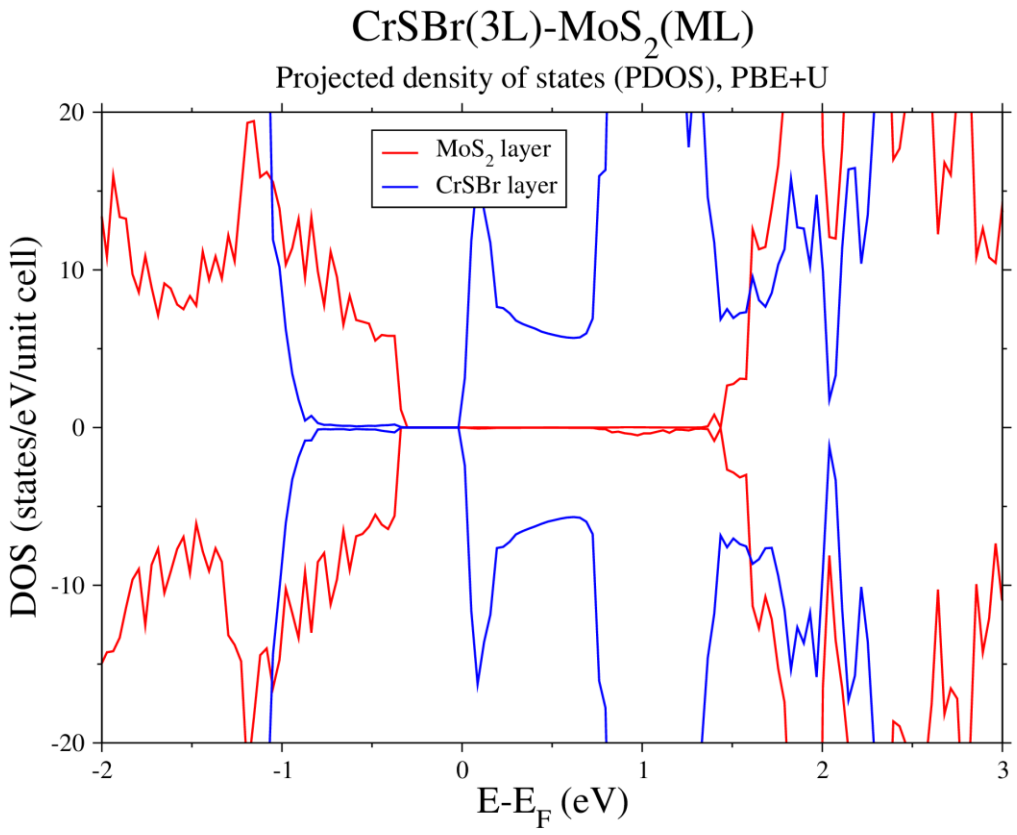


Contribution near VBM mainly due to upper layer

Electronic structure: DOS



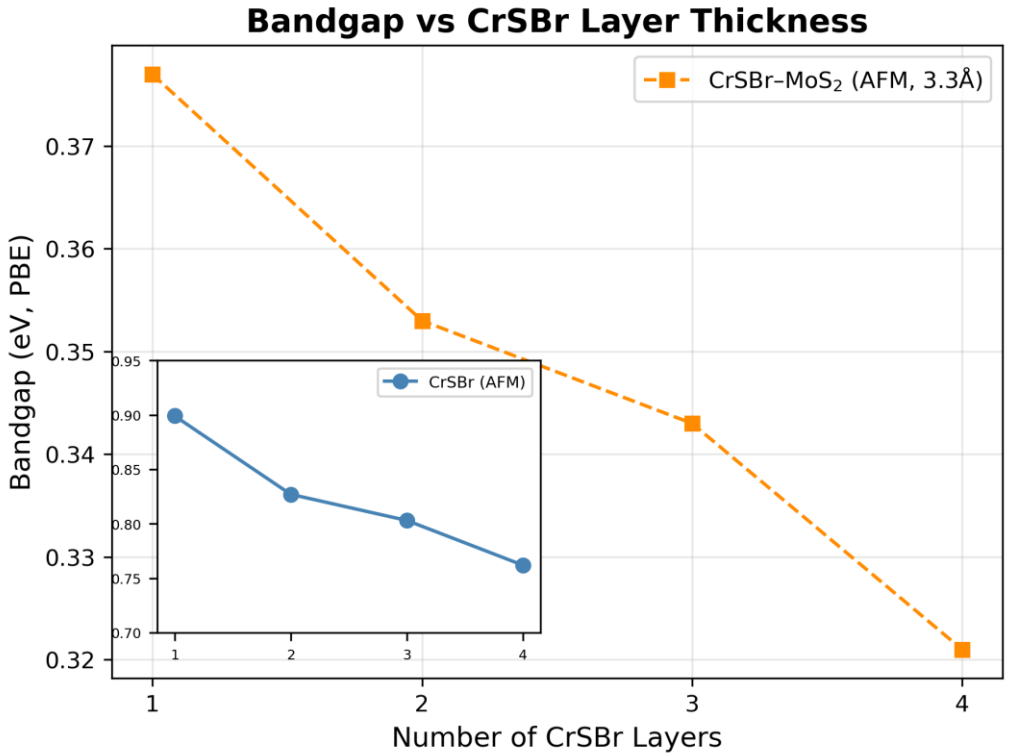
CBM of CrSBr is below CBM of MoS_2 and VBM of CrSBr is below VBM of MoS_2 (staggered gap)



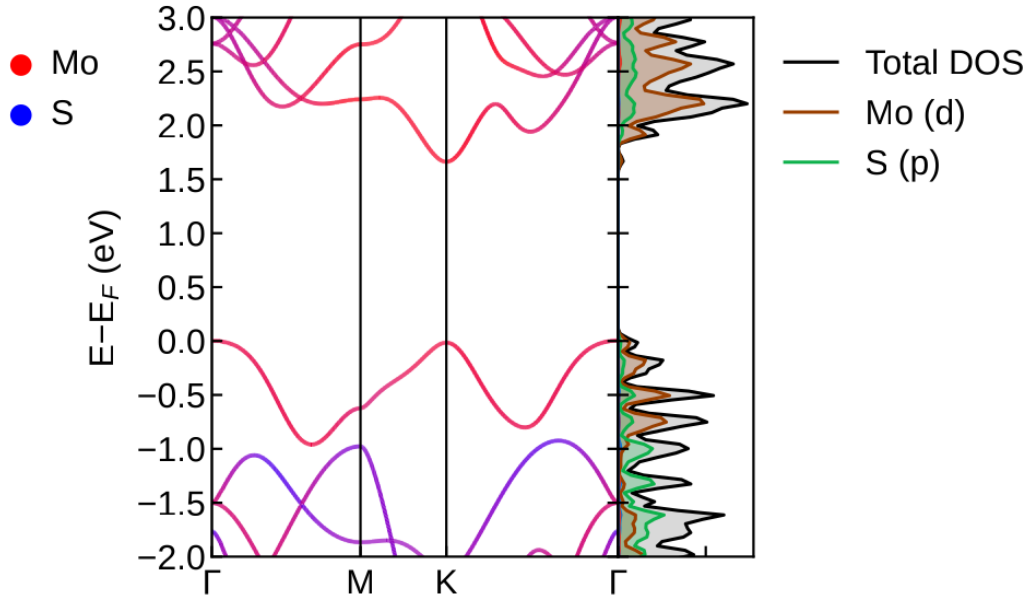
"Asymmetry" in MoS_2 part (states in gap) due to magnetic proximity

Electronic structure: bandgaps

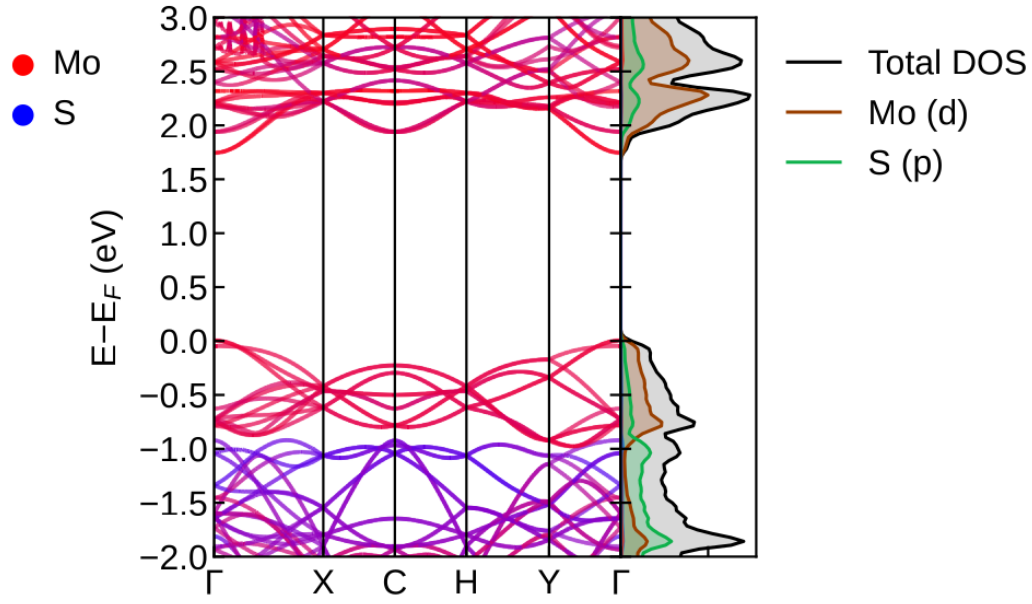
Structure	E_g PBE (eV)	E_g HSE06 (eV)
Bulk CrSBr AFM	0.765	1.636
Bulk CrSBr FM	0.621	
CrSBr(1L) AFM	0.883	
CrSBr(1L)-MoS_2 AFM 3.2 Å	0.377	
CrSBr(2L) AFM	0.818	
CrSBr(2L)-MoS_2 AFM 3.3 Å	0.353	
CrSBr(3L) AFM	0.797	1.622
CrSBr(3L) FM	0.662	
CrSBr(4L) AFM	0.759	
CrSBr(4L) FM	0.648	
MoS_2(ML) non-magnetic	1.737	2.146
CrSBr(3L)-MoS_2 AFM 3.3 Å	0.343	0.949
CrSBr(3L)-MoS_2 FM 3.3 Å	0.283	
CrSBr(3L)-MoS_2 AFM 2.3 Å	0.038	
CrSBr(4L)-MoS_2 AFM 3.3 Å	0.321	
CrSBr(4L)-MoS_2 FM 3.3 Å	0.290	



Band structure



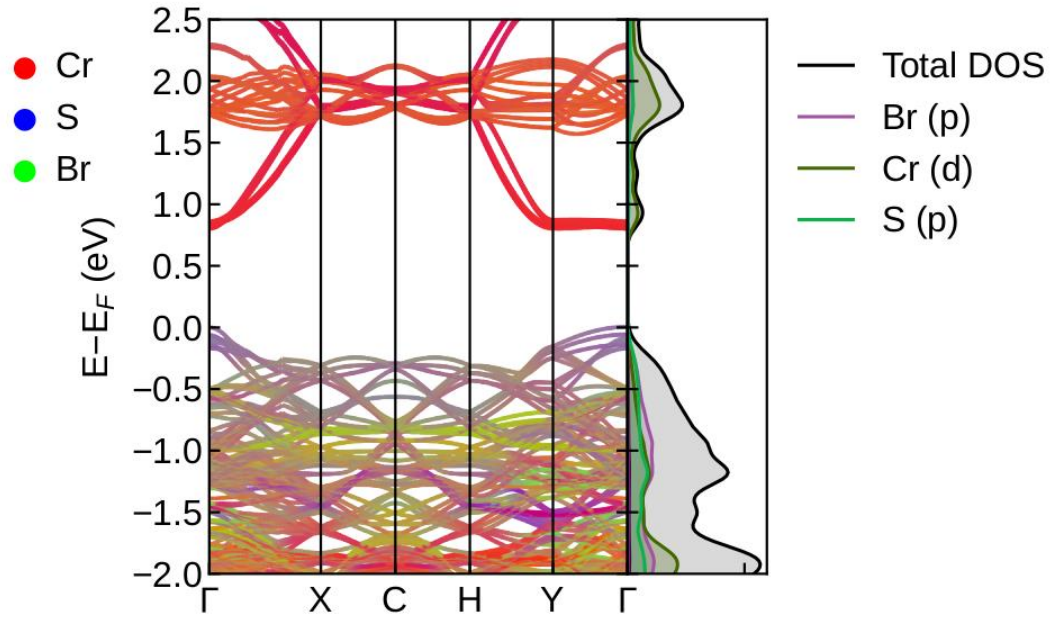
MoS_2 (ML) primitive unit cell



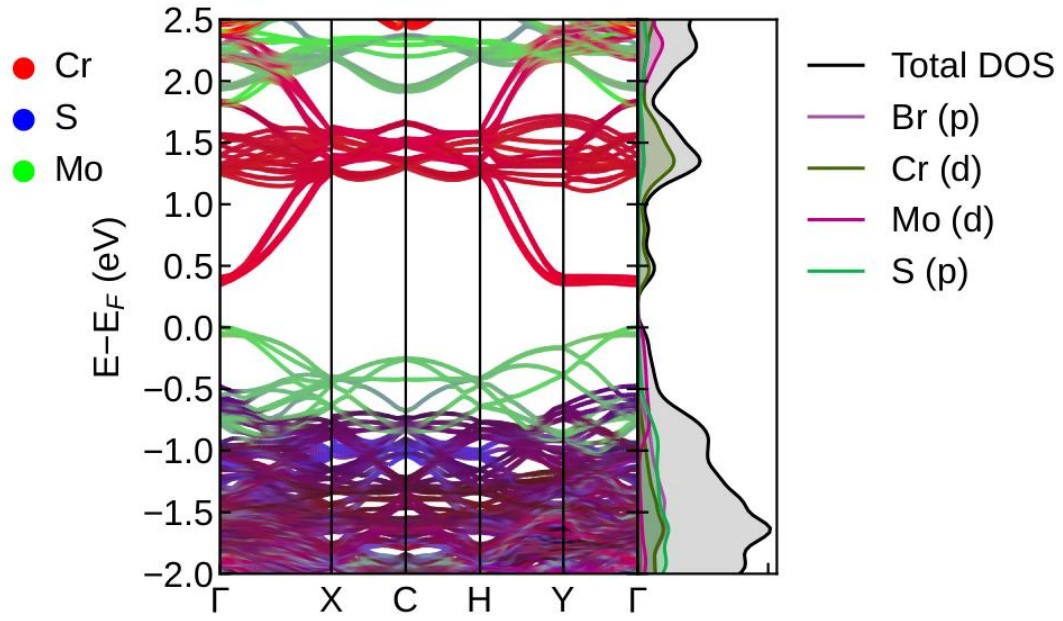
MoS_2 (ML) supercell

Larger unit cell → Smaller BZ, lowering of symmetry → Different k-path
Bands are folded → Direct bandgap appears on Γ point instead on K

Band structure

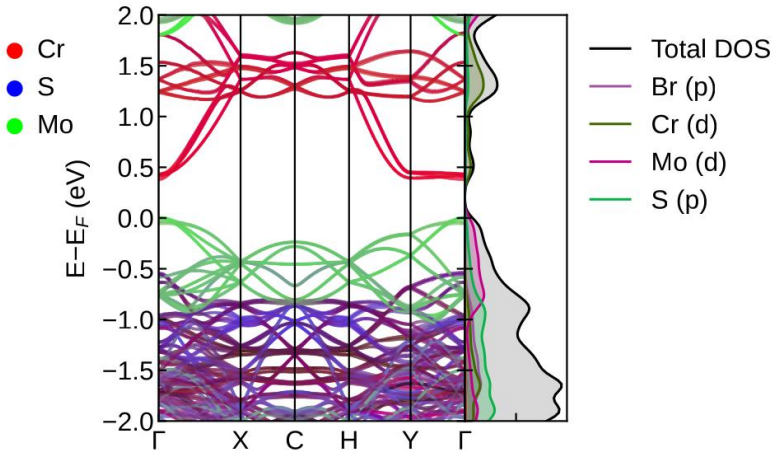


CrSBr (3L)

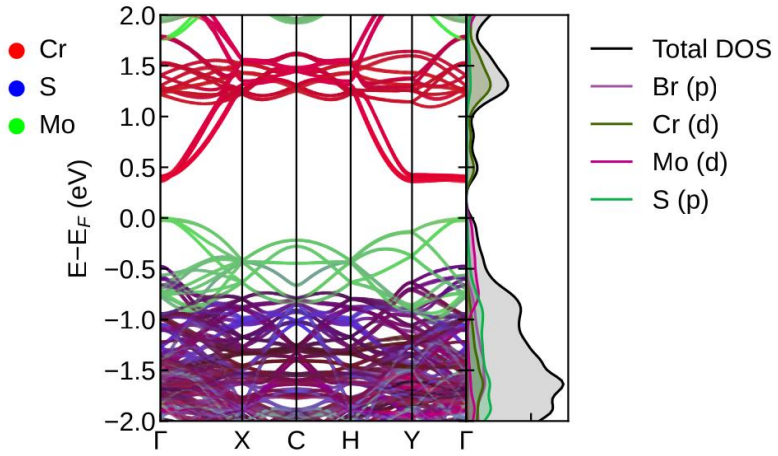


CrSBr(3L)-**MoS_2** HS

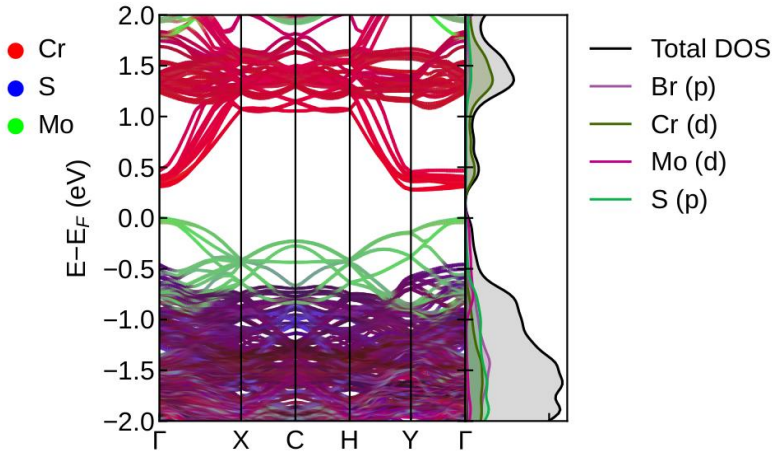
Band structure



CrSBr(1L)-**MoS$_2$** HS

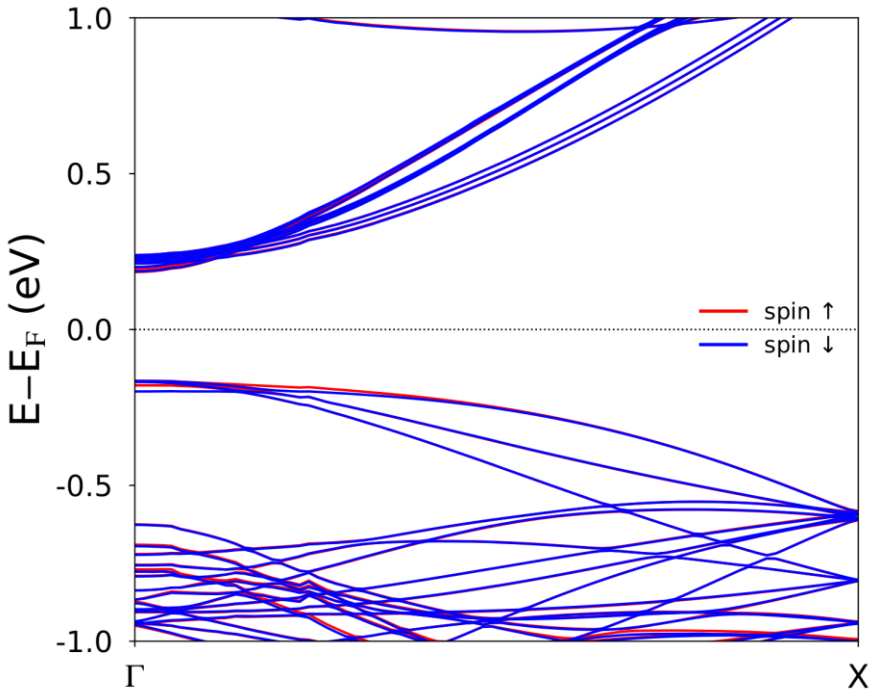


CrSBr(2L)-**MoS$_2$** HS

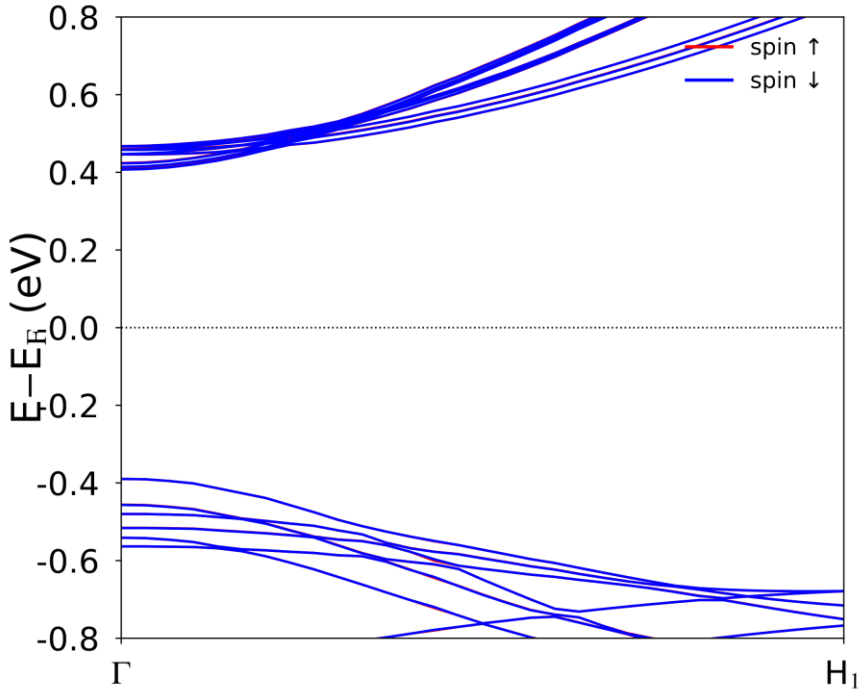


CrSBr(4L)-**MoS$_2$** HS

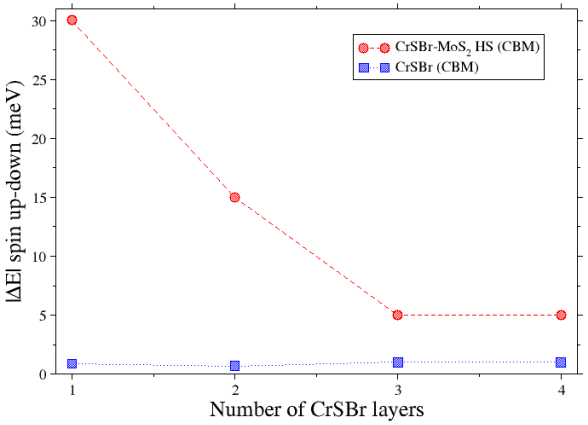
Spin-resolved band structure



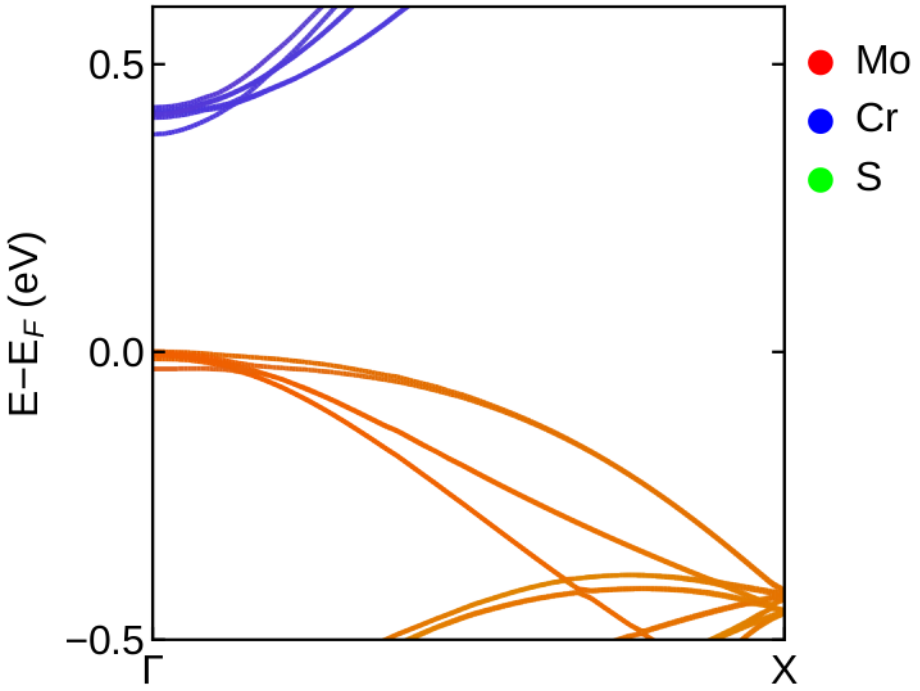
CrSBr(3L)-**MoS_2** HS



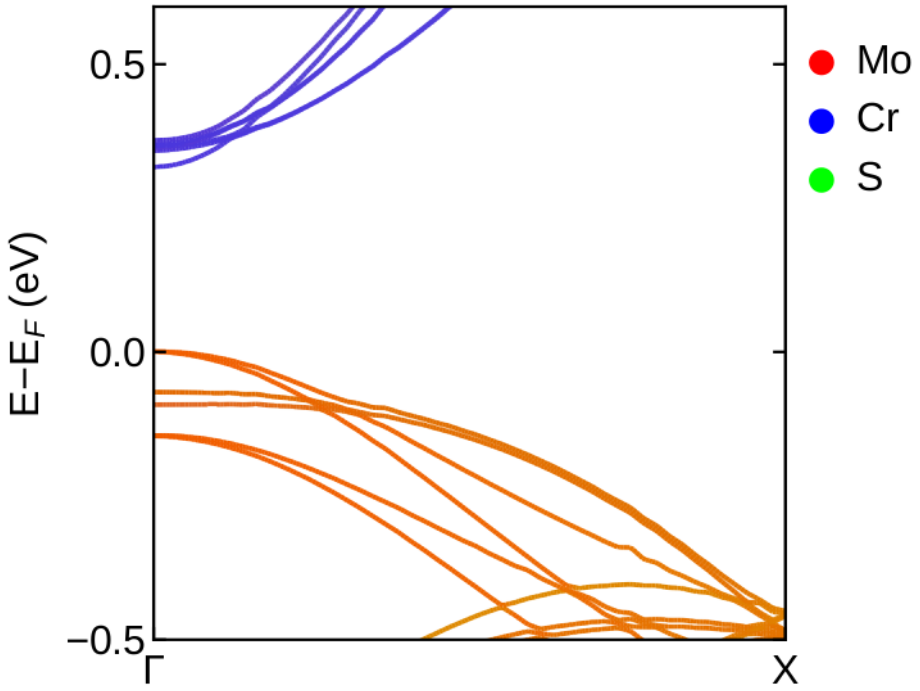
CrSBr(3L)



Spin-resolved band structure (SOC)

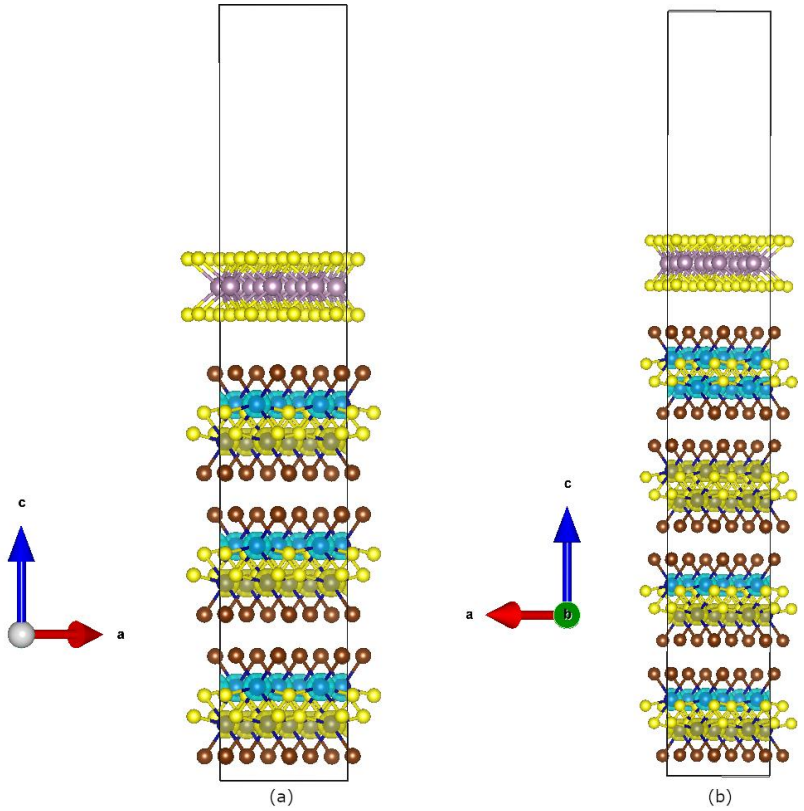


CrSBr(1L)-**MoS$_2$**

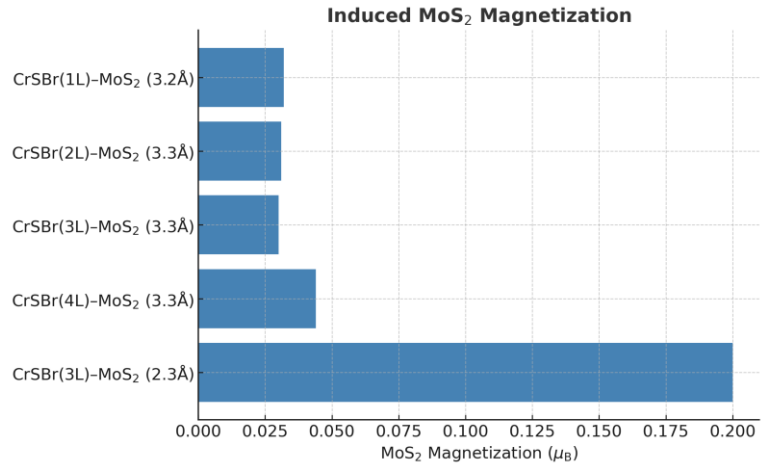


CrSBr(1L)-**MoS$_2$** (SOC)

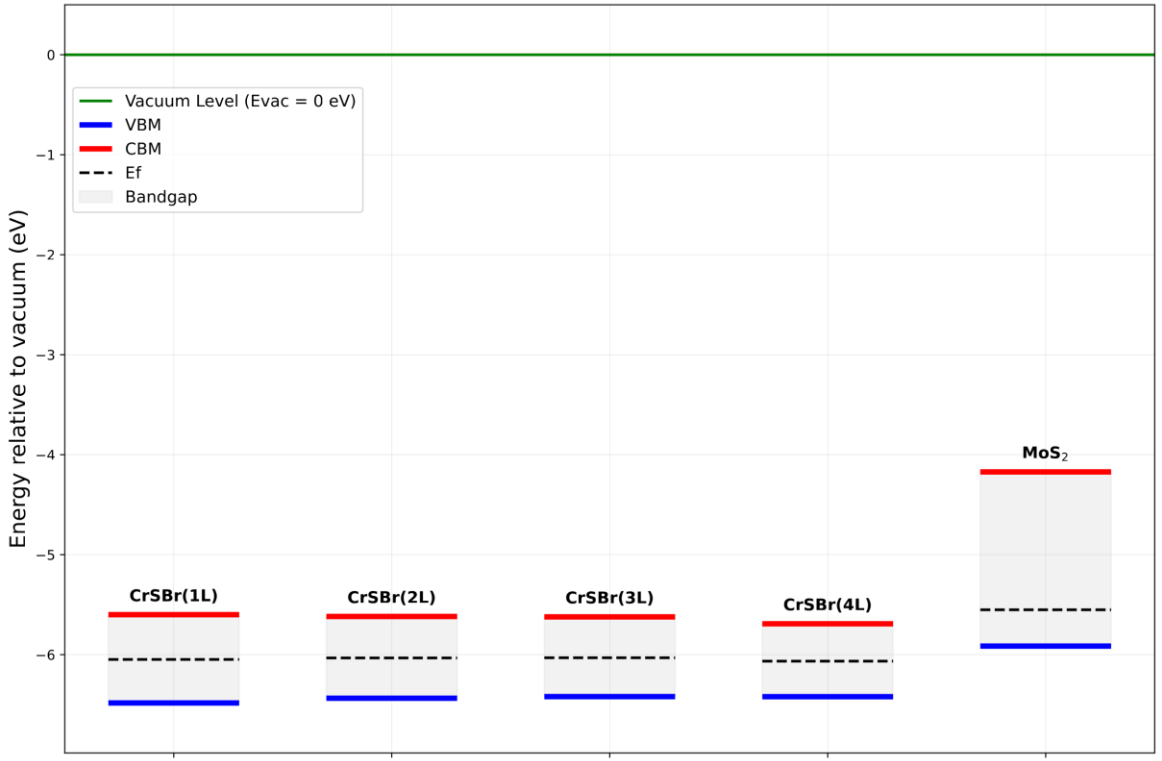
Induced MoS_2 magnetization



Structure	MoS_2 magnetization PBE (μ_B)
CrSBr(1L)-MoS_2 3.2 Å AFM	0.032
CrSBr(2L)-MoS_2 3.3 Å AFM	0.031
CrSBr(3L)-MoS_2 3.3 Å AFM	0.030
CrSBr(3L)-MoS_2 3.3 Å FM	-0.030
CrSBr(3L)-MoS_2 2.3 Å AFM	0.200
CrSBr(4L)-MoS_2 3.3 Å AFM	0.044



Work function and band alignment

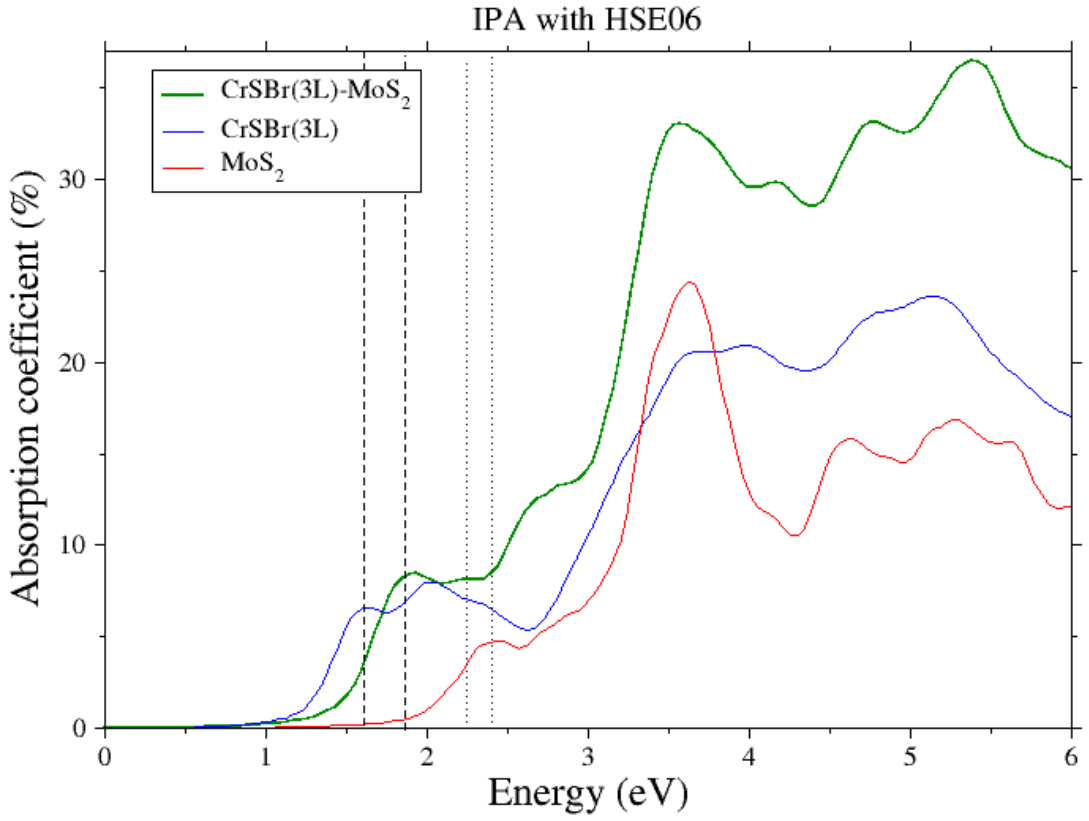


Structure	Φ (eV)	E_{vac} (eV)	E_F (eV)
CrSBr (1L)	6.048	2.672	−3.376
CrSBr (2L)	6.033	4.130	−1.903
CrSBr (3L)	6.031	5.061	−0.970
CrSBr (4L)	6.065	5.215	−0.850
MoS_2 (ML)	5.551	3.251	−2.300
CrSBr(1L)–MoS_2 (3.2 Å)	6.361	5.091	−0.652
CrSBr(2L)–MoS_2 (3.3 Å)	5.753	5.702	−0.051
CrSBr(3L)–MoS_2 (3.3 Å)	5.762	5.926	0.164
CrSBr(4L)–MoS_2 (3.3 Å)	5.787	5.933	0.146

$$\Phi = E_{vac} - E_F$$

- Band alignment Type-II (staggered)
- Indicates charge transfer from MoS_2 to CrSBr

Optical absorption



- Independent Particle Approximation: absorption coefficient from imaginary part of dielectric tensor
- Dashed lines: CrSBr part
- Dotted lines: MoS2 part
- Optical gap peaks in PL spectra: 1.1 & 1.9 eV
- Optical gaps in HS: blueshift for CrSBr, redshift for MoS2

Extensions

- MoS_2 bilayer/CrSBr multilayer
- Spin-orbit coupling
- GW+BSE for excitons

Acknowledgments

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Thank you for your kind attention!