

Tuning the Electronic and Chemical Properties of Monolayer MoS₂ Adsorbed on Transition Metal Substrates

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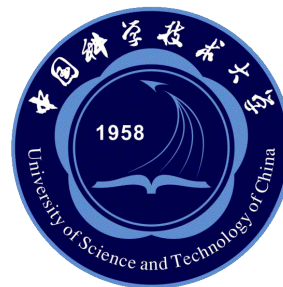
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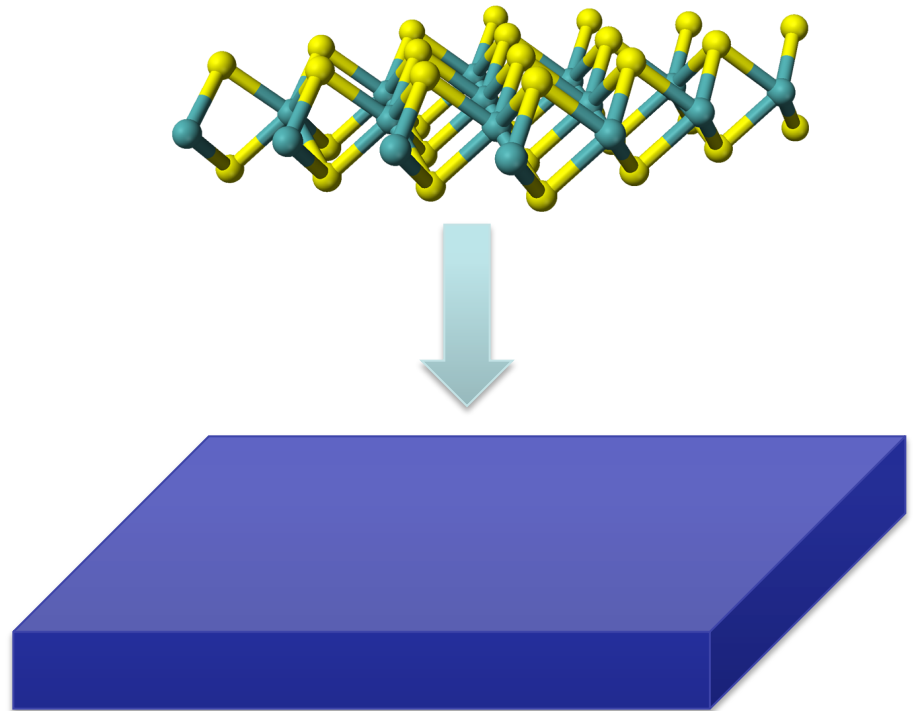
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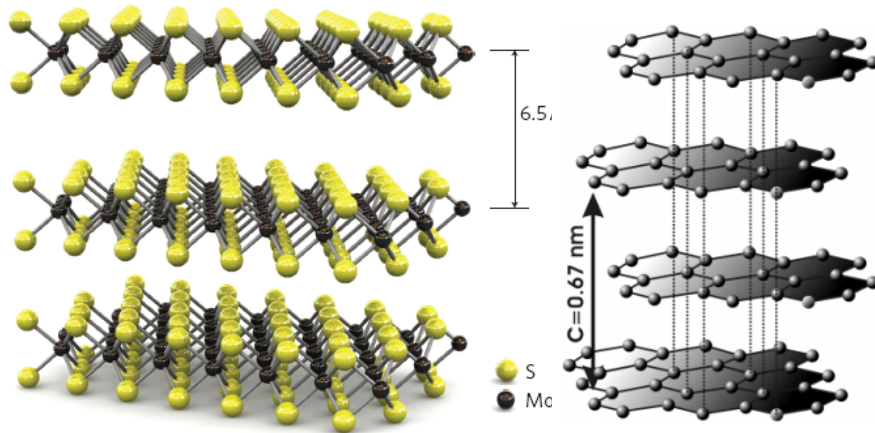
Outline

1. Background
2. Monolayer MoS_2 - Metal Interfaces
3. Electronic Properties
4. Chemical Properties
5. Conclusion

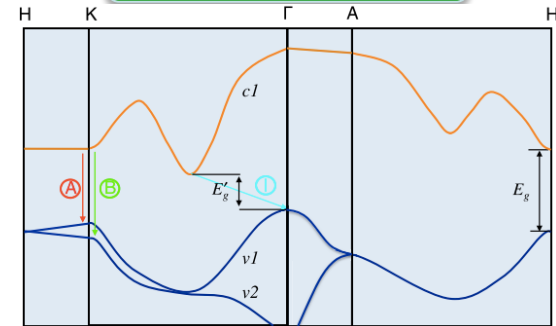


Background & Motivation

Geometric



Electronic

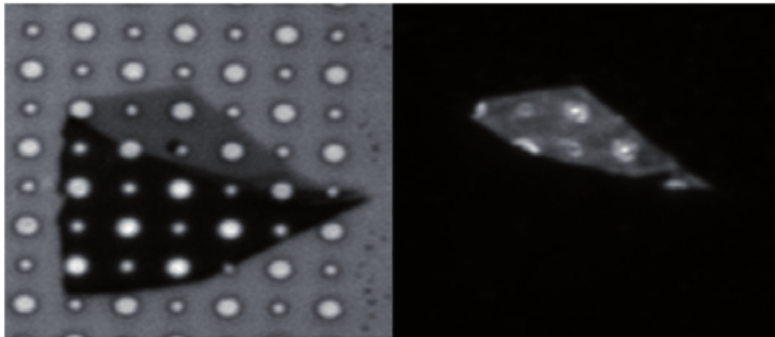


Bulk
1.29 eV indirect gap
~1.7 eV direct gap

Monolayer
~ 1.9 eV indirect gap
~ 1.8 eV direct gap

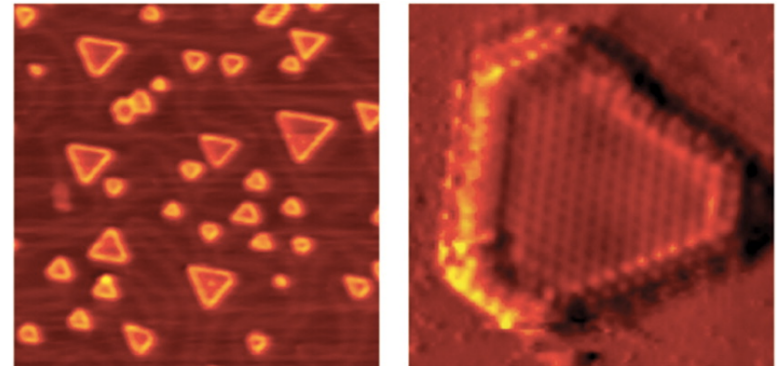
K. F. Mak *et al.*, *PRL* (2010)

Optical



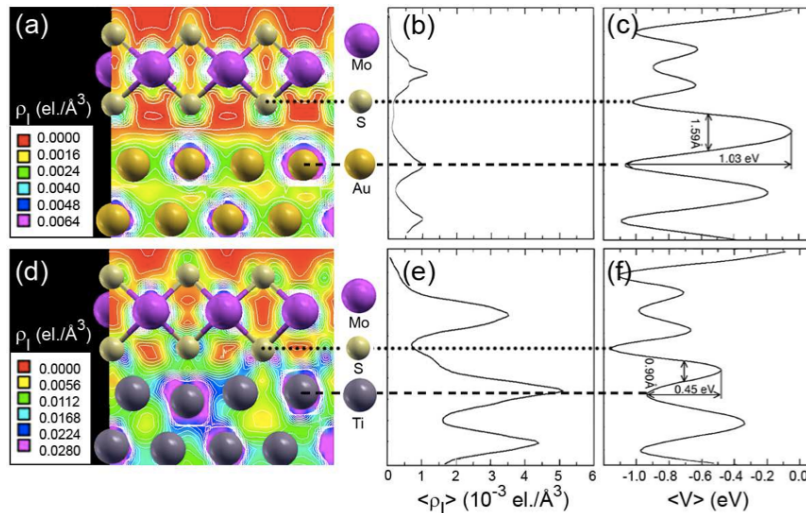
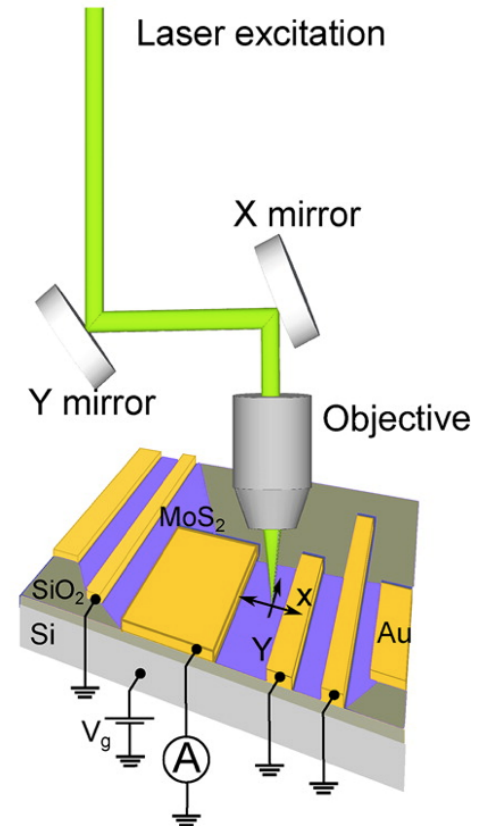
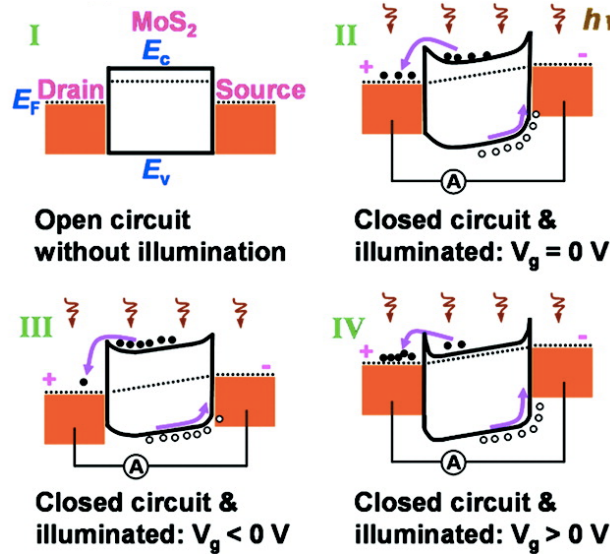
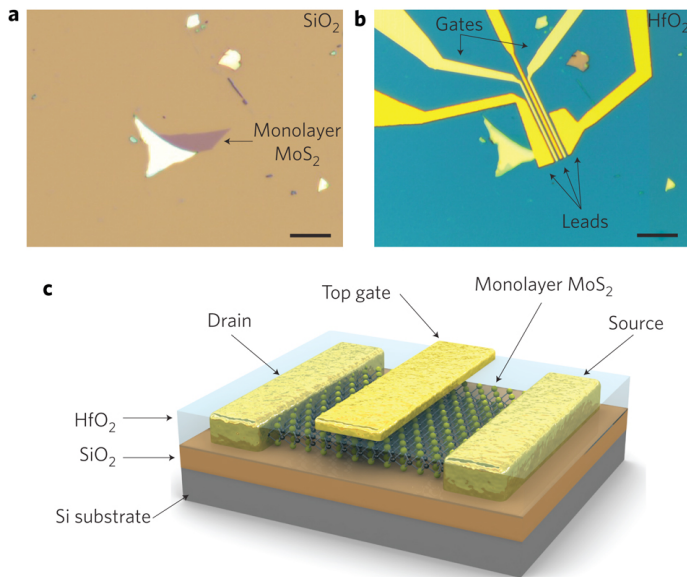
K. F. Mak *et al.*, *PRL* (2010)
H. Zeng *et al.*, *Nat. Nanotechnol.* (2012)
K. F. Mak *et al.*, *Nat. Nanotechnol.* (2012)

Catalytic



T. F. Jaramillo *et al.*, *Science* (2007)

Monolayer MoS₂-based Electronics



"Single-Layer MoS₂ Transistors", B. Radisavljevic *et al.*, *Nat. Nanotechnol.* (2011)

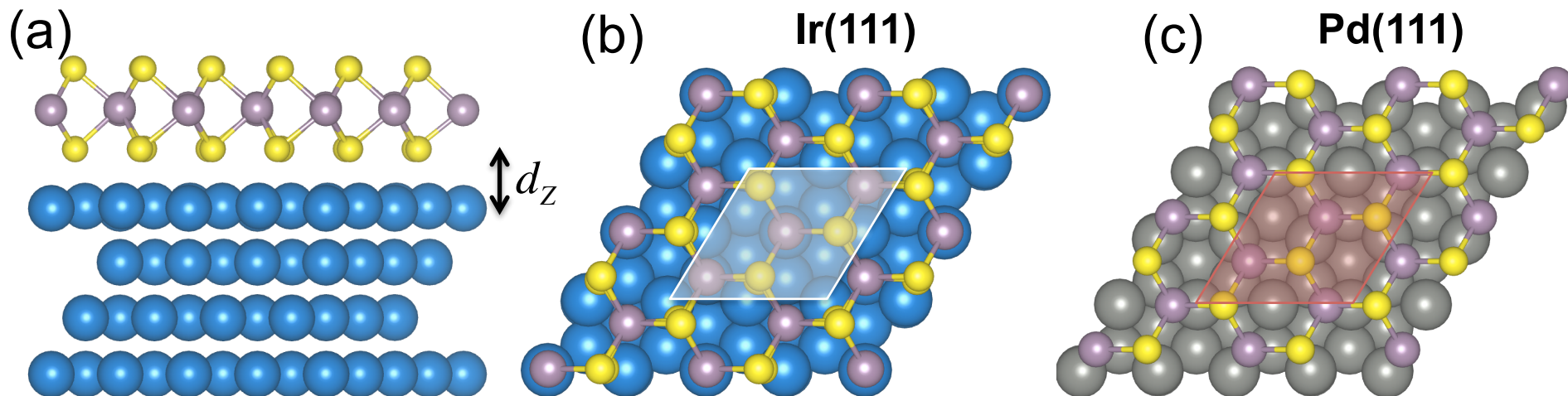
"Single-Layer MoS₂ Phototransistors", Z. Yin *et al.*, *ACS Nano* (2012)

"Large and Tunable Photothermoelectric Effect in Single-Layer MoS₂", M. Buscema *et al.*, *Nano Lett.* (2013)

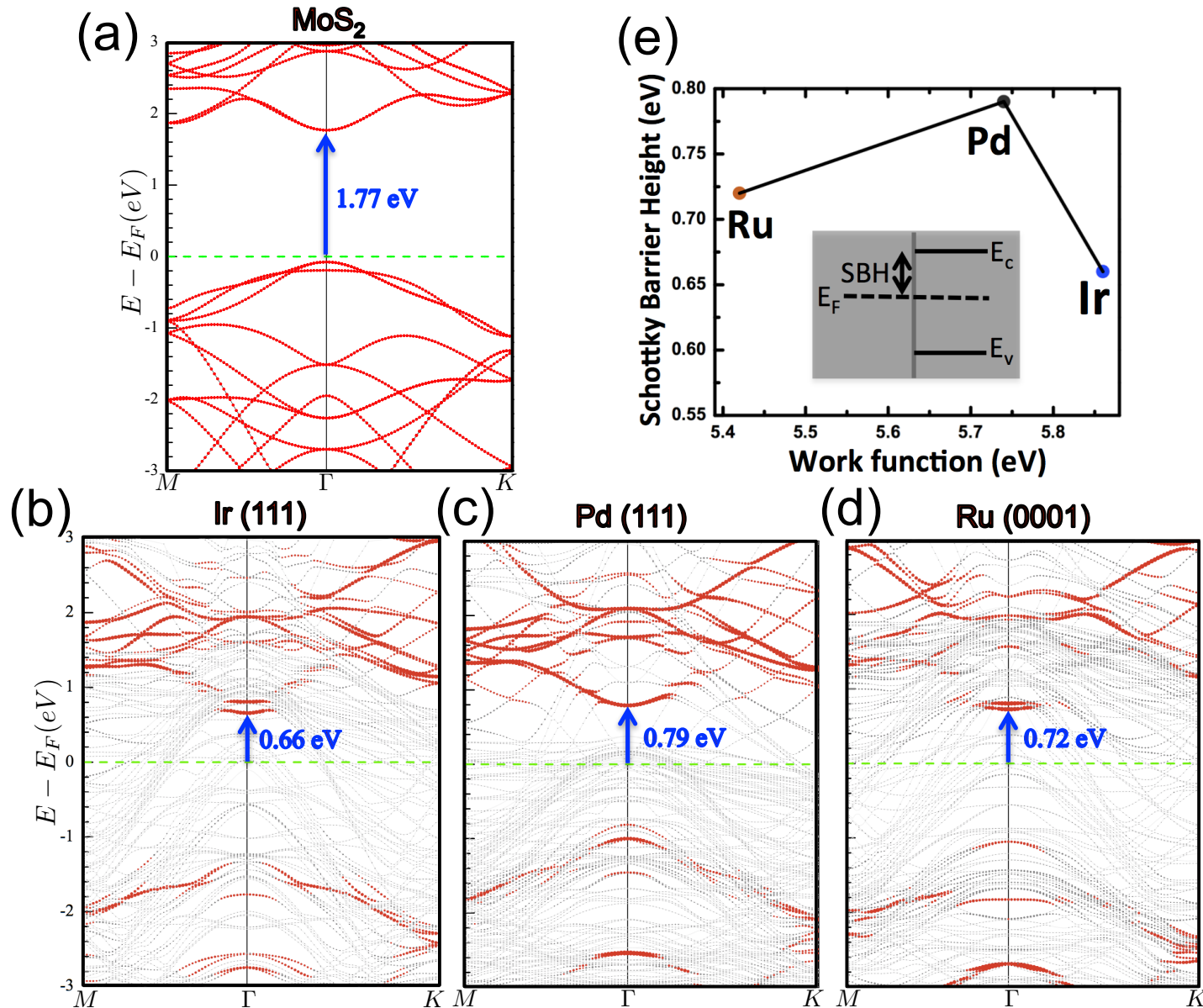
"Designing Electrical Contacts to MoS₂ Monolayers: A Computational Study", I. Popov *et al.*, *Phys. Rev. Lett.* (2012)

Monolayer MoS₂ – Metal Interfaces

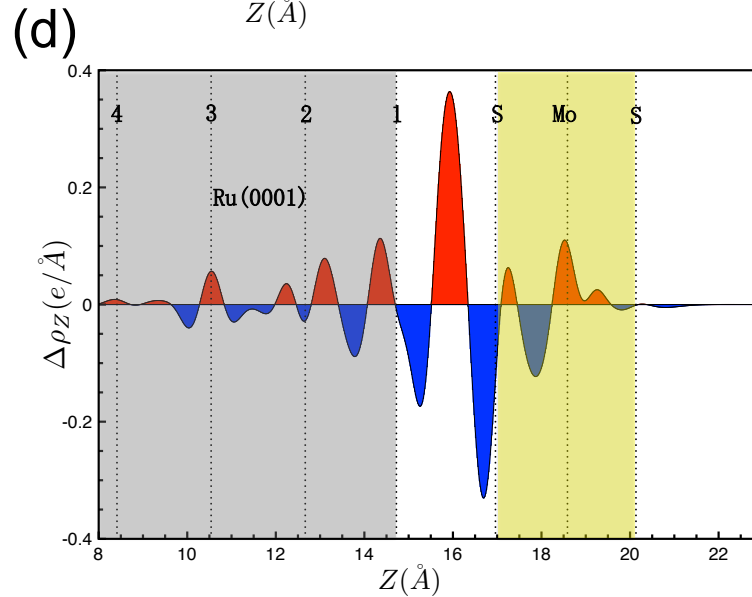
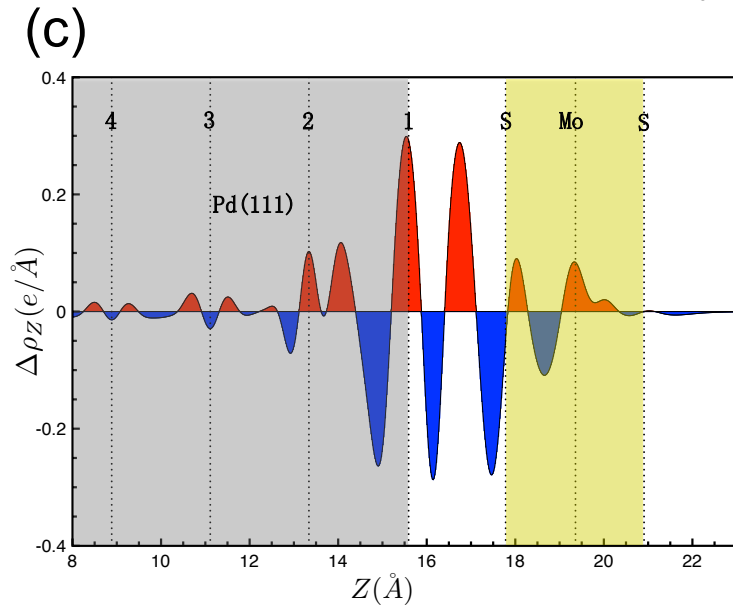
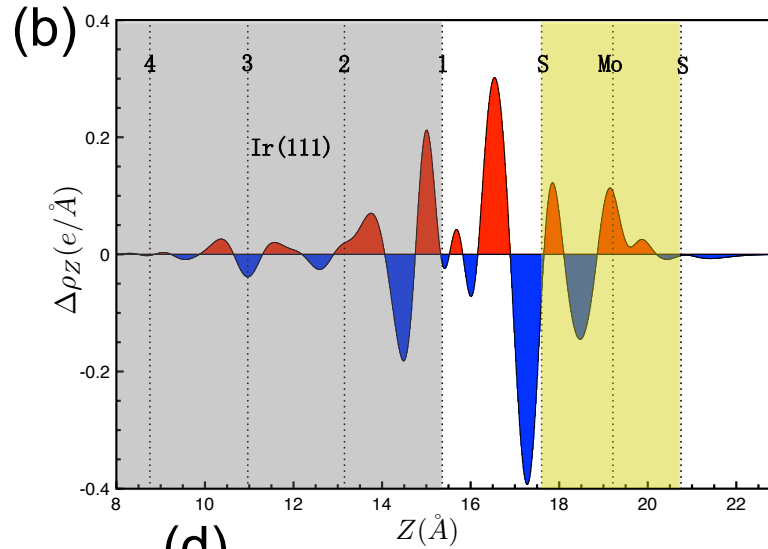
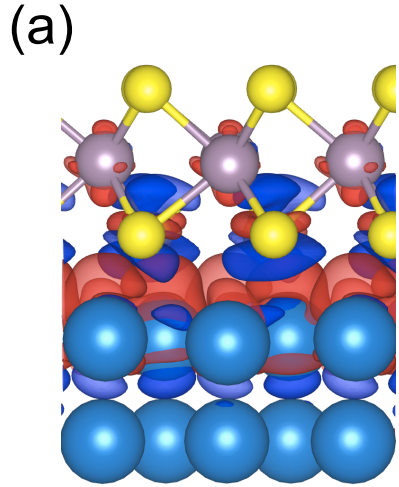
- DFT Package: VASP
- Functional: LDA, checked by PBE-vdW
- Supercell: $(\sqrt{3} \times \sqrt{3})$ MoS₂ + (2×2) Metal (mismatch < 1.2 %)
- Systems: MoS₂-Ir(111); MoS₂-Pd(111); MoS₂-Ru(0001)
- Spin-polarized calculations
- Spin-Orbital Coupling (SOC) effect checked



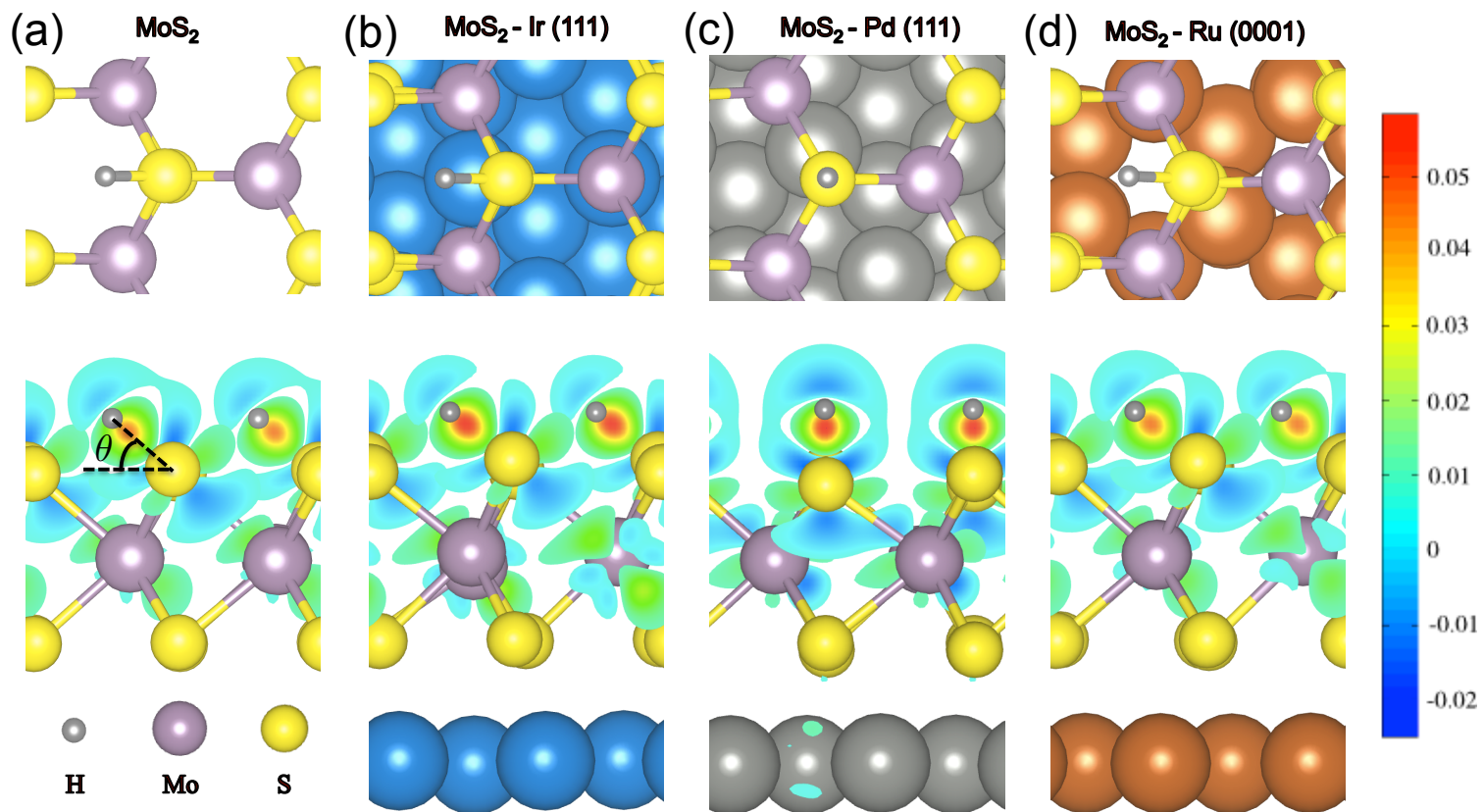
Band Structures



Charge Redistribution



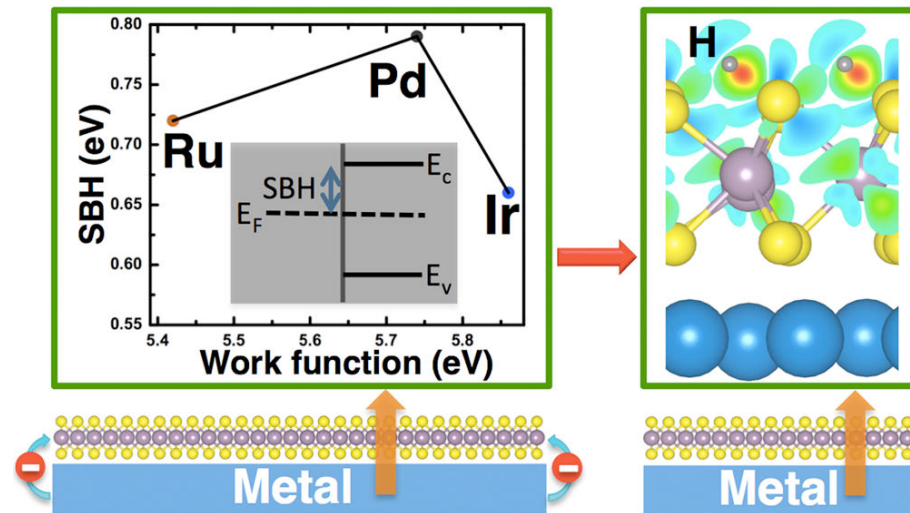
Hydrogen Adsorption



	E_b (eV)	d_z^0 (Å)	d_z^H (Å)	E_a (eV)	$L_{\text{H-S}}$ (Å)	θ (deg)
free-standing MoS_2				1.07	1.46	40.2
$\text{MoS}_2/\text{Ir}(111)$	0.62	2.23	2.20	1.44	1.43	37.2
$\text{MoS}_2/\text{Pd}(111)$	0.74	2.17	2.09	1.39	1.39	89.1
$\text{MoS}_2/\text{Ru}(0001)$	0.82	2.25	2.20	1.33	1.46	38.2

Conclusion

- The contact nature is Schottky-barrier type when monolayer MoS₂ adsorbed on Ir(111), Pd(111), or Ru(0001).
- The dependence of barrier height on the metal work function exhibits a partial Fermi-level pinning picture.
- Using hydrogen adsorption as a testing example, the chemical reactivity of MoS₂ can be substantially altered by the underlying metal substrates.
- The enhanced binding of hydrogen, is attributed in part to the enhanced H-S bonding, and in part to a stronger MoS₂-metal interface by H adsorption.



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Thank you!