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Engineering Hybrid Two-Dimensional TMDC/Perovskite Heterostructures from First Principles

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The integration of transition-metal dichalcogenide (TMDC) monolayers with thin perovskites nanosheets offers a versatile platform for engineering the electronic and excitonic properties of van der Waals heterostructures. First-principles simulations play a key role in understanding the microscopic mechanisms governing interfacial interactions and in guiding the design of new hybrid materials.
In this contribution, we present a series of density functional theory (DFT) studies of TMDC-based heterostructures performed in close collaboration with experimental groups. Structural optimization and electronic structure calculations reveal how the atomic arrangement, dielectric environment, and charge-compensating species determine the band alignment and interlayer coupling. For $WS_2/(PEA)_2PbI_4$, the calculations predict an unconventional band alignment where the organic spacer suppresses electron transfer while enabling efficient hole transfer through a valence-band cascade. Extending this approach to $MoSe_2$/2D-perovskite heterostructures demonstrates how the relative alignment of electronic and excitonic states controls the competition between charge transfer and energy transfer, providing design principles for tailoring excitation pathways. Simulations of $WSe_2/(BA)_2PbI_4$ further identify hybridized interfacial electronic states responsible for tunable spin-dependent interlayer exciton emission observed experimentally. Finally, recent first-principles investigations of the emerging $WSe_2/[Ca_2Nb_3O_{10}]^-$ heterostructure reveal weak interfacial coupling but a strong sensitivity of the electronic structure to the charge-compensating environment of the oxide nanosheet, highlighting the importance of defects and local chemistry in modifying excitonic properties.
Together, these studies demonstrate how atomistic simulations provide predictive insight into the structure–property relationships of hybrid TMDC heterostructures and establish computational guidelines for designing next-generation optoelectronic, spintronic, and quantum photonic materials.