

Spin-Orbit-Coupled Electronic and Vibrational Properties of ZnO/ZnS Quantum-Dot Heterostructures

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ZnO/ZnS quantum-dot heterostructures combine a polar oxide core with a wide-bandgap sulfide shell. Quantum confinement, local strain, and chemical bonding at the interface can change band-edge states and lattice vibrations. We connect Raman spectroscopy with first-principles calculations to study these coupled electronic, vibrational, dielectric, and spin-dependent effects.

We construct an isolated 2,870-atom $\text{Zn}_{1366}\text{O}_{310}\text{S}_{1194}$ model from a MatterSim-relaxed geometry. PBEsol/SIESTA calculations test the large heterostructure. A 42-atom $\text{Zn}_{14}\text{O}_{28}$ reference uses our published Quantum ESPRESSO workflow [1]: PBE+U, fully relativistic norm-conserving pseudopotentials, noncollinear spin-orbit coupling, and density-functional perturbation theory. These calculations provide band-edge orbital character, phonon modes, Raman tensors, Born effective charges, and dielectric response. Room-temperature 532 nm Raman spectra provide the experimental mode reference.

The ZnO reference has a calculated 4.54 eV HOMO-LUMO gap. Its valence edge is mainly O 2p, its conduction edge is mainly Zn 4s, and Zn 3d states lie several electronvolts below the Fermi level. Spin-orbit coupling slightly narrows the gap and lifts valence-edge degeneracies by about 0.1 eV. In the ZnO/ZnS structure, O occupies the inner region, S forms the outer shell, and Zn spans both regions. This geometry provides a direct basis for testing how interface strain and local bonding reshape band-edge and Raman-active states. The combined workflow links measured phonons to atomic structure, dielectric screening, and spin-dependent electronic behavior. These links support rational interface design for ultraviolet optoelectronics, sensing, and quantum technologies.

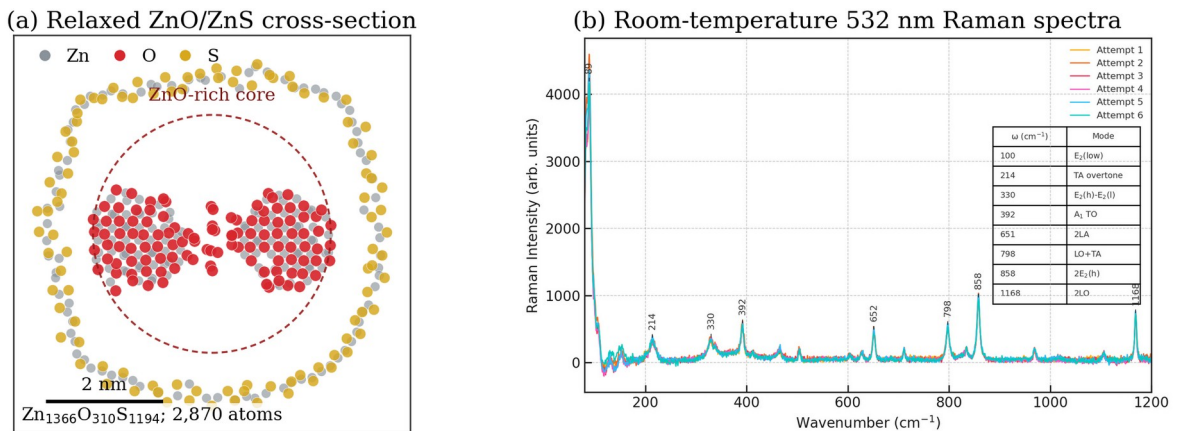


Figure 1. (a) Central cross-section of the relaxed 2,870-atom ZnO/ZnS model. Gray, red, and gold markers denote Zn, O, and S. (b) Room-temperature 532 nm Raman spectra of the ZnO quantum-dot reference. Labeled bands guide the experimental-computational mode comparison.

[1] W. Benson, C. Adams IV, B. Baral, and P. Misra, *AIP Adv.* **16**, 025236 (2026), doi: 10.1063/5.0303211.

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