Surface Geometry-driven Quantum Confinement in III-V Colloidal Quantum Dots

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Colloidal quantum dots (QDs) exhibit unique optoelectronic properties arising from quantum confinement (QC) within their nanoscale dimensions. The particle-in-a-box (PIB) model traditionally predicts tetrahedral QDs should display stronger QC than spherical ones. However, recent experiments on InAs QDs unexpectedly show that spherical QDs possess larger confinement energy than their tetrahedral counterparts, challenging the conventional PIB interpretation.[1, 2]

Here, we demonstrate that ligand-induced surface reconstruction significantly alters the QD electronic structure by using first-principles calculations. Specifically, ligand passivation triggers reconstruction on (100) surfaces while leaving (111) surfaces as is. Consequently, (111) surfaces show strong hybridization between in-gap surface states and bulk conduction band, effectively narrowing the band gap. Furthermore, we employed full three-dimensional QD models, to confirm the surface-originated QC effect in the synthesized QD. These insights highlight the importance of surface geometry and passivation for tailoring the optoelectronic properties of colloidal QDs.

References

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