A First-Principles Investigation of Dual-Ligand Passivation for Engineering Surface Defects in InAs Colloidal Quantum Dots

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InAs colloidal quantum dots (CQDs) have emerged as a promising Pb-free infrared (IR) absorber for photovoltaics, offering tunable optoelectronic properties. Despite this advantage, as-synthesized InAs CQDs exhibit strong *n*-type behavior due to intrinsic surface defects, which create trap states and limit photovoltage. Here, we conduct a density functional theory (DFT) analysis of multi-ligand passivation strategies to address these defects. Our calculations identified two major donor-type defects, each requiring distinct passivation. While 3-Mercaptopropionic acid (MPA, X-type) could suppress one donor, additional $ZnCl_2$ (Z-type) passivation is required for complete passivation. The distinct binding mechanisms of these ligands effectively mitigate multiple surface defects. Using an optimized architecture incorporating this passivation strategy, we achieved the first Pb-free IR photovoltaic devices that surpass silicon in IR harvesting.[1] This understanding of passivation mechanisms is expected to further expand the applicability of CQDs in optoelectronic devices.

References

1. Park, Y.; Kim, J.; Jeong, M.; Shin, D.; Jung, J.; Kim, H.; Jeong, H.; Kim, H.; Kim, Y.-H.; Jeong, S. *Advanced Energy Materials*, **2024**, 2404141.