Advancing Quantum Dot Simulations: from DFT insights to Machine Learning

Ivan Infante[1,2]

[1]BCMaterials, Bld. Martina Casiano, Barrio Sarriena s/n, 48940, Leioa, Spain
[2]Ikerbasque, Plaza Euskadi 5, 48009, Bilbao, Spain

Ivan.infante@bcmaterials.net

Colloidal quantum dots (QDs), which were awarded the 2023 Nobel Prize in Chemistry, exhibit complex electronic, optical, and structural properties, making them essential in optoelectronics, photovoltaics, and nanomedicine. Despite advances in understanding QD surface chemistry and trap state formation, key questions remain, particularly regarding surface effects on electronic properties. Addressing these challenges requires accurate theoretical modeling.[1, 2]

In our group, extensive density functional theory (DFT) studies have explored QDs up to ~4.5 nm, revealing that increasing system size leads to band gap collapse and facet-specific localization of frontier orbitals.[3] We also found that introducing surface vacancies induces reconstructions that widen the band gap and delocalize charge carriers, emphasizing the critical role of surface geometry in defining QD properties. However, DFT-based approaches are computationally expensive, limiting their application to larger, more realistic systems and longer timescales.

To overcome these limitations, we are preparing to employ machine learning force fields (MLFFs) trained on DFT datasets [4]. A key aspect of this approach is the inclusion of long-range electrostatic interactions during the training process to ensure the structural stability of QDs, which is crucial for accurately capturing their surface and bulk properties. These MLFFs aim to provide DFT-level accuracy at significantly reduced computational costs, enabling the study of larger QD systems and their dynamic behavior over extended timescales.

We anticipate applying ML-based models to various QD compositions, including CdSe, InP, PbSe, and $CsPbBr_3$, facilitating more efficient investigations into QD growth and optoelectronic integration. This transition highlights the transformative potential of machine learning in advancing the computational toolkit for nanomaterial design.

References

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Figures

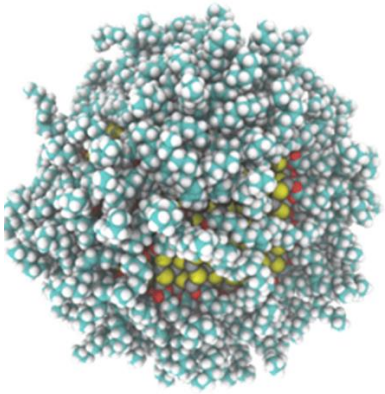

Figure 1. A typical QD model that resembles closely experimental observations.