Structures of CdSe and CdS nanocluster by ab initio random structure searching

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The properties of colloidal semiconductor quantum dots (QDs) continue to stimulate a lot of interest in a wide variety of field including materials, photonics, energy applications etc. Over the last few years, there has been a lot of interest in ultra-small QDs that do not follow the usual nucleation- and-growth model during the synthesis - so-called Magic-Sized Clusters (MSCs). These MSCs have an advantage of being mono-diapers systems with a number of unusual optical properties including temperature-dependent emission switching [1]. However, despite significant level of interest, the identifying atomic structure of these systems is still a challenge.

We have therefore turned to ab initio structure prediction as a useful tool to aid the analysis of experimental observations and develop an understanding of the structures of the dots. A systematic first principles structure prediction study of $(CdSe)_n$ and $(CdS)_n$ nanoclusters has not been attempted previously, instead guesses are often obtained starting from sections of the bulk crystal that are subsequently relaxed using density-functional theory (DFT). In our research, we have explored the possible space of stable structures of $(CdSe)_n$ and $(CdS)_n$ (n up to 34) more completely using ab initio random structure searching (AIRSS) [2, 3]. Using AIRSS we build libraries and benchmarks for the ground state structures in a more objective way and also provide some reference to predict the nanoclusters structures with AIRSS method. I will present demonstrate how AIRSS allows us to understand the transition from cage to bulk-like structures.

References

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