Towards the modelling of the optical properties of sensitized solar cells: a self-consistent periodic electrostatic embedding method

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Abstract: Over the last few decades, the need to overcome the limitations of the silicon based solar cell, first and foremost the high manufacturing costs, has pushed the research towards the development of sensitized solar cells (SSCs). Among them, Quantum Dot sensitized solar cells (QDSCs), characterized by the use of inorganic semiconductor nanoparticles (so called quantum dot or QD) as sensitizer, has attracted increasing attention over the last years due to its excellent opto-electronic properties such as tunability of band gap energy, narrow emission spectrum and multiple exciton generation, which come from the quantum confinement effect experienced by the nanoparticle because of its reduced and controlled size. The experimental characterization of QD optical properties is quite challenging and methods for modelling this kind of systems still remain particularly computationally-demanding. From a theoretical point of view therefore, multilayer QM/MM or QM/QM' approaches divide a molecular cluster in different layers to which different levels of theory are applied. Despite being widely used, these methods do not consider the effect of an infinite potential generated by the periodic system on the molecular cluster. In this context, a few years ago, Wilbraham et al. [1] developed a computational protocol, the SC-Ewald method, which seeks to reproduce the electrostatic effect produced by an infinite periodic environment using a finite array of point charges. A cluster of atoms, extracted from a periodic structure, is embedded in an inner shell of constant point charges and an outer shell of variable charges defined to reproduce the electrostatic potential of an infinite periodic system. The procedure has been generalized to deal with systems periodic in one, two and three dimensions.

Hereby, we present a couple of examples concerning the application of the SC-Ewald method to compute the optical properties of two basic components of QDSC:

1. CdSe zinc blende nanoplatelets, 2D nano-sized structures used in QDSC as light harvesting component [2];
2. benzoic acid derivatives adsorbed on a TiO_2 surface, recently considered for perovskite solar cell application as linkers to graft the perovskite to the TiO_2 substrate [3];

The SC-Ewald procedure allowed us to successfully apply Time Dependent DFT to such complex systems, in order to compute the UV-vis absorption spectra of extended periodic 2D objects at low computational cost, in good agreement with the available experimental data.

References

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