

Electronic Structure of Dye-Sensitized TiO₂ Clusters from G_0W_0

Noa Marom

*Center for Computational Materials, Institute for Computational Engineering and Sciences
(ICES), The University of Texas at Austin, Austin, TX 78712, USA*

The development of new types of solar cells is driven by the need for clean and sustainable energy. In this respect dye sensitized solar cells (DSCs) are considered as a promising route for departing from the traditional solid state cells. The physical insight provided by computational modeling may help develop improved DSCs. To this end it is important to obtain an accurate description of the electronic structure, including the fundamental gaps and level alignment at the dye-TiO₂ interface. This requires a treatment beyond ground-state density functional theory (DFT).

In this talk I will present a many-body perturbation theory study of TiO₂ clusters, dye molecules, and dye-sensitized TiO₂ clusters, within the G_0W_0 approximation. At this level of approximation the one-particle Green's function, G , and the dynamically screened Coulomb interaction, W , are calculated non-self-consistently based on DFT orbitals and eigenvalues. G_0W_0 calculations based on a hybrid functional are in excellent agreement with photoemission spectroscopy (PES) experiments. For TiO₂ clusters, this enables the identification of the isomers observed in PES as those with the highest vertical electron affinity [1]. For transition metal phthalocyanine dyes G_0W_0 calculations, unlike DFT, correctly predict the energies of orbitals associated with the metal d-states [2]. For dye-sensitized TiO₂ clusters, G_0W_0 calculations provide reliable predictions for the fundamental gaps and the level alignment at the dye-TiO₂ interface [3].

References:

[1] Phys. Rev. Lett. **108**, 106801 (2012)

[2] Phys. Rev. B **84**, 195143 (2011)

[3] Phys. Rev. B **84**, 245115 (2011)