Toward Computational Design of Cluster-Based Functional Nano-Structures

Noa Marom
Department of Physics and Engineering Physics, Tulane University, New Orleans, LA 70118

Computational materials design offers tremendous potential for discovery and innovation. This powerful concept relies on computational exploration of the vast configuration space of materials structure and composition to identify promising candidates with desired properties for target applications. In fact, many applications do not rely on a single material but on the combination of several materials in a functional nano-structure. Examples for functional nano-structures include the dye-oxide interface, at which charge separation is achieved in dye-sensitized solar cells, and nanocatalysts based on clusters dispersed on a large surface area support. Therefore, we would like to design not just a material, but a functional nano-structure. This requires the combination of accurate electronic structure methods with efficient optimization algorithms.

The electronic properties and the resulting functionality of a nano-structure cannot be deduced directly from those of its isolated constituents. Rather, they emerge from a complex interplay of quantum mechanical interactions that depend on the local environment at the nano-scale. Describing these effects requires a fully quantum mechanical first principles approach. In the first part of the talk, many-body perturbation theory within the *GW* approximation, where *G* is the one-particle Green's function and *W* is the screened Coulomb interaction, is used to elucidate the size effects in the energy level alignment at the interface between dye molecules and TiO_2 clusters of increasing size [1,2].

In the second part of the talk, a new approach is presented for computational design of clusters using property-based genetic algorithms (GAs). These algorithms perform optimization by simulating an evolutionary process, whereby child structures are created by combining fragments ("mating") of the fittest parent structures with respect to the target property. Property-based GAs tailored to search for low energy, high vertical electron affinity (VEA), and low vertical ionization potential (VIP) are applied to TiO_2 clusters with up to 20 stoichiometric units. Analysis of the resulting structures reveals the structural features associated with a high VEA and a low VIP and explains the absence of the expected size trends [3,4].

References:

[1] T. Körzdörfer and N. Marom, PRB 86, 041110(R) (2012)
[2] N. Marom, T. Körzdörfer, X. Ren, A. Tkatchenko, and J.R. Chelikowsky, J. Phys. Chem. Lett. 5 2395 (2014)
[3] N. Marom, M. Kim, and J. R. Chelikowsky, Phys. Rev. Lett. 108 106801 (2012)
[4] S. Bhattacharya, B. H. Sonin, C. J. Jumonville, L. M. Ghiringhelli, and N. Marom, PRB 91, 241115(R) (2015)