Local Excitations in Point Defects and Surface Binding using a Periodic Density Matrix Embedding Framework

Abhishek Mitra[a], Matthew Hermes[b], Hung Pham[c], Valay Agarwal[d] and Laura Gagliardi*[e]

[a] *Department of Chemistry, Chicago Center for Theoretical Chemistry, University of Chicago.*
[b] *Department of Chemistry, Chicago Center for Theoretical Chemistry, University of Chicago.*
[c] *Bytedance, Mountain View, CA.*
[d] *Department of Chemistry, Chicago Center for Theoretical Chemistry, University of Chicago.*
[e] *Department of Chemistry, Pritzker School of Molecular Engineering, James Franck Institute, Chicago Center for Theoretical Chemistry, University of Chicago.*

Accurate and affordable correlated wave function-based methods are often necessary for understanding the electronic structure of localized perturbations in crystalline materials, such as solid-state defects and adsorbate molecules on solid surfaces. These perturbations break translational symmetry, which can make the use of large supercells prohibitively expensive with traditional correlated wave function-based methods. Quantum embedding offers a solution by using a high-level method to treat the perturbations while using a mean field method for the rest of the system. We have recently developed strongly correlated electronic structure solvers, specifically, complete active space self-consistent field (CASSCF), n-electron valence state second-order perturbation theory (NEVPT2) and multiconfiguration pair-density functional theory (MC-PDFT) within the density matrix embedding theory (DMET) algorithm for studying electronic excitations in solid-state defects and binding energies of gas molecules on solid surfaces. Our results show good agreement with non-embedded approaches and demonstrate that DMET can be used to calculate excitation energies for large supercells that are otherwise intractable with conventional methods.