Quantum Embedding with pair-Coupled Cluster Doubles-Based Methods

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In the quantum embedding approach, the whole molecular structure is bifurcated into a system part studied by more accurate methods and the environment part modeled by low-level methods. Thus, we circumvent the problem of treating the whole structure with computationally expensive methods by focusing on the more chemically important region. Here, we propose a unique embedding scheme, augmenting the orbital optimized pair-coupled cluster doubles (OO-pCCD)-based methods as the wave-function-theory (WFT) component for the system part with density functional theory (DFT) approximations for the environment. The pCCD method shows reliable accuracy for strongly correlated systems, albeit with mean-field-like computational scaling. Specifically, in our WFT-in-DFT approach, we use a static embedding scheme, where the WFT part is represented by pCCD along with its dynamic correlation correction variants. We test our embedding methods for selected complexes featuring a UO_2^{2+} unit. The accuracy of the proposed models is assessed against the dipole moments and excitation energies.

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