Quantum-chemical methods for biochemical systems

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Linear- and sublinear-scaling methods ranging from DFT, MP2 to RPA allow - in particular in combination with graphics processing units (GPUs) - for the description of biochemical systems at QM and QM/MM levels. Here, large QM spheres with typically 500-1000 atoms are necessary for reliable studies [1]. Linear-scaling methods are briefly outlined with special focus on our RPA (random-phase approximation) [2, 3] and beyond RPA [4] reformulations for describing electron-correlation effects using atomic orbitals and Cholesky-decomposed densities. Furthermore, we introduce a new resolution-of-the-identity (RI) scheme for MP2 and RPA which avoids entirely the huge errors in absolute energies of standard RI approaches. In contrast to standard RI that typically relies on error cancellations for relative energies, our RI scheme for MP2 and RPA is almost error free, while the cost is only slightly increased [5]. Applications of our linear-scaling quantum-chemical methods range from understanding the discrimination within the nucleotide addition reaction in RNA polymerase II [6], over the dynamical behavior of PYP (photoactive yellow protein) [7], to a new base-independent DNA repair mechanisms [8]. Finally, a widely applicable approach for localizing free energy changes is introduced [9] that allows for novel insights into molecular transformations.

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