Relativistic equation of motion coupled cluster theory

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Résumé

Heavy elements (Z > 31) are widely used in technological applications (catalysts, strong magnets, nuclear fuels etc) and, as such are intimately linked to major societal issues (nuclear waste, pollution). This makes is a pressing concern to understand their behavior in complex environments, and nowadays simulations play a key role in such efforts.
These elements, however, are often not easy to simulate since, in order to understand their properties it is essential to incorporate relativistic effects (scalar relativistc effects and spin-orbit coupling) into the electronic structure calculations. This has to be done at the same footing as the treatment of electron correlation, as both effects are now understood to be non-additive [1].

In this contribution we discuss our current work in the development of the equation of motion coupled cluster approach based on 4-component Hamiltonians [2], including its combination with quantum embedding approaches based on the frozen density embedding framework [3], so that both ground and excited states of complex systems can be accurately treated-and predicted whenever experiments are too difficult to perform.

These approaches are showcased with examples of the determination of electron binding energies and electronically excited states of actinides in the gas phase [4], and of electron binding energies halogenated species in water droplets [5]. We will also outline our current efforts to treat larger molecular systems and other molecular properties in the DIRAC (http://diracprogram.org) program.

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