MRChem: Quantum Chemistry at the basis set limit with Multiwavelets

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The accuracy of Density Functional Theory calculations is governed by two factors: the **functional** employed and the **basis set**.
Despite the great popularity and success of some very well known functionals, the universal functional remains elusive. A vast number of functionals is currently available, sometimes tailor-made to target specific systems or properties. Assessing the accuracy of a modern functional, requires also a basis set which is capable of achieving an even greater precision – ideally approaching a complete basis – efficiently and systematically.
The two main families of basis sets (plane waves and Gaussian-type orbitals) have drawbacks which limit them, especially when high precision is required. A very attractive alternative is constituted by by grid-base methods[1] such as Multiwavelets[2, 3]. They combine conceptual simplicity (basis functions are standard polynomials) with the ability to reach complete basis set results within any given, predefined precision. Multiwavelets are fully orthonormal (like plane waves) and localized (like Gaussian-type orbitals).
With the development of MRChem,[4] – a multiwavelet code – we have been able to carry out extensive benchmarks of energy and properties, consistently achieving the basis set limit[5, 6].
In this presentation, we will summarize the theoretical multiwavelet framework, with emphasis on our implementation of the SCF optimizer and linear response solver, we will give an overview of some recent applications, and we will discuss the hurdles which shall be overcome to extend the Multiwavelet framework to large systems (thousands of electrons or more).

References

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