Design of the `molsturm` quantum-chemistry framework:

A step towards method development for arbitrary basis functions

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State-of-the-art program packages for modelling the electronic structure of molecules predominantly employ Gaussian-type basis functions for the discretisation. Gaussians are computationally cheap and usually reliable, but exhibit a few drawbacks, e.g. with respect to describing the physics near the nuclei or at large nucleus-electron distances. Thus investigating alternative basis function types and testing their applicability in such cases is desirable.

As will be discussed with focus on the self-consistent field (SCF) procedure, all it should take to add a new basis type into an existing quantum-chemical software package is to change certain integral routines [1]. In practice, however, both the size and the sparsity structure of the arising numerical linear algebra problems is affected as well, such that other approaches for solving these might be required. Especially in this respect, many existing quantum-chemistry software use hard-coded procedures tailored towards Gaussian-type functions, leading to assumptions about this basis scattered around the large codes. Consequently, incorporating other basis functions becomes challenging if not impossible.

This talk presents our solution to this issue, namely the flexible and light-weight quantum-chemical method development framework `molsturm` [1]. It will be explained how a concept we call lazy matrices [1, 2] allows us to completely separate `molsturm`'s SCF code from the basis-type dependent integral routines. As a result, it is possible to add new basis functions in a plug and play fashion, i.e., by just implementing a link to an existing integral code. Conversely an SCF scheme, for example, only needs to be programmed once and can subsequently be used with all basis functions available.

It is explicitly *not* the goal of `molsturm` to become yet another full-featured quantum-chemistry package. Instead `molsturm` has been designed such that readily available interfaces allow to integrate well with existing third-party codes [1, 2], e.g. Post-HF methods. This will be demonstrated by presenting examples of `molsturm`'s usage in our recent investigation of Coulomb-Sturmian-type basis sets [2]. `molsturm` is well-suited for building simple stand-alone programs for prototyping and teaching, but could be used for making plug-in modules hosted in other quantum chemistry software as well.

Related works

[1] M. F. Herbst, A. Dreuw and J. E. Avery. J. Chem. Phys., **149**, 84106 (2018).

[2] M. F. Herbst. Ph.D. thesis, Ruprecht-Karls-Universität Heidelberg (2018).