

VeloxChem: an efficient implementation of real and complex response functions at the level of Kohn–Sham density functional theory

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With the ease of Python library modules, VeloxChem offers a front end to quantum chemical calculations on contemporary high-performance computing (HPC) systems and aims at harnessing the future compute power within the EuroHPC initiative. At the heart of this software lies a module for the evaluation of electron-repulsion integrals (ERIs) using the Obara-Saika recurrence scheme, where a high degree of efficiency is achieved by employing architecture-independent vectorization via OpenMP SIMD pragmas in the auto-generated C++ source code. The software is topology aware and with a Python-controlled work and task flow, the idle time is minimised using an MPI/OpenMP partitioning of resources.

In the second software layer, we have implemented a highly accurate SCF start guess based on atomic densities and a first-level of iterations in a reduced version of the user-defined basis set, leading to a very smooth convergence in the subsequent standard DIIS scheme. This layer also includes vectorised and OpenMP/MPI parallelised modules for efficient generation of DFT grid points and weights as well as kernel integration.

In the third software layer, we present real and complex response functions as to address dispersive and absorptive molecular properties in spectroscopy. The kernel module in this layer is the iterative linear response equation solver that we have formulated and implemented for a combination of multiple optical frequencies and multiple perturbation operators. With efficient use of computer memory, we enable the simultaneous reference to, and solving of, in the order of 1,000 response equations for sizeable biochemical systems without spatial symmetry, and we can thereby determine electronic response spectra in arbitrary wavelength regions, including UV/vis and X-Ray, without resolving the sometimes embedded excited states in the spectrum. E.g. the electronic CD spectrum (involving the Cartesian sets of electric and magnetic perturbations) over a range of some 10 eV is obtained at a computational cost comparable to that of determining the transition energy of the lowest excited state, or optimising the electronic structure of the reference state.

Multi-frequency/gradient linear response equation solver

