Autonomous Active Space Calculations through AutoCAS

M. Mörchen[1], M. Reiher[1]*

[1]Department of Chemistry and Applied Biosciences

In order to describe strongly correlated systems correctly, the choice of an active space imposes one of the greatest problems in multi-configurational quantum chemistry.
In the AutoCAS algorithm, concepts from quantum information theory are exploited in order to automatically and consistently select orbitals for an active space.
We present our Python-based AutoCAS [1, 2, 3, 4] module, which can be employed in existing workflows to streamline multi-configurational calculations in a black-box manner.
Due to the black-box-like selection of active spaces, post-active space methods like Tailored Coupled Cluster [5, 6, 7] or second order perturbation theory [8] can be routinely applied to recover dynamic correlation.
Furthermore, in the AutoRXN workflow [9], a workflow for the exploration of chemical reaction networks, we automatically validated CCSD(T) energies with potential multi-reference character through the AutoCAS module.

[1] Christopher J. Stein, Markus Reiher, *Journal of Computational Chemistry*, **2019**, 40, 2216-2226.
[2] Christopher J. Stein, Markus Reiher, *Journal of Chemical Theory and Computation*, **2016**, 12, 1760-1771.
[3] Christopher J. Stein, Vera von Burg, Markus Reiher, *Journal of Chemical Theory and Computation*, **2016**, 12, 3764-3773.
[4] Christopher J. Stein, Markus Reiher, *Chimia*, **2017**, 71, 170-176.
[5] Tomoko Kinoshita, Osamu Hino, Rodney J. Bartlett, *Journal of Chemical Physics*, **2005**, 123, 074106.
[6] Libor Veis, Andrej Antalík, Jiří Brabec, Frank Neese, Örs Legeza, Jiří Pittner, *Journal of Physical Chemistry Letters*, **2016**, 7, 4072–4078.
[7] Maximilian Mörchen, Leon Freitag, Markus Reiher, *Journal of Chemical Physics*, **2020**, 153, 244113.
[8] Leon Freitag, Stefan Knecht, Celestino Angeli, Markus Reiher, *Journal of Computational Chemistry*, **2017**, 13, 451-459.
[9] Jan P. Unsleber, Hongbin Liu, Leopold Talirz, Thomas Weymuth, Maximilian Mörchen, Adam Grofe, Dave Wecker, Christopher J. Stein, Ajay Panyala, Bo Peng, Karol Kowalski, Matthias Troyer, Markus Reiher, *Journal of Chemical Physics*, **2023**, 158, 084803.