Quantum Chemical Exploration of Catalytic Reaction Networks

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A variety of different approaches have been proposed for the automated exploration of complex reaction networks of a variety of chemical processes in the last decade.[1] In such an endeavor, we have followed a specific path which combines massively automated exploration[2] based on first-principles heuristics with interactive steering[3] through real-time quantum mechanics[4]. As results of individual calculations can no longer be carefully inspected owing to the huge amount of data produced, we improved on standard algorithms in order to make orbital optimizations more robust[5] and fast[6] and to push the stability of transition-state optimization[7]. We equipped all steps with error estimation procedures[8] that together with seamless uncertainty quantification[9] allow for kinetic modelling and mechanism deduction from noisy reaction networks[10]. A specific design feature of our approach has been the drive to provide algorithms that are agnostic w.r.t. the type of molecules considered and that can be extended when needed. In my talk, I will describe this integrated approach that led to the SCINE project (www.scine.ethz.ch).

References

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