1. PHD PROJECT DESCRIPTION (4000 characters max., including the aims and work plan)

Project title: A theoretical perspective on the structure-property relationship in organic electronics

1.1. Project goals

- Devising a geometry optimizer for pCCD-based methods including their eclectic extensions
- Implementing ground and excited state molecular properties for post-pCCD methods and their eclectic extensions
- Extending the derived models to open-shell electronic structures
- Prediction of structure-property relations in large-scale organic electronics

1.2. Outline

Experimental studies on the advancement of organic electronics heavily rely on a trial-and-error approach. This disadvantage entails large workloads and depletes a lot of time and consumables. A more efficient approach is to exploit quantum chemistry to guide the synthesis of new materials and accelerate the development of novel compounds. The computational models, however, are difficult primarily because conventional highly-accurate approaches are technically limited to small model compounds and demand user control on an expert level. In large-scale modeling of new materials, DFT is hence considered the method of choice. Yet, DFT may predict unreliable electronic structures and molecular properties as heavy-element-containing organic electronics may feature a substantial amount of strong correlation. In this project, we will extend our novel hybrid wave-function-based approaches to scrutinize the structure-property relationship in heavy-element-containing organic electronics, like dye-sensitized solar cells or phosphorescent organic light-emitting diodes. Specifically, we will develop a geometry optimizer for various pCCD-based methods and implement molecular properties, like optical gaps, dipole strengths, etc. The proposed models will be implemented in PyBEST and will (i) enhance our understanding of the relation between a molecular structure and its properties and how changes in the molecular structure influence molecular properties, (ii) shift the computational paradigm towards wave-function-based quantum chemistry calculations (beyond DFT), and (iii) accelerate the discovery of new materials.

1.3. Work plan

The proposed Ph.D. project includes the following work tasks:

- Geometry optimizer for ground-state pCCD (with and without orbital optimization)
- Extension to ground-state post-pCCD models
- Extension to open-shell pCCD-based methods
- Generalization to electronically excited state models for the improved pCCD-based methods
- Implementation of molecular properties for all available pCCD-based approaches

- Application of the derived and implemented methods to large-scale organic electronics

1.4. **Literature** (max. 10 listed, as a suggestion for a PhD candidate)

- *Molecular Electronic-Structure Theory, T. Helgaker, P. J*ørgensen, and J. Olsen, John Wiley & Sons, New York, 2000.
- Many-Body Methods in Chemistry and Physics, MBPT and Coupled-Cluster Theory, I. Shavitt and R. J. Bartlett, Cambridge University Press, Cambridge, 2009.
- A. Leszczyk, P. Tecmer, and K. Boguslawski (2019) New Strategies in Modeling Electronic Structures and Properties with Applications to Actinides. In: Broclawik E., Borowski T., Radoń M. (eds) Transition Metals in Coordination Environments, vol 29. Springer, Cham.

1.5. Required initial knowledge and skills of the PhD candidate

- Basic knowledge of molecular electronic structure theory like Hartree-Fock theory, multiconfigurational self-consistent field theory, coupled cluster theory, etc.
- The candidate should be familiar with Linux, bash, and basic terminal commands
- The candidate should have basic programming skills, preferably in Python
- The candidate should be fluent in spoken and written English (the project language will be English)

1.6. Expected development of the PhD candidate's knowledge and skills

- The candidate will gain expertise in unconventional electronic structure methods, like the density matrix renormalization group algorithm, geminal-based wave function ansätze, and various coupled cluster flavors
- The candidate will gain fundamental knowledge in state-of-the-art many-body-theory for ground and excited states for both closed- and open-shell systems
- The candidate will be actively involved in software development of our own open-source software package written in Python and C++ (using the version-control system git and GitLab).
- The candidate will be well-trained in electronic structure calculations (using wave function-based methods) at various scales