Embedding methods in quantum chemistry - pushing the boundaries of modeling molecular properties of complex systems with heavy elements.
Abstract for the general public

Modern world, now more then ever, brings new threatening challenges in the areas of health, environment and well-being of societies, rendering scientific developments indispensable to inspire and drive change towards innovative solutions involving new functional materials, targeted drug design and clean industries. At the heart of these developments is basic research, aimed at understanding properties and behaviors of complex molecular systems under various conditions. Of special interest in this context are molecular systems involving heavy elements – from the bottom of the Periodic Table – due to their use as energy sources, nuclear agents in medicine, catalysts and sensors for toxic moieties in living cells and in the atmosphere, to name but a few.

Efficient design of new molecules and materials starts with their computational modeling, aimed first at a deep understanding of the relation between structure, function and properties of molecular assemblies in realistic environments and under various conditions, for instance in solution, on interfaces, embedded in complex structures, and additionally affected by the presence of external perturbations. This knowledge is best gained from calculations employing ab-initio quantum chemistry methods, which are based only on the physical laws. Particularly helpful in this context are calculations of molecular properties which can be measured in various spectroscopic experiments, and which connect the information on how a molecular system responds to various perturbations with its electronic structure. Unfortunately, the applicability of accurate quantum chemistry methods for this purpose is limited to small molecules, due to unfavorable scaling of such methods with the size of the system. The development of cost-effective methods is one of the main challenges in computational chemistry. Additional challenges arising due to the presence of heavy elements in these systems - such as the importance of the so-called relativistic effects and effects of electron correlation - further increase the complexity of the problem. A promising way to address all these challenges is to employ embedding methods, relying on the partitioning of the molecular system into smaller subsystems, which can then be tackled separately by carefully chosen quantum chemistry models. In addition to the possibility of performing multi-level calculations at a significantly lower cost, embedding methods have many more advantages. For instance they allow to address multi-scale problems, involving phenomena happening at various time and length scales, and offer a deeper insight into mechanisms governing molecular phenomena, which could otherwise remain hidden due to the complexity of the problem. The latter supports frequent strategy in chemistry which is to gain understanding from studying atoms and fragments in molecules. All these advantages contribute to a growing popularity of quantum embedding techniques and to an increasing number of successful applications.

This project proposes method developments in the field of quantum embedding techniques and combines these developments with the studies of molecular response properties in the relativistic framework. An essential part of this project is also the adaptation of analysis tools from applied mathematics for a better understanding of the strengths and deficiencies of developed embedding schemes and for gaining a deeper insight into the nature of complex systems and phenomena. An example of such tool is the Topological Data Analysis (TDA), which will be employed to extract, quantify and compare topological features of unperturbed and perturbed densities calculated for different systems and with various embedding models.

While the project centers around method development in quantum chemistry, it also involves a thorough testing on well-understood molecular systems and high-throughput benchmark calculations of the electronic structure and molecular properties of systems with heavy elements exhibiting various types of noncovalent interactions with their environment. It embodies a new paradigm in basic research in the era of data - combining the design of accurate models, advanced analysis, large high-throughput calculations and applications to meaningful problems.