Intermolecular interactions and homogeneous catalysis

Noncovalent interactions and chemical bond theory

In our group we combine state-of-the-art quantum mechanical methods for the calculation of accurate energies and electron densities with tools that allow for their decomposition into additive chemical meaningful components, with the final aim of contributing to a unified understanding of intermolecular interactions. Our interest ranges from small model systems of importance for gaining understanding of the basic principles of the interaction to large and complex molecules with practical interest in catalysis and biology.

An important aspect concerns the quantification of the elusive London dispersion component of the interaction and the identification of the so-called "Dispersion Energy Donors". In this context, our strategy involves the calculation of accurate DLPNO-CCSD(T) energies at the CBS limit and their decomposition by means of the recently introduced Local Energy Decomposition analysis. This scheme exploits the locality of the internal and external space in the DLPNO-CCSD(T) framework for quantifying, among other things, the London dispersion component of the interaction. As an example, we recently applied this approach to the interaction of Lewis acids and bases in both classical Lewis adducts and Frustrated Lewis Pairs (FLPs). It was found that, by counteracting the destabilizing energy contribution associated with the deformation of the monomers, London dispersion drives the stability of many Lewis adducts.
Complex chemical reactions in homogenous catalysis

We are developing protocols for the calculation of accurate geometries, electronic energies, entropies and solvation effects in complex chemical reactions that are challenging for current mainstream computational strategies. To do that, we combined advanced electronic structure calculations with explicit and implicit solvation models.

Our research interests include organo- and organometallic catalysis, with emphasis on reactions for C-C bond formation and C-H bond activation. We combine a detailed mechanistic understanding with an in-depth analysis of the underlying interactions, with the final aim of aiding in the development of designing principles for catalysts with well-defined bonding features and reactivity.

Validation of DLPNO-based schemes

An important activity in the group is devoted to the validation of DLPNO methodologies for applications in homogenous and heterogeneous catalysis. As an example, we recently developed a protocol for the accurate treatment of sub-valence correlation effects in the DLPNO-CCSD(T) framework. This method permits the study of sub-valence correlation effects in large and complex systems for which canonical CCSD(T) is not feasible.