Challenges and opportunities for quantum chemistry in understanding catalytic processes

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Quantum chemistry (QC) allows studying properties and transformations of chemical species at the most basic, molecular level. The knowledge gained enables knowledge-based alternations that aim to improve, e.g., selectivity or yield of the system under study. However, the size and complexity of molecules involved in various catalytic reactions limit the choice of QC methods that can be applied to a specific problem. Over the years, heterogeneous and homogeneous catalysis developed their own strategies to study systems of interest, mainly based on the density functional theory (DFT) in periodic or aperiodic formulations, respectively. Unfortunately, the lack of exact functional of the density and therefore no systematic way to improve results makes the common approach unpredictable in some instances. Moreover, emerging types of catalysis often involve interfaces (e.g., electrocatalysis) and require special care. One cannot approach them only from the side of homogeneous or only from heterogeneous perspective. In the CoopCat group at the Institute of Physical Chemistry PAS, we use and develop high-level quantum chemical methods and computation protocols to provide quantitative data of controllable accuracy. Our key concern is the transferability of the methods between various catalytic systems that ensure consistent accuracy irrespective of the reagents' phase.

The seminar will cover two key factors that one needs to consider to achieve high accuracy: (i) level of quantum chemical approximation and (ii) completeness and adequacy of the molecular model. The two choices are not independent as the size of the structure under study limits methods that can be applied. Our strategy typically involves combining embedding techniques with wave function methods that explore the locality of electron correlation. In this way, we target reactivity and spectroscopy at periodic and aperiodic surfaces such as titanium dioxide [1] or graphene oxide [2]. Other applications covered in the presentation include the chemistry and properties of biologically-relevant iron-sulfur clusters [3] and our contributions to understanding cis/trans isomerization of retinoids under various catalytic conditions [4].

References

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