Research Statement

13 February 2026

My research applies the tools of computational chemistry to explore phenomena such as gas-phase reactions, reactions in solution and at solid surfaces. My interests have evolved from first-principles based kinetic studies of gas-phase reactions and kinetic modelling to chemistry of surfaces and interfaces.

Summary of completed research in the last 10 years

In the last 10 years, I applied methods of density functional theory (DFT), *ab initio* molecular dynamics (AIMD) simulations, and microkinetic modelling to study complex mechanisms of heterogeneous catalytic reactions on various transition-metal catalysts, including Pt group metals and coinage metals, most notably, on Au-Ag alloys. My research efforts were largely focused on nanoporous gold (np-Au), a novel catalyst proposed for environmentally friendly applications but also an interesting material from the standpoint of fundamental research on gold. Since the discovery of the catalytic activity of Au on the nanoscale, as opposed to very inert bulk Au, there has been an ongoing debate in the scientific community, whether gold on its own (without any support material) can be catalytically active.

My research contributed to this scientific debate and related questions about the interplay between the topology, composition, and catalytic properties of alloyed nanostructures. I was able to show that the small amount of Ag impurities in the material can facilitate adsorption and dissociation of O_2. This finding allowed me and my experimental collaborators (the group of M. Bäumer at Universität Bremen) to argue that Ag impurities are responsible for the unexpectedly high catalytic activity of np-Au [1-3]. My co-authors and I published a computational study [3] where we proposed a new mechanism of CO oxidation on np-Au via a direct bimolecular reaction between CO and O_2 on the catalyst's surface. In the proposed mechanism the energetically unfavourable O_2 dissociation step is replaced by OCOO formation and its subsequent dissociation to CO_2 and surface atomic O. In another recent study on methanol oxidation [4], my co-authors and I, in a similar manner, suggested an alternative mechanism for methanol reaction with O_2, avoiding O_2 dissociation step but forming an OOH species instead. The O-O bond in surface OOH can dissociate with a considerably lower barrier than in O_2.

In a search for a lower activation barrier for O_2 dissociation on some special surface sites, my co-authors and I conducted further studies on the surface composition of Au-Ag alloy surfaces, concentrating on the role of surface and subsurface atomic oxygen, oxygen induced Ag segregation, and formation of –O–Au–O– phases [2,5-8]. The latter topic is quite interesting on its own because it again illustrates the special properties of gold. We found that –O–Au–O– chains show more covalent bonding as compared to analogous –O–Ag–O– chains. Despite the higher affinity of Ag to oxygen, in bimetallic Au-Ag systems –O–Au–O– chains are more thermodynamically favourable than –O–Ag–O– chains. An experimental study by the group of Friend [9], for the first time visualised such chains on the pure Au(110)-(1x2) surface by means of STM. Very recently, we have computationally demonstrated that formation of –O–Au–O– along the steps of gold surfaces can influence the selectivity of alcohol partial oxidation on Au-based catalysts [8]. My contribution to the broader understanding of np-Au catalysis is reflected in the recent comprehensive Chem. Rev. 2023 article (IF 55.8) [10], where I co-authored the section on computational studies, summarizing key developments in the field including my own contributions.

Some of my research related to np-Au can also be extended to other types of Au-based catalysts, such as catalysis on Au nanoparticles. In this regard, I mention my work on propylene epoxidation [11], where I have given an explanation why H_2 or water dramatically improve the selectivity of propene epoxidation, on the basis of extensive DFT calculations of various possible reaction pathways. My work shows that surface HOO species (formed via H_2 or H_2O reaction with O_2) is a more selective oxidising agent than atomic O and it thus allows for

the formation of the desired propylene oxide rather than undesired products, e.g. acetone, acetaldehyde, and CO_2.

Recently, I also worked on oxide or metal-oxide systems, which are more difficult to model theoretically due to limitation of current DFT approaches in describing strongly correlated systems [12, 13]. The method of choice offering a trade-off between accuracy and computational cost is the DFT+U method including empirical on-site Hubbard correction. I mention our recent publication, in which we successfully simulated the complete catalytic cycle of CO oxidation on a two-component inverse ceria-gold model catalyst. The low-energy pathway was discovered with the help of *ab initio* MD simulations.

Another line of research that I was pursuing recently is related to battery research and electrocatalysis. My group collaborated with three experimental groups in China working on the design of improved electrocatalysts and novel electrode materials for energy storage. I would like to mention our collaborative work on the improvement of the MoB electrocatalyst for hydrogen evolution reaction (HER) [14]. Molybdenum-based electrocatalysts are currently emerging as a promising class of noble-metal free catalysts for HER. Our work proposed and demonstrated that introducing a layer of n-type semiconductive g-C_3N_4 with a wide band gap of 2.7 eV on the surface of a MoB electrocatalyst leads to a Shottky junction, which enhances the charge separation and transport and leads to a superior HER activity. Our theoretical DFT-based simulations demonstrated that introducing n-type semiconductive g-C_3N_4 on MoB catalyst leads to enriched surface electron density of MoB, such that the proton access to active sites is facilitated, and the kinetic barrier for H^+ reduction is lowered, significantly boosting the surface hydrogen generation. This development opens a new avenue for activating metallic electrocatalysts. Our article has already been cited >400 times since 2018. In a collaborative work with the group of Prof. Mai [15], we helped to demonstrate that silicene structural subunit in $MoSi_2$ serves for boosting HER activity in alkaline media. Our DFT-based calculations indicate that the formation of Si vacancies on silicone subunit edges weakens the covalent character of Mo–Si bonds and enhances the metallicity and catalytic activity of Mo. In another collaborative work with the group of Prof. Wei Shi [16] we used ab initio molecular dynamics simulations to reveal the mechanism of increased Li storage capacity of novel anode materials. These materials are based on one-dimensional polymeric coordination compound of Ni with suitable organic ligands containing aromatic rings and capable of noncovalent weak hydrogen bonding or $\pi\cdots\pi$ stacking, so that both metal centres and the ligands are simultaneously contributing to enhanced Li storage. With Prof. Z.-J. Zhao's group (Tianjin University), we published a series of studies 1720] on Cu-based electrocatalysts for CO_2 electroreduction, elucidating mechanistic pathways and scaling relations.

In summary, I use theoretical tools to provide a mechanistic understanding of chemical reactivity in the gas phase, in solution, and on the interface between gas or liquid with solid surfaces. Many of these mechanistic insights are difficult, if not impossible, to access experimentally. My research thus assists experimentalists in the design of improved catalysts for a "green" and sustainable future.

Directions for future Research

I plan to direct my future research to computational studies of industrially important oxidation reactions, such as CO oxidation, preferential CO oxidation, water-gas shift reaction with two groups of heterogeneous catalysts: gold-based alloys and perovskite oxides. Research on perovskites will also include applications to electrocatalysis. I am particularly interested in exploring gold-based alloys, rare earth oxides, mixed metal oxides, and high-entropy alloys. Research on mixed metal oxides will also include applications to electrocatalysis. These materials hold great potential for understanding the relationship between atomic composition and nanoscale structure, as well as serving as active and stable alternatives to costly Pt-group catalysts in various industrial processes.

My interest in gold and functionalised gold goes back to my long-term collaboration with the research groups of Marcus Bäumer at Universität Bremen and Thomas Risse at Freie Universität Berlin and also to my participation in the DFG research unit NAGOCAT where I continued to participate even after my move to South Africa.

I would also be happy to contribute to modelling of homogenous catalytic reactions. While my research focus to date was on heterocatalytic systems, I have necessary experience due to my past work on modelling structure and reactivity of bare and solvated inorganic complexes [21- 24], including a very sophisticated study of spectral transitions [25]. I have recently applied computational tools to describe the structure, aurophilic bonding and photophysical properties of luminescent dinuclear Au complexes with phosphine ligands [26].

Overall, my ongoing and planned projects illustrate the continuity of my expertise in gold-based catalysis, the expansion into rare-earth oxide and molecular catalyst systems, and the exploration of new frontiers such as AI-driven catalyst discovery. These activities will not only advance fundamental understanding but also strengthen my international collaborations and contribute to training the next generation of computational chemists in South Africa.

In the long term, I aim to position my research at the forefront of computational catalysis by deepening the integration of atomistic modeling, data-driven methods, and experimental collaboration. This strategy will not only enable breakthroughs in understanding catalytic mechanisms but also accelerate the design of novel materials with societal relevance, particularly in energy conversion and environmental catalysis. A central objective will remain the training and mentorship of postgraduate students and postdoctoral fellows, equipping the next generation of South African scientists with cutting-edge skills to contribute to the global research community.

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