First-principles study of the electronic and optical properties of Sn-halide perovskite solution precursors

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Metal halide perovskites have shown great promise for next-generation opto-electronic applications but the predominant employment of Pb poses a problem of environmental sustainability. Replacing Pb with Sn represents a viable solution, however, our knowledge on Sn-based perovskites and its precursors is insufficient to date despite recent efforts in this direction [1]. In a first-principles work based on time-dependent density-functional theory coupled to the polarizable continuum model, we systematically investigate electronic and optical properties of SnI_n^{2-n} (n=1-4) complexes, considered the minimal units for Sn-halide perovskite precursors. We find that the electronic structure is hardly affected by the halogen species, which only cause an overall upshift of the energy eigenvalues upon increasing size. A similar trend is reflected also in the calculated optical absorption spectra, which we analyze in terms of allowed electronic transitions based on quantum-mechanical selection rules. The identified trends are consistent with those found for Pb-halide perovskite solution precursors [2], thereby motivating future work on tin-halide complexes explicitly bound to solvent molecules.

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A Lewis Acidic Catalyst Aluminum Chlorofluoride in C-F Bond Activation and Its Industrial Application

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As science and industry developed in the past decades, fluorinated compounds play an essential role in organic synthesis, agriculture and pharmacy[1]. Therefore, C-F bond activation becomes an inevitable challenge to achieve further derivatives[2]. Among various avenues of C-F bond cleavage, high Lewis acidic nanoscopic aluminium chlorofluoride (ACF, $AlCl_xF_{3-x}$, x ≈ 0.05-0.3), as an efficient catalyst, makes it easy to obtain different kinds of products[3].

Different fluorinated organic compounds can be catalyzed by ACF in the presence of various main group compounds, such as $HSnBu_3$, $ClSiEt_3$ and so on. Even though fluorinated organic compounds are difficult to be activate, with the help of ACF the reactions can be operated in mild reaction conditions with relatively high yield. With fruitful reaction results, the mechanism behind is also worth to research on. MAS NMR studies suggest the formation of silylium-like species at the surface of ACF, which facilities C-F bond cleavage. In addition, TGA studies also support an initial silane coordination at the ACF surface. In this research, not only the catalytical methodology is easier than traditional C-F bond activation conditions, but the application of the reactions can also be widely used in industry, such as drug intermediates synthesis and greenhouse gas removal.

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Ambient Forcing: Probabilistic Stability of Power Grids with constraint Phase Spaces

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Ambient Forcing is a novel method to sample random local perturbations of nodes in power systems with loads. These initial states enable the calculation of probabilistic stability measures of power systems with loads, which was not yet possible, but is important as these measures have become a crucial tool in studying power systems. We find that loads fundamentally influence the dynamical stability of power systems.

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Charge distribution and electronic structure of ZIF-8
and its derivatives from first principles

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Metal organic frameworks are novel materials with high potential in many fields of applications, ranging from gas storage and catalysis to optoelectronics. In spite of their growing popularity, there is still a lack of fundamental understanding of the physico-chemical properties of these materials, even in the most established subgroups such as the zeolitic imidazolate frameworks (ZIFs). To shed light into the nature of the chemical bonds in these systems, we performed a first-principles study based on density-functional theory investigating the charge distribution in ZIF-8 and its derivatives with Br, Cl, and H terminations replacing CH3. Our results indicate the presence of an ionic bond connecting the Zn to the organic scaffold, which is held together by covalent bonds. Control calculations performed on the building blocks of ZIF-8 indicate that the aforementioned characteristics are quantitatively influenced by the extended nature of the Framework. The presence of halogen substituents, carrying around an excess of negative charge, leads only to a negligible redistribution of the charge density. On the other hand, the electronic structures computed for these systems indicate a reduction of about 0.5 eV of the band-gap in the Br and Cl-terminated systems with respect to ZIF-8.

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Constellation: An Open-Source Web Application for Unsupervised Systematic Trend Detection in High-Resolution Mass Spectrometry Data

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The rise in the popularity and relative affordability of HRMS machines has led to a concurrent rise in custom software solutions to process, interpret and reveal the many layers of information contained in high-resolution mass spectra. Although there are numerous software packages for peak-picking, calibration and formula-finding routines, there are additional layers of information in the mass defect space, particularly when searching for polymers, molecules with repeating structures, and products of chemical or biochemical transformations that exhibit systematic, serial chemical changes of mass. Constellation is an open-source, Python-based web application that attempts to fill this gap by expanding HRMS data into the mass defect space, after which a trend finding algorithm can be used for supervised or unsupervised detection of repeating motifs. Adjustable trend finding parameters allow the user to tailor their trend-search to target particular chemical moieties or repeating units, or search for all potential motifs within certain limits. Visualization tools built on the open-source Dash/Plotly platform allow the user to interrogate their data and any trends and patterns in the mass defect space to a highly specific degree and save publication-quality images directly from the interface. Thorough these new developments, we are excited to add to a growing body of open-source software specialized in extracting useful information from complex datasets without the high costs, technical knowledge, and processor-demand typically associated with such tools.

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Deciphering microbiological influenced corrosion processes on steel with single cell-ICP-ToF-MS

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Microbiologically influenced corrosion (MIC) is a highly unpredictable process dictated by the environment, microorganisms, and the respective electron source. Interaction pathways between cells and the metal surface remain unclear.[1] The development of this novel single cell-inductively coupled plasma-time of flight-mass spectrometry analytical method and a MIC-specific staining procedure facilitate the investigation of steel-MIC interactions. With this it is possible to analyze the multi-elemental fingerprint of individual cells. The detection method revealed elemental selectivity for the corrosive methanogenic archaeal strain Methanobacterium-affiliated IM1. The interface between material and environmental analysis thus receives special attention, e.g., when considering MIC on solid steel. Hence, the possible uptake of individual elements from different steel samples is investigated. Results showed the cells responded at a single-cell level to the different types of supplemented elements and displayed the abilities to interact with chromium, vanadium, titanium, cobalt, and molybdenum from solid metal surfaces.[2] The information obtained will be used in the future to elucidate underlying mechanisms and develop possible material protection concepts, thus combining modern methods of analytical sciences with materials research.

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Regioselective Oxidative carbon-oxygen bond cleavage by lytic polysaccharide monoxygenase (LPMO)-inspired copper complex

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One of the most important industrial challenges for a low fossil fuel economy future is the conversion of biomass into fuels and other useful chemicals. First-generation biofuels are well established, but their dependence on food source suitable for human consumption means that they should be substituted. Because of its low cost, abundance and renewability, the degradation of lignocellulosic biomass (LB) to produce fuels and other useful chemicals is a very promising perspective. One of the major obstacles of LB valorisation is its surprisingly high recalcitrance to enzymatic hydrolysis caused by the heterogeneous multi-scale structure of plant cell walls. The discovery of lytic polysaccharide monooxygenases (LPMOs) has revolutionized our view on how cellulose is degraded [1]. In contrast to typical cellulases, which are hydrolytic enzymes, LPMOs cleave the β-1,4-glycosidic bonds via the oxidation of the C1- or C4 atom making the substrate tractable to hydrolases, apparently without the need for de-crystallization. In doing so, LPMOs boost the activity of canonical glycoside hydrolases by up to two orders of magnitude, thereby greatly reducing the financial and environmental penalties associated with the use of recalcitrant polysaccharides as a feedstock. This placed LPMOs at the centre of biochemical/bioinorganic research. However, our knowledge on the functioning of these enzymes is rather limited, the identity of the reactive species, the key steps in the oxidative mechanism and even the identity of the biologically relevant oxidizing co-substrate (O_2 or H_2O_2), are still unknown. To this end, the goal of this project is to develop an heterogenous catalyst inspired by the catalytic site of the lytic polysacchride monooxygenase (LPMO). The catalytic site of this enzyme is composed of a copper (II) ion bound by two histidine and a solvent molecule. The first histidine is bound by its imidazole side chain and the second one is bound in a clamp like coordination, by the N-terminal amine and an imidazole side chain. In this type of catalytic core called ''Histidine Brace'', the copper possesses a distorted geometry between square planar and tetrahedral which favors the copper redox cycling (Cu(I)/Cu(II)). This system was successfully synthesized and investigated using several spectroscopic tools. Our kinetic results indicate that the copper complexes in the presence of hydrogen peroxide as oxidising co-substrate, possess significant LPMO-like activity both at pH 7.4 and 10.5. Their enzyme-like behaviour was confirmed, as the kinetic data could be described by the Michaelis-Menten kinetics.

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Influence of the Anti-Solvent Drop on the Film Formation Processes in Mixed-Halide Perovskites by *in-situ* multimodal Spectroscopy and X-Ray scattering

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Metal-halide perovskites evolved over the last decade due to their outstanding optical properties as a photoactive material in various optoelectronic devices. All devices require high-quality thin films, mainly morphological but also regarding ionic homogeneity.

While mixed materials of the $MAPb(Br_xI_{1-x})_3$ series can be synthesized, recent studies have highlighted microscopic ionic heterogeneity, originating from the synthesis of the sample. Intrinsic heterogeneous formation processes via competing pathways, rationalize ionic heterogeneities within the $MAPb(Br_xI_{1-x})_3$ series prepared without an anti-solvent drop. Depositing an anti-solvent is an established method to prepare high-quality perovskite films.

We rationalize the formation mechanism by utilizing the three anti-solvents types for the $MAPb(Br_xI_{1-x})_3$ series via structural and optical in-situ characterization. We answer the questions: Does the anti-solvent facilitate more homogeneous samples ionically by "quenching" or worsen the inhomogeneity by inducing the system to crystallize in different phases? The answer strongly depends on how the anti-solvent acts, either as a medium forcing the whole system into oversaturation, including the formation of solvate phases, or by removing the original solvent, possibly preventing the formation of solvate phases. Rationalizing the underlying mechanisms of the anti-solvent drop helps to understand and control their effect on homogeneity and morphology of final thin films.

Catalysis and Quantum Chemistry

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We present an overview of how atomic scale simulations are used to understand processes and structures in heterogeneous catalysis. Our goal is to understand fundamental concepts that can guide us to more selective, lower energy processes involving abundant, non-toxic elements. Additionally, this poster was produced from an interdisciplinary collaboration between scientists and designers in order to better introduce our topic and facilitate a broader discussion about the communication of complex ideas. Come and take a look!

Luminescent Polymer Microbeads – Synthesis & Applications

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Luminescent polymer microbeads, either with organic dyes or luminescent nanoparticles included in the polymer matrix, are used for various (bio)analytical and diagnostic applications.[1] The preparation of these spherical microparticles is important for the realization of optically distinguishable barcodes that can be read out, e.g., by a flow cytometer or fluorescence microscope.

Fluorescent nanoparticles such as semiconductor quantum dots or nanoplatelets absorb light in a broad wavelength range and show narrow emission bands, which enables simultaneous excitation of differently colored particles and facilitates a spectral discrimination in emission compared to organic dyes. Furthermore, nanocrystals are typically more photostable than dyes. This encouraged us to explore simple and effective approaches to luminescent nanocrystal encoding of polystyrene microbeads and identify suitable polymerization conditions and nanocrystal surface ligands to tackle the presenting challenges. Best results were obtained with nanocrystals bearing a polymer compatible surface ligand dispersed in hydrophobic monomer droplets.[2] Aiming for a minimized loss in nanocrystal fluorescence and prevention of nanocrystal leaking, we systematically investigated the fluorescence properties of the resulting nanocrystal-encoded beads and identified conditions yielding highly fluorescent polystyrene microbeads firmly encoded with different, luminescent nanocrystals suitable for future applications. Possible fields of use include immunoseparation processes in combination with magnetic nanoparticles, tracking cell uptake or identifying biomolecules/proteins for detection of e.g., virus DNA.

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On-Demand Ride-Pooling Dynamics and its Influence on Sustainable Mobility

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Ride-pooling is an efficient technique to lower negative aspects like CO_2-emissions or noise of individual traffic by cars. With ride-pooling similar rides are bundled, implying that the number of vehicles necessary and the number of rides overall decreases. Furthermore, it can be built fast, as the required infrastructure is already constructed in the form of the street networks. To increase the understanding about the system dynamics, my work begins with an investigation about the behaviour of vehicles in a ride-pooling system. Thereby, I will focus on transitions into taxi systems or bus systems, which have been shown in [1] and [2]. Secondly, the combination of ride-pooling with other public transportation systems like railways is explored. On the one side, ride-pooling could function as system along other public transportation systems, which offers users another, more comfortable, way of traveling from one place to another. On the other side, ride-pooling could also be a system that serves only as a supplement for other systems, for example, in last-mile travel. The last part will be a study about the savings that ride-pooling can accomplish, compared to private cars. Savings which will be investigated are the CO_2-emissions, the number of cars necessary, the amount of traffic and the travel time of passengers. This part is complemented with a research about the mode choice of customers. As basic experiment I will use an agent-based ride-pooling simulation, techniques from network theory and game theory as well as methods from stochastics.

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Optimal DAPI Control Topology for Power Grids with MCMC

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Due to climate change, a fundamental transformation of our energy system will be necessary in order to decrease emission of greenhouse gases such as carbon dioxide and meet the goals of the Paris agreement [1]. Besides examining the physical power grid construction, there are also approaches to promote existing grids to so-called Smart Grids, which call for effective control mechanisms [2]. The design of these control topologies is the subject of current research [3, 4].

This work analyses the stability of different power grid and power grid control topologies regarding perturbations to the operating frequency. The synchronisation performance of the grid is characterised by a high survivability, or a low synchronisation norm, or both. Starting from embedded synthetic networks, edges are added and/or removed with the simulated annealing algorithm, thereby improving the transient performance.

The results show that in the droop controlled case, minimising the synchronisation norm in the simulated annealing algorithm significantly improved the survivability compared to the initial grid. However, a trade-off between high survivability and low synchronisation norm was discovered in the proportional integral (PI) controlled systems. The results from applying the simulated annealing algorithm to lower the synchronisation norm show an interesting combination of centralised and decentralised averaging. Control topologies that comprise all edges of the underlying power grid plus connections of all other nodes to a central controller node are shown to reach lower synchronisation norm values than evenly distributed averaging PI control for networks of order $8 \leq N \leq 512$.

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Optimizing energy cell's flexibility potential with stochastic programming

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Role of ancillary services and flexibility in the energy grid grows with the transition to renewable energies. We analyze energy cell's potential to provide ancillary services by using not only reserve storage but also components used in the system operation. Stochastic programming is applied to the optimization problem to model the stochastic nature of ancillary service requests. The investment decision is shown to be sample-independent. Flexibility cost reduction and improvement of flexibility potential given by stochastic optimization are analyzed.