Universität Basel

Research Project

Optical plasmonic nanostructures for enhanced photochemistry

Third-party funded project

Project title Optical plasmonic nanostructures for enhanced photochemistry
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Aim of this PhD project is to combine the experience and know-how of the Constable-Housecroft Group in materials photochemistry and photophysics with the expertise of CSEM Muttenz in simulation, large-area manufacturing, and characterization of optical plasmonic nano-structures, in order to study and demonstrate the potential of these innovative nano-optical surfaces for the production of solar-fuels.

Solar-fuels is a collective term for chemical energy sources created by harnessing solar-energy. Solar-fuels can be used to store the energy in a chemical form and regain it by a chemical process, such as oxidation, as in classical combustion processes. The concept of solar-fuels is quite intuitive in approaching the question on energy storage from the energy density point of view. The highest known energy mass densities can be found in nuclear fission and nuclear decay followed by chemical energy, which exhibits a 10-100 times higher energy mass density then electrochemical energy sources, such as batteries. In nature, photosynthesis is one of the key photochemical reactions for life, although the overall efficiency is low. Since 2007, solar-fuels have raised increasing interest in the scientific community, thus resulting in a tremendous increase of publications, with a particular emphasis on artificial photosynthetic systems.

Main focus of this PhD project will be water splitting to create H2 from water as a solar-fuel: this process yields the maximum energy density for a chemical fuel. In particular, it requires two electrons, while for other solar-fuels like methane or ethanol, multiple electrons per mole of target are required, thus significantly reducing the efficiency of the photo-synthesis process. It is therefore reasonable to focus on the water-splitting approach. In the Constable-Housecroft Group, significant know-how has been developed on the synthesis of catalysts for water-splitting [1]. This PhD project will build on the existing background and aim to develop functional demonstrator devices to prove and assess the overall concept. For the two electron process, it is important to have an overlap in time of two excited electrons that can contribute to the reaction. Recent publications have been based on proton-coupled electron transfer [2,3]. The PhD student will study this latter approach on the efficiency increase that can be achieved using optical plasmonic nano-structures.

Several limitations to the efficiency of the water-splitting are known [4]. One is a low absorption of many semiconductors due to a large and/or not overlapping bandgaps. Others are reflection due to a high refractive index of the semiconductor material and recombination due to a short minority carrier distance. The challenges are similar to those observed in organic (OPV) and Si (Si-PV) photovoltaics, two fields, where CSEM is particularly active through its PV Center. Plasmonic enhancement of the conversion efficiency have been studied for both PV systems. Water-splitting can benefit from this existing know-how: plasmonic structures and/or nanoparticles may lead to a significant enhancement of the conversion efficiency. Plasmonic nanoparticles are already reported in literature as giving significant enhancement to the conversion efficiency. Very similar to the enhancement mechanism in OPV, the nanoparticles act like antennas receiving the light and transferring the plasmon energy to a nearby absorber.

Plasmonic nano-structures in the range of approx. 100 – 200 nm can address similarly the known limitations by injecting hot electrons and holes into the semiconductor [4,5]. Furthermore, nano-structures can be used to increase the effective optical path of photons propagating through photochemical active materials in order to increase the probability of absorption, and thus the overall efficiency. In contrast to the enhancement obtained using plasmonic nanoparticles [6], to the best of our knowledge, both the theoretical and the experimental proof-of-concept of large scale optical plasmonic nano-structures for enhanced water splitting have to not been investigated so far. Plasmonic nano-structures yield the potential for cost effective

and high reproducibility production of large areas, which will be a key for competitiveness of the described technology.

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CSEM Muttenz has a strong background in both refractive and diffractive micro- and nanooptics, as well as in optical plasmonic nano-structures. Design and simulation tools and large-area manufacturing processes are available in-house, which can be used to create well-controlled plasmonic nano-structures and to replicate them on large surfaces. Due to its activities in OPV, CSEM Muttenz is fully equipped with solar simulators and has a strong background in light-management for photovoltaics. The PhD student can therefore rely on proven experimental setups for characterization of the conversion efficiency.

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