Capturing Sunlight for Sustainable Fuel Generation

The Reisner Group

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Artificial Photosynthesis

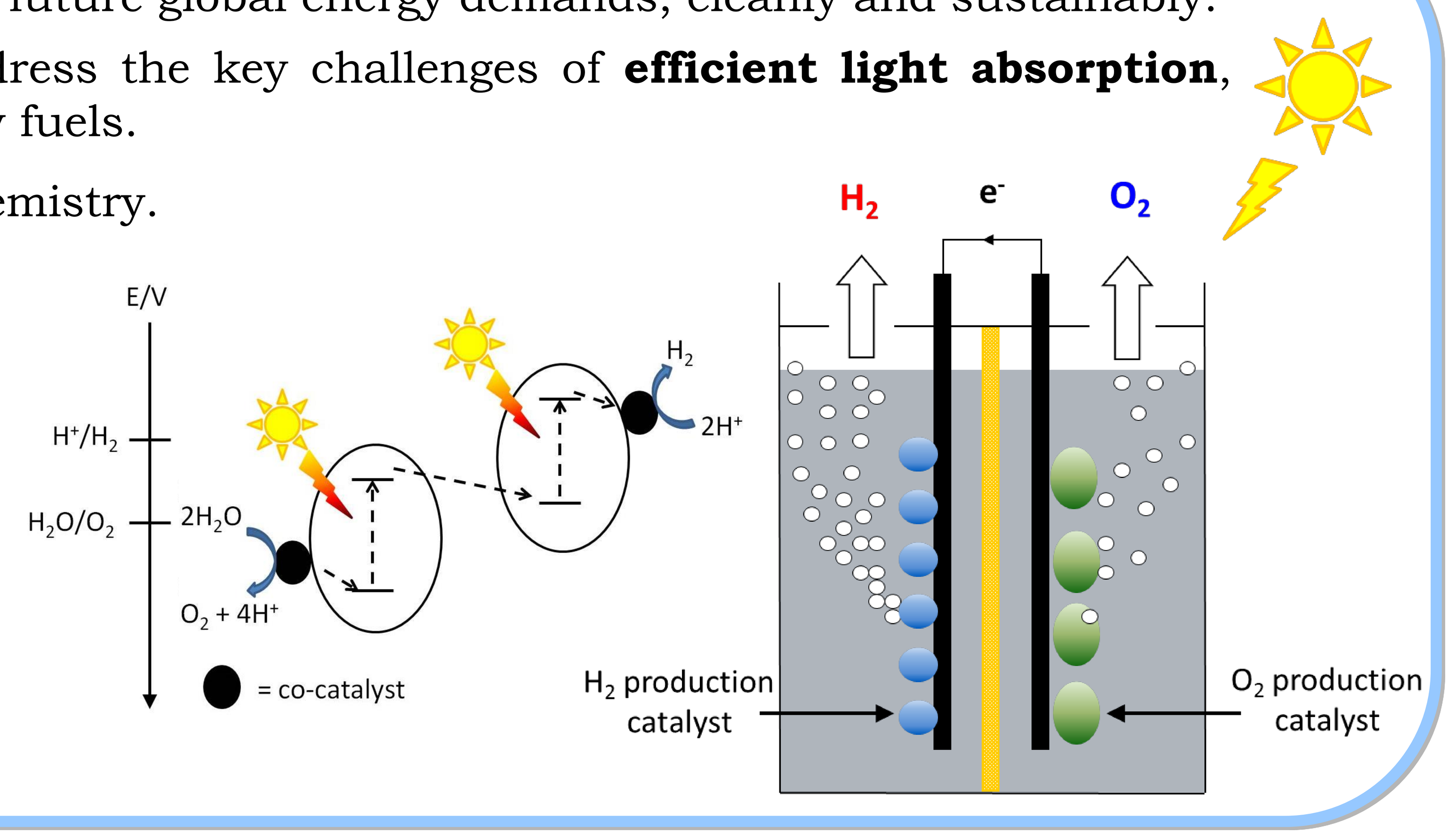
Conversion of solar energy into fuels is one of the most promising means of meeting future global energy demands, cleanly and sustainably.

We develop hybrid systems for each half of the water splitting reaction that address the key challenges of **efficient light absorption**, **effective charge separation**, and **efficient redox catalysis** to generate high energy fuels.

Our research is multi-disciplinary, spanning biological, synthetic, and materials chemistry.

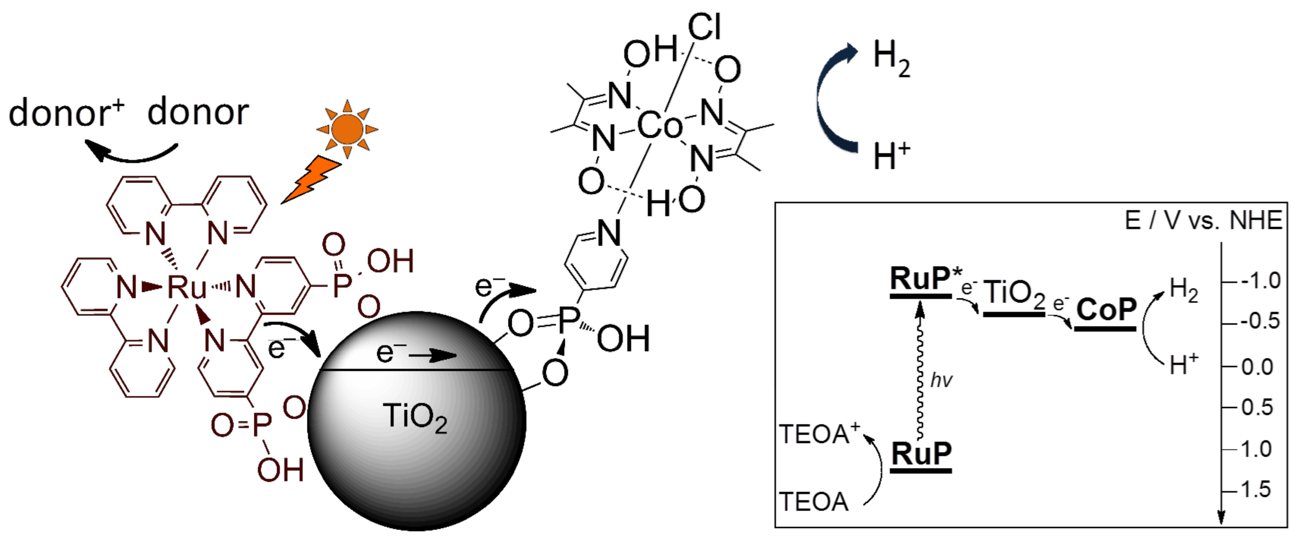
- **Biological materials:** · Incorporated as catalysts · Studied to inspire catalyst design
- **Transition metal complexes:** · Photosensitizers · Solution and surface-bound redox catalysts
- **Nanostructured materials:** · Photoactive materials · Conductive support/electron relay for catalysts

Our ultimate goal is to integrate these systems into a photoelectrochemical cell for complete photocatalysis.



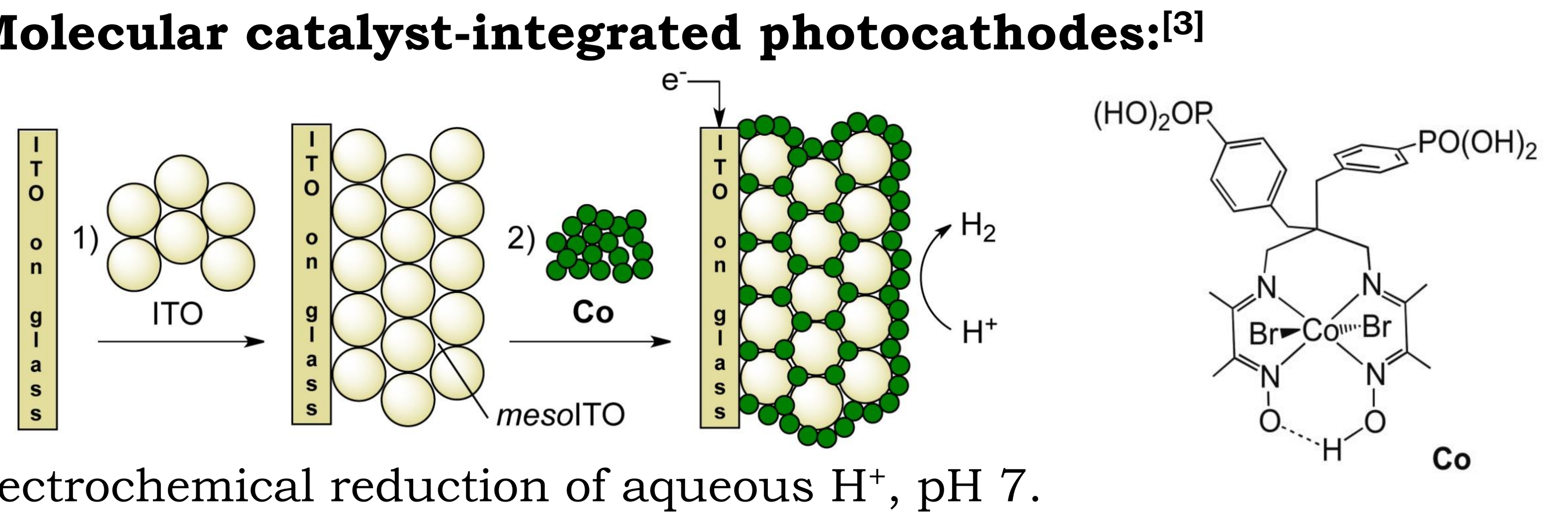
H^+ Reduction

Dye-sensitized nanoparticle-catalyst systems:[1]



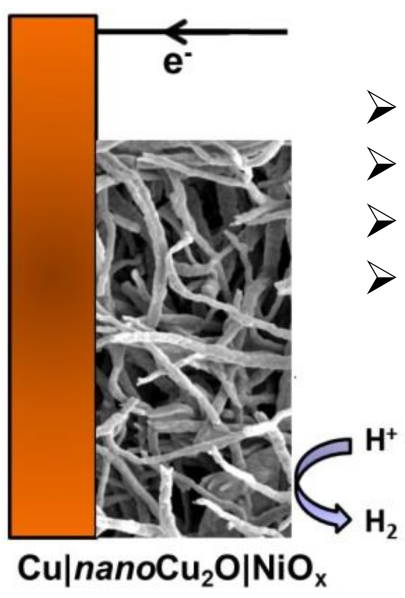
- Solution-based aqueous system, pH 7.
- Highly efficient with both small molecule catalysts (e.g. CoP, shown) and an enzyme known as hydrogenase.
- Dye and catalyst bound *via* groups with high surface binding affinity.
- Dye-sensitized TiO_2 allows for long-lived charge-separated state (up to 1 s) and electron transfer from TiO_2 to CoP is rapid (10 μs).

Molecular catalyst-integrated photocathodes:[3]



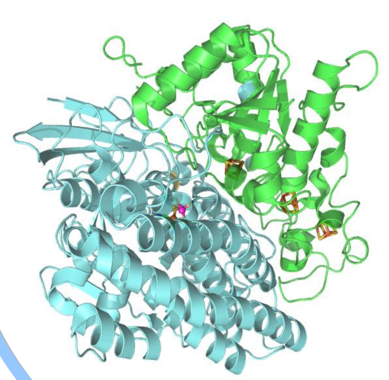
- Electrochemical reduction of aqueous H^+, pH 7.
- Cobalt catalyst has high affinity for nanostructured surface.
- Remains molecular species on *meso*ITO
- Excellent stability and multi-colouration for electrochromic devices

All-inorganic nanostructured photocathodes:[5]

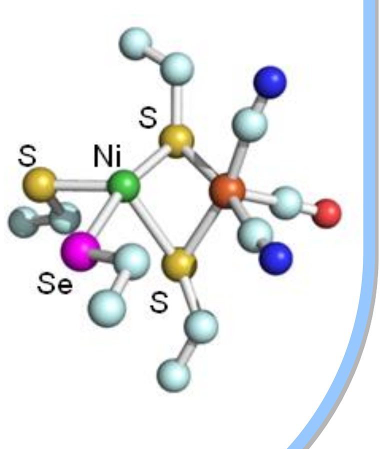


- Nanostructured Cu_2O hybrid electrodes, pH 6.
- High current density due to high surface area.
- NiO_x acts as catalyst and enhances stability.
- Successfully coupled to WO_3 nanosheet photoanode for **complete water splitting**

[NiFeSe] hydrogenase and enzyme mimics:[6]

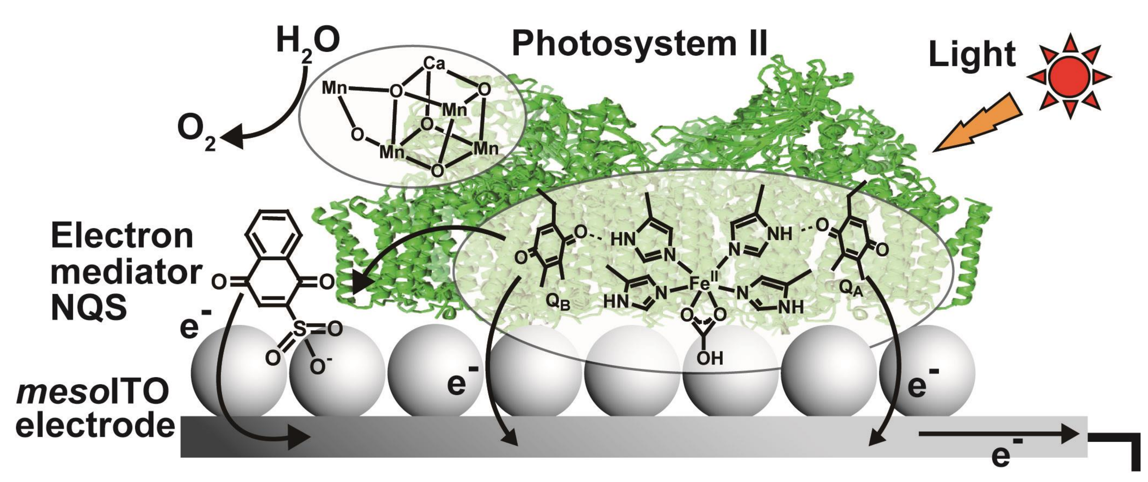


- Visible light driven H_2 evolution with O_2 tolerant [NiFeSe] hydrogenase
- [NiFeSe] active site mimics as precursors for active Ni-nanoparticle films.



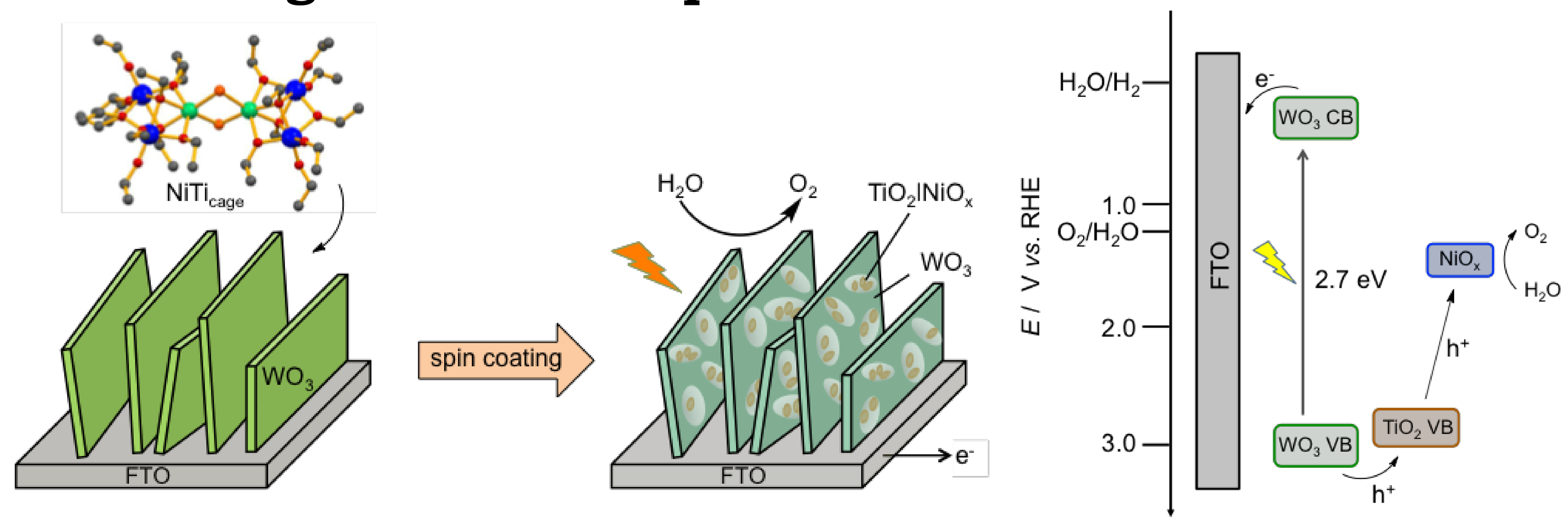
O_2 Evolution

Photosystem II (PSII) integrated photoanodes:[2]



- Photosystem II-film photoelectrochemistry, pH 6.5.
- *meso*ITO is transparent, conducting and allows for high PSII loading
- Direct electron transfer from PSII to electrode
- Electron transfer enhanced by mediator, orientation or covalent binding.

All-inorganic nanocomposite electrodes:[4]



- Leaf-like nanostructured electrode, pH 9.2.
- One-step procedure introduces stability layer and co-catalyst
- Mimics key functional features of PSII.
- $TiO_2|NiO_x$ enhances charge separation and stability.

CO_2 Reduction

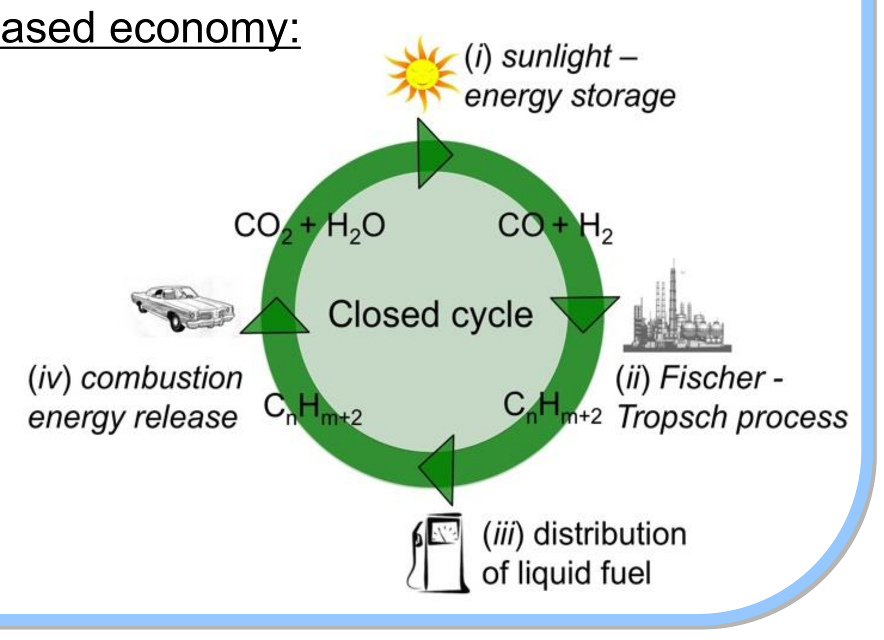
$$CO_2 + H_2O \longrightarrow O_2 + H_2 + CO \quad \text{SynGas}$$

- Reduction of aqueous CO_2 gives renewable C source
- Developing catalysts for CO_2 transformation is a key target.

<u>Towards a sustainable carbon-based economy:</u>

Christian Doppler Forschungsgesellschaft

OMV



[1] (a) Reisner, E. *et al. J. Am. Chem. Soc.* **2009**, *131*, 18457; (b) Lakadamyali, F. and Reisner, E. *Chem. Commun.* **2009**, *47*, 1695; (c) Lakadamyali, F.; *et al. Angew. Chem. Int. Ed.* **2012**, *51*, 9381; (d) Lakadamyali, F. *et al. Chem. Eur. J.* **2012**, *18*, 15464.
[2] (a) Kato, M. *et al. J. Am. Chem. Soc.* **2012**, *134*, 8332; (b) Kato, M.; *et al. J. Am. Chem. Soc.* **2013**, *in press* (DOI:10.1021/ja404699h)
[3] (a) Muresan, N. *et al. Angew. Chem. Int. Ed.* **2012**, *51*, 12749; (b) Scherer, M. R. J. *et al.*, in preparation.
[4] (a) Lai, Y.-H. *et al. Chem. Commun.*, **2013**, 49, 4331; (b) Lai, Y.-H., *et al. Chem. Eur. J.* **2013**, *in press* (DOI: 10.1002/chem.201302641)
[5] Lin, C.-Y., *et al. Chem. Sci.*, **2012**, *3*, 3482.
[6] (a) Sakai, T., *et al. submitted;* (b) Wombwell, C. *et al. submitted.*

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