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Towards Understanding Heterogeneous Catalysis: Surface Reactions on Model Nanostructured Catalysts

The selectivity of multi-pathway surface reactions depends on subtle differences in the activation barriers of competing reactive processes, which are difficult to control. One strategy to overcome this problem is introducing a specific, selective interaction between the reactant and the catalytically active site, directing the chemical transformations towards the desired route. This interaction can be imposed via functionalization of a solid catalyst with ligands, promoting the desired pathway via steric constrain and/or electronic effects. The microscopic-level understanding of the underlying surface processes is an important prerequisite for a rational design of this new class of ligand-functionalized catalytic materials.

In my talk, I will present two case studies. First, hydrogenation of α,β -unsaturated carbonyl compound acrolein over Pd surfaces will be discussed with a particular focus on the selectivity towards C=O vs. C=C bond hydrogenation.¹ We show that selective hydrogenation of the C=O bond in acrolein to form the desired reaction product – an unsaturated alcohol propenol – is possible over Pd(111) with nearly 100 % selectivity. However, this process requires a very distinct modification of the metal surface with a densely packed overlayer of ligand-like surface species -- oxopropyl groups -- that are formed from acrolein during the initial stages of reaction and turn the metal surface highly selective towards propenol formation. By applying pulsed, multi-molecular beam techniques and in-operando IRAS we have identified the chemical nature of the surface ligands and the reactive surface intermediate.

In the second part, I will discuss the mechanisms of keto-enol tautomerization of acetophenone over catalytically active metal surfaces.² Tautomerization to their enol counterparts on metal surfaces is theoretically predicted to enable an easier route for normally very difficult hydrogenation of the C=O bond. We observed that the enol form of acetophenone can be formed on Pt(111) by building a ketone-enol dimer, where one molecule is present in the enol form and is stabilized via H bonding to the carbonyl group of the second ketone molecule. We achieved a clear spectroscopic identification of the ketone-enol dimers and showed that the enol species can exist only if it is stabilized by the neighboring ketone molecule.

References:

1. Dostert, K.-H.; O'Brien, C. P.; Ivars-Barceló, F.; Schauerermann, S.; Freund, H.-J., *JACS*. **2015**, *137*, 13496.
2. Attia, S.; Schmidt, M.-C.; Schröder, C.; Pessier, P.; Schauerermann, S., *Angew. Chem.* **2018**, *57*, 16659.

All interested colleagues are welcome to this seminar lecture (45 min. presentation followed by discussion).
Friedrich Aumayr (LVA-Leiter) Ulrike Diebold (Seminar Chair)