O 67: Gerhard Ertl Young Investigator Award (talks of the selected canditates)

Time: Thursday 10:30–13:00 Location: H37

O 67.1 Thu 10:30 H37

Restructuring of Catalyst Surfaces under Reaction Conditions — •Feng Tao[1,2], Miquel Salmeron[2], and Gabor Somorjai[1,2] — [1]Department of Chemistry, University of California, Berkeley, California, USA, 94720 — [2]Materials Science Division, Lawrence Berkeley National Lab, Berkeley, California, USA, 94720

The surface structure of catalysts and their evolution under reaction conditions is one of the crucial issues in understanding catalytic mechanisms at molecular and atomic levels. Here bimetallic nanoparticles (Rh0.5Pd0.5 and Rh0.5Pt0.5) were used as model catalysts for the in-situ study of surface structure under reaction conditions. The as-synthesized Rh0.5Pd0.5 nanoparticles are core-shell structured. However, there is no significant depth-dependence of atomic fractions of Rh and Pt in Rh0.5Pt0.5 nanoparticles. Surface structures of two Rh-based bimetallic nanoparticle catalysts during catalysis, oxidation, and reducing reactions have been systematically studied in-situ using XPS in Torr pressure ranges. The Rh0.5Pd0.5 and Rh0.5Pt0.5 nanoparticles undergo reversible changes in both atomic fractions and chemical states corresponding to the switch of reaction environments between oxidizing and catalysis (or reducing) conditions. Rh atoms in Rh0.5Pd0.5 and Rh0.5Pt0.5 segregate to surface region and are largely oxidized under oxidizing conditions, while in reducing atmospheres the Pd atoms in Rh0.5Pd0.5 and Pt atoms in Rh0.5Pt0.5 segregate to the surface regions and Rh oxides are largely reduced. The capability of restructuring nanoparticles through chemical reactions suggests a new method for the development of new catalysts.

O 67.2 Thu 11:00 H37

Charge Measurement of Atoms and Atomic Resolution of Molecules with Noncontact AFM — •Leo Gross[1], Fabian Mohn[1], Nikolaj Moll[1], Peter Liljeroth[1], Jascha Repp[2], Franz J. Giessibl[2], and Gerhard Meyer[1] — [1]IBM Research - Zurich, 8803 Rüschlikon, Switzerland — [2]Institute of Experimental and Applied Physics, University of Regensburg, 93040 Regensburg, Germany

Individual gold and silver adatoms and pentacene molecules on ultrathin NaCl films on Cu(111) were investigated using an atomic force microscope (AFM), equipped with a qPlus sensor, operated at 5K.

Charging a gold atom by one electron charge increased the force on the AFM tip by a few piconewtons. Employing Kelvin probe force microscopy (KPFM) we also measured the local contact potential difference (LCPD) which is shifted depending on the sign of the charge. This effect allows the discrimination of positively charged, neutral, and negatively charged atoms.

To image pentacene molecules we modified AFM tips by means of vertical manipulation techniques, i.e. deliberately picking up known atoms and molecules. Using a CO terminated tip we resolved all individual atoms and bonds within a pentacene molecule. By comparison with density functional theory (DFT) calculations we found that atomic resolution is obtained due to the contributions from the Pauli repulsion.

O 67.3 Thu 11:30 H37

Probing the surface state and reactivity of an active catalytic site: from single-site catalysts to nanoparticles — •Elena Groppo — Department of Inorganic, Physical and Material Chemistry, NIS Centre of Excellence and INSTM Unità di Torino, University of Torino

The utility of applying advanced spectroscopic techniques in determining the properties of the active catalytic sites, the difficulties encountered and the possible strategies to overcome the problems, will be discussed in this contribution by considering two case studies, which are among the simplest examples of single-site and multiple-site catalysts, respectively:

(i) the CrII/SiO2 system, which catalyses the ethylene polymerization. Interaction with CO was studied since decades by in situ FT-IR spectroscopy and more recently by Raman spectroscopy, both giving a clear, although indirect, proof that an important rearrangement of the local environment of the CrII site at the silica surface occurs upon CO adsorption. Very recently, XAS experiments offer the first direct experimental evidence of adsorption induced mobility of CrII sites, which is the key for understanding the catalytic activity of CrII sites in presence of adsorbates.

(ii) supported Pd nanoparticles, which catalyse hydrogenation reactions. EXAFS, UV-Vis and FT-IR spectroscopies, coupled with TEM microscopy, are used to investigate the structural, optical and vibrational properties of two Pd/polymer systems in all the synthetic steps, starting from the hosted Pd(OAc)2 precursors up to Pd nanoparticles.

O 67.4 Thu 12:00 H37

Assembly of core-shell superparamagnetic iron oxide nanoparticles and their application to smart materials — •Erik Reimhult — ETH Zürich, Zürich, Switzerland

Application of superparamagnetic iron oxide nanoparticles (SPIONs) as biomedical imaging contrast agents and as smart materials require them to retain high stability even in extremely dilute suspensions, high salt and elevated temperatures. Furthermore, many applications require close control over their hydrodynamic diameter and interfacial chemistry. These requirements can only be met by irreversible steric stabilization. We describe how such stabilization can be obtained using dispersants anchored by biomimetically inspired, electronegatively substituted catechols, which provide irreversible binding of a high density polymer shell to the particle core by grafting to assembly, in contrast to previously described approaches using e.g. DOPA or dopamine. The novel dispersant stabilization allowed creation of multifunctional SPIONs for targeted biomedical imaging with controlled number of ligands and individually optimized core and shell thicknesses. Furthermore, we present how assembly of core-shell nanoparticles at fluid hydrophobic-hydrophilic interfaces provides a route to create hierarchical smart materials, by allowing control of the geometry of monolayer films of inorganic cores serving as nanoantennas for external stimuli within a responsive matrix. We show how irreversible binding affinity of the hydrophobic or hydrophilic dispersant shell greatly increases NP loading into lipid, polymer and NP only membranes and allows the emergence of well characterized responsive properties.

O 67.5 Thu 12:30 H37

The structure of surfactants at the nanoscopic oil-in-water droplet interface and the consequence for the interfacial tension — •Sylvie Roke — Max-Planck Institute for Metals Research Heisenbergstrasse 3, 70569 Stuttgart, Germany

Surfactants such as sodium dodecylsulphate (SDS) can reduce the interfacial tension between bulk water and bulk n-hexadecane by 42 mM/m. Although it is commonly expected that interfacial tension lowering should also take place on the interface of nanoscopic oil droplets in water vibrational sum frequency scattering experiments indicate otherwise. Vibrational sum frequency scattering is a method that can be used to obtain local structural information from the chemical species present at the interface on nanoscopic oil droplets in water. In these measurements we have directly measured the adsorption of SDS onto hexadecane oil droplets with an average radius of 83 nm. We find that the interfacial density of adsorbed SDS is at least one order of magnitude lower than that at a corresponding planar interface. The derived maximum decrease in interfacial tension is only 5 mN/m.