

The active sites in catalysis

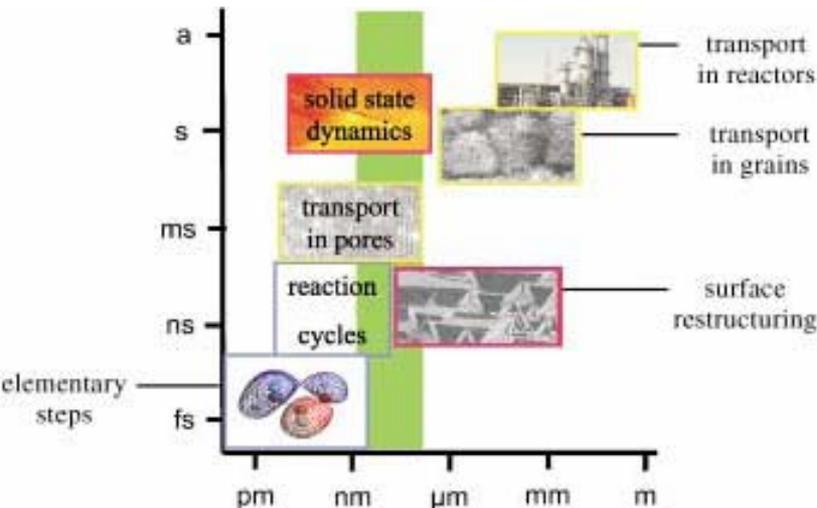
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Ammonia synthesis

- the largest energy-consuming process in the industrial world
- requires ~1% of the world power production

Catalytic phenomena occur at many scales of length and time



Schloegl & Hamid, Angew. Chem. 43(2004)1628

- Catalysts are typically very complex materials
- The catalytic site often forms @ reaction conditions

The active sites

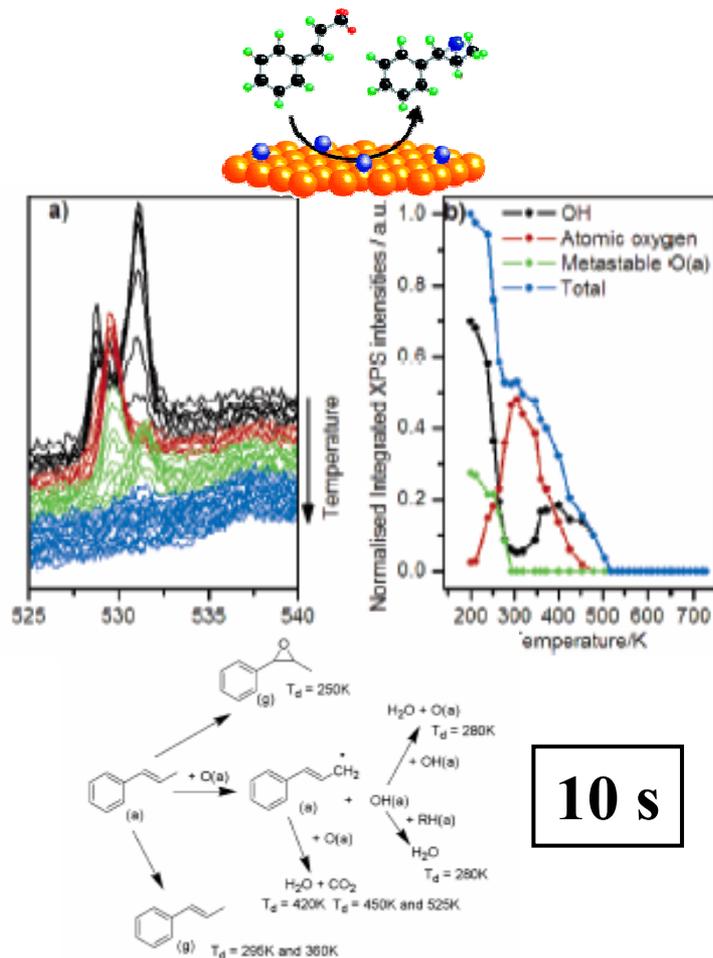
- Static*: steps, vacancies, dislocations
 - Dynamic*: transient surface structures
- The actual active site exists only in presence of the reactants and products

Challenge

- characterize materials and processes at relevant scales (time, space, composition) and under reaction conditions
- establish structure-activity relationship

Fast or high-pressure XPS

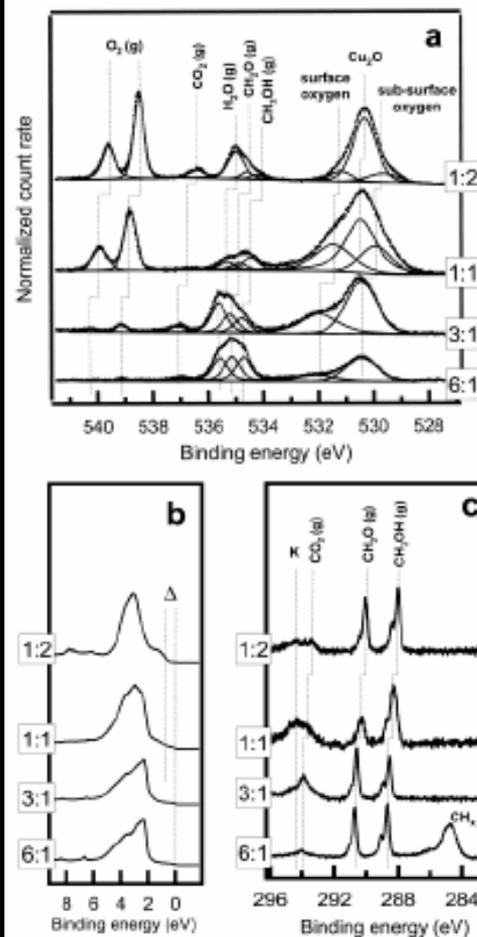
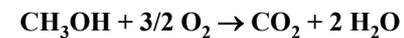
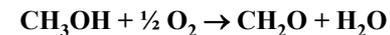
Epoxidation of trans-methyl styrene (Cu(111)) Evolution of surface species: time, temperature



10 s

Lambert et al. in JACS 127(2005)6069

Methanol oxidation on Cu: hp XPS



Subsurface oxygen
only in situ
depth profiling by hv

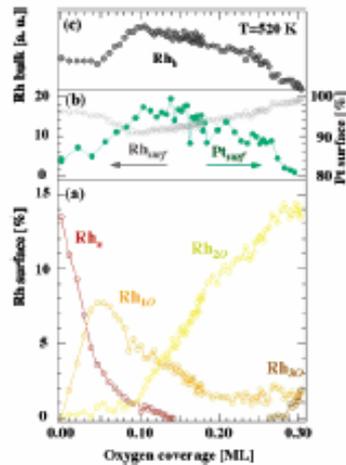
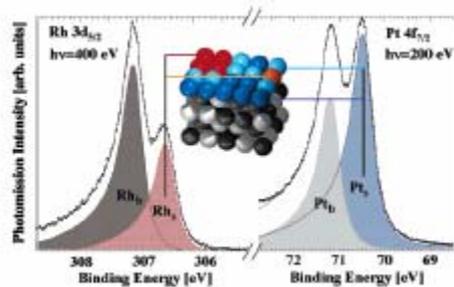
< 10 mbar

Schlogl, Salmeron et al. in JPC 108(2004)14340

Fast high-resolution XPS

Hydrogen + oxygen reaction on Pt₅₀Rh₅₀(100) alloy

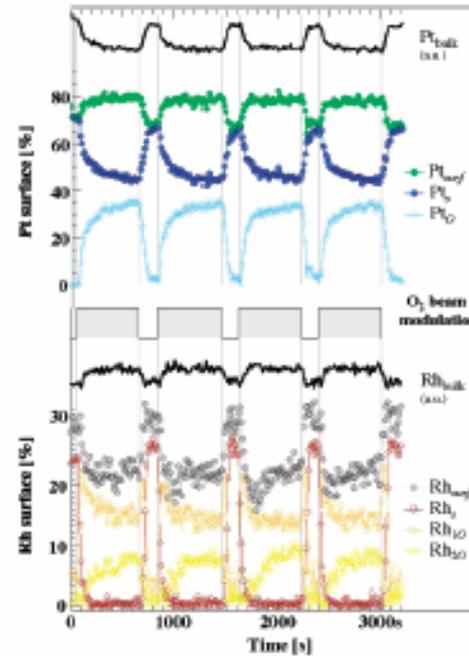
Surface composition/structure depends on the gas environment



6.6×10^{-8} mbar H₂

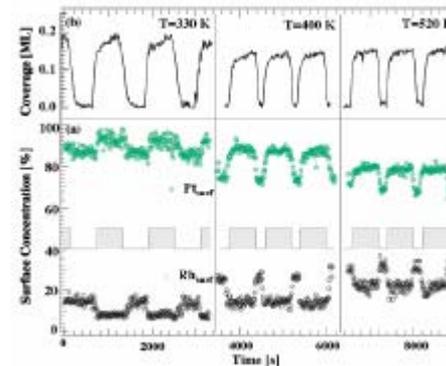
2.5×10^{-7} mbar O₂

Pt enrichment



Surface sensitivity

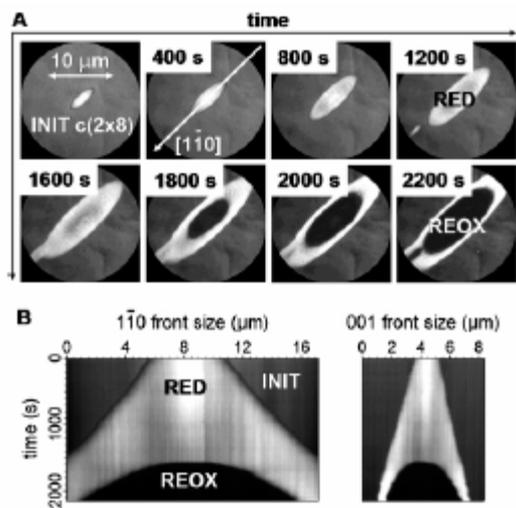
Continuous exchange:
subsurface-surface
in oxygen



15 s
100 meV

LEEM & Photoelectron spectromicroscopy

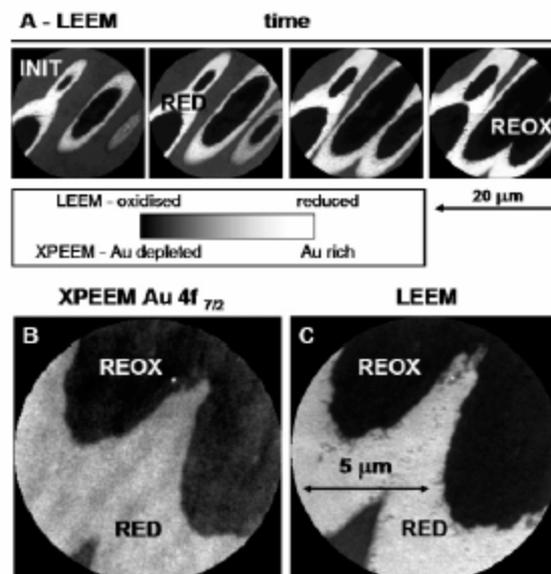
Hydrogen + oxygen reaction on Au/Rh (110)



Ignition and propagation of a reduction front on Au/O/Rh surface followed by reoxidation front

$\Theta_{\text{O}} = 0.5 \text{ ML}$; $\Theta_{\text{Au}} = 0.5 \text{ ML}$
 $3.8 \times 10^{-7} \text{ mbar H}_2$
 $4.4 \times 10^{-7} \text{ mbar O}_2$
 720 K

Temporal evolution of patterned surface: Au-rich and Au-poor areas, reactivity different

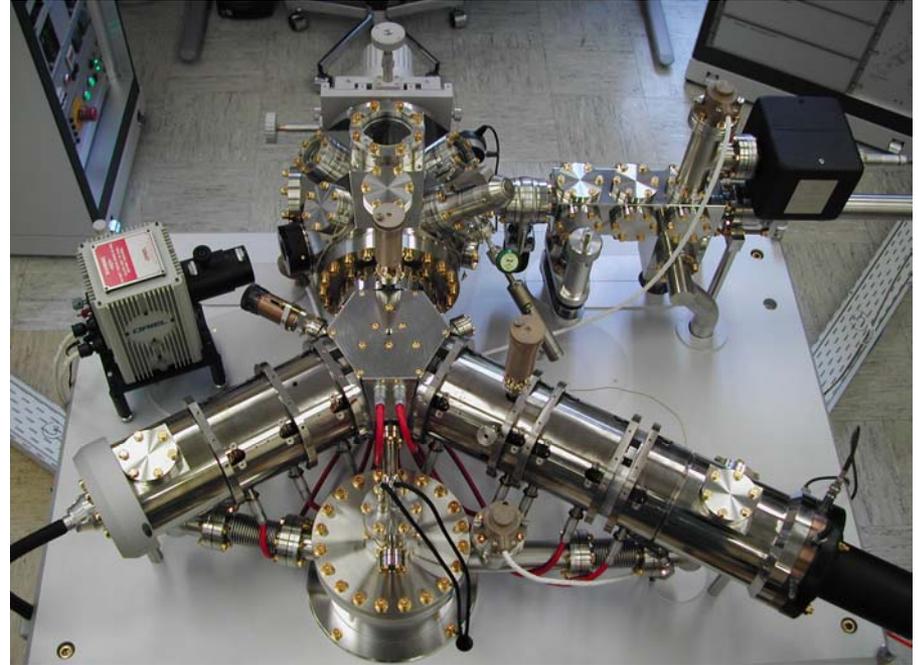
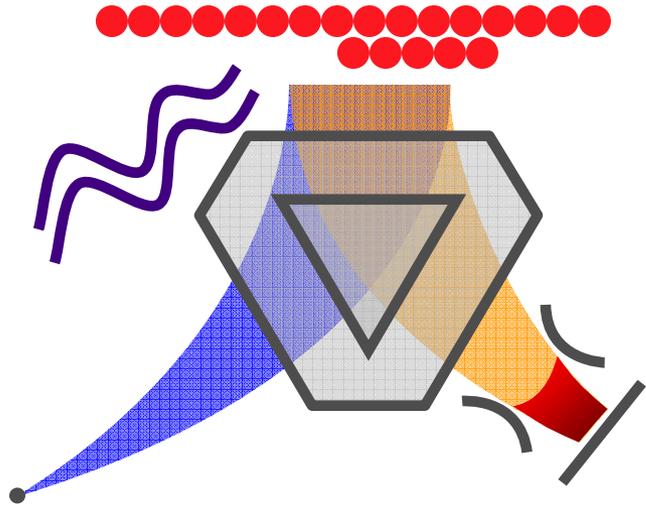


Evolution of reaction fronts on Au/O/Rh surface towards stationary state; Au redistribution

$3.8 \times 10^{-7} \text{ mbar H}_2$
 $4.4 \times 10^{-7} \text{ mbar O}_2$
 620 K

100 nm
< 500 meV
50 ms

New LEEM instrument at CFN/NSLS



Real-time surface imaging while heating, gas dosing

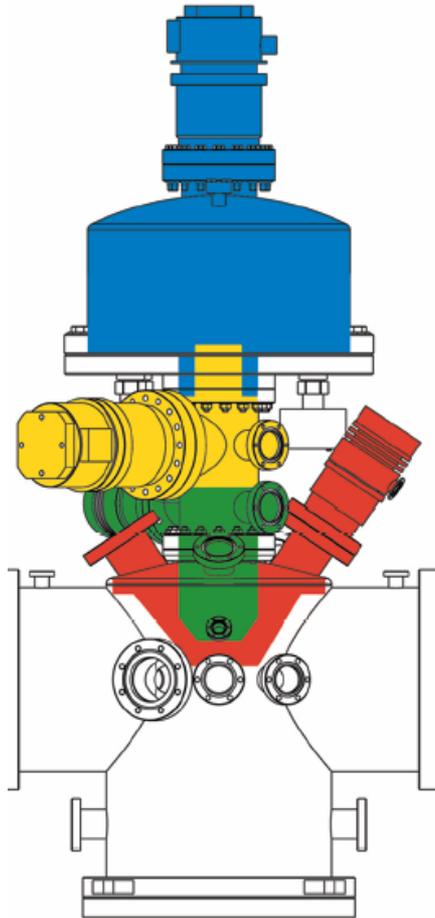
7 nm lateral resolution; Sub-nm vertical resolution

Surface sensitive

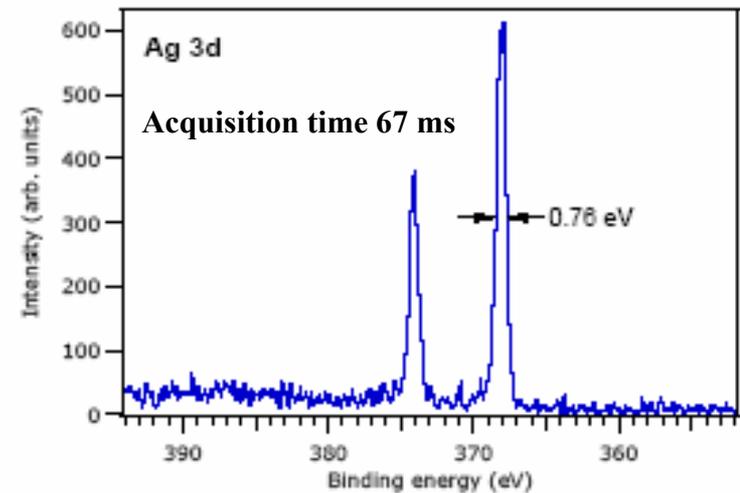
Low-energy electrons

UV/X-ray photons (2006) (Peter Sutter)

Fast high-pressure high-resolution XPS



On our wish list



< 100 ms
3 meV
< 10 mbar