TOWARDS FULL, ATOMIC-SCALE UNDERSTANDING OF CATALYTIC PROCESSES UNDER INDUSTRIAL CONDITIONS

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Under relevant industrial conditions, typically involving high gas temperatures and high gas pressures, the structure and composition of a catalyst surface can be very different from those under the artificial conditions of low temperatures and vacuum that are required for most traditional surface sensitive experimental techniques. This can have dramatic consequences for the chemical reaction mechanisms at work and, as a result, for the performance of the catalyst.

In this talk, I will discuss how we use a combination of surface X-ray diffraction (SXRD) and scanning tunneling microscopy (STM) to obtain direct, atomic-scale information on the structure and the atomic and molecular processes that occur on the surfaces of working catalysts. We have developed special purpose instrumentation for both techniques, SXRD [1] and STM [2], the latter now reaching atomic resolution on metal model catalysts under reaction conditions.

Results will be shown for the oxidation of CO and the reduction of NO on Pt and Pd surfaces under atmospheric pressures at elevated temperatures [2-5]. Our STM- and SXRD-movies demonstrate that at high O_2 pressures these surfaces all oxidize, the catalyst simultaneously switching between two different reaction mechanisms and the oxide being most reactive. We resolve various ultrathin oxide structures, which only appear under high partial pressures of oxygen and others that are only stable under the simultaneous presence of O_2 and CO.

Our findings provide a surprising explanation for the self-sustained oscillations in the reaction rate that appear spontaneously for several of these surfaces. The oscillation mechanism identified here involves a novel role for atomic steps in catalysis [6].

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