

Surface Science Investigations on Structures and Adsorption Sites of Single Crystal PdGa Surfaces

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With its highest technological and economic importance, catalysis is an extremely active research area, which yields in a great impact on the development of new catalyst systems with the aim to produce more efficient and selective chemical processes. Thereby, a key issue is to gain detailed knowledge of the underlying molecular mechanisms which relate the structure, composition, and chemical bonding of the topmost catalytically active surface layer with the reactant.

Recently, intermetallic Pd-Ga compounds were presented as extraordinary selective catalysts for the semi-hydrogenation of acetylene [1], a key process in the polyethylene production. These intermetallic compounds exhibit specific separations of the catalytically active sites [2] and thereby reconcile the usually mutually excluding properties of high activity and concomitant high selectivity. Additionally, the crystal structure of PdGa belongs to the $P2_13$ space group and therefore this system features an intrinsic chirality and polarity [2,3].

As a first step, the stable surface terminations were determined and explored, since they define the activity and selectivity of the catalyst. For this purpose, we investigated the surface atomic and electronic structures of low Miller indexed PdGa single crystals, serving as model catalysts.

In a next step, high-resolution scanning tunneling microscopy (STM) enabled to directly visualize the adsorption geometry and conformational changes in small hydrocarbon molecules upon reaction. In combination with DFT calculations different extent of changes in the adsorption site for the different molecules on the PdGa(hkl) surfaces were discovered [4]. Special emphasis was put on the role of step edges as they are usually most reactive for molecular adsorption and catalytic processes.

Our investigations relay mainly on UHV techniques such as STM, X-ray photoelectron spectroscopy and diffraction (XPS and XPD), angle-resolved photoelectron spectroscopy (ARPES), I(V)-Low Energy Electron Diffraction (LEED-I(V)) and atomistic simulations. Thereby, the precise surface stoichiometry and the identification and characterization of relevant adsorption sites were revealed and lead to a better understanding of the materials properties giving rise to a high selectivity towards a specific reaction product, even though, it remains a challenge to bridge the gaps that were opened by the idealized conditions typically applied in these studies.

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