In situ Microscopy of Oxide Growth and Transformation under Reaction Conditions

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Current research in heterogeneous catalysis aims to provide a deeper understanding of all the components in a real catalytic system, which depending on the conditions typically comprises both metals and oxides in nanoparticulate form. Significant progress has been achieved by studying carefully devised model systems that facilitate detailed investigation of the structure and chemistry of the individual constituents in controlled environments. While traditional approaches have focused on the role of oxide-supported metal nanoparticles, in the so-called inverse configuration a nanosized metal oxide is supported on a transition metal, thereby allowing us to assess the properties of the nanoscale metal oxide and its defect chemistry as well as to gain complementary access to the oxide-metal interface.

In this presentation, we will focus on the epitaxial growth of ultrathin metal oxide films and nanostructures on transition metals, with a special emphasis on rare-earth oxides. These materials have attracted considerable attention owing to their rich chemistry and enhanced reducibility in proximity to transition metals. We will demonstrate that real-time monitoring of their synthesis under vacuum conditions as well as their structural and chemical modifications on the nanometer scale in reactive gaseous environments is possible using low-energy electron microscopy and related methods [1]. Primary examples will address reversible structural transformations in ruthenium, platinum, and copper supported cerium oxide inverse model catalysts [2-5] upon thermal and chemical reduction, e.g., from CeO_2 via the cubic Ce_2O_3 phase to hexagonal Ce_2O_3, concomitant with partial dissolution of the cerium oxide particles and considerable dispersion of metallic cerium on the substrate, resulting in irreversible morphological changes. Similar sesquioxide and dioxide phases are present directly after deposition of praseodymium oxide on the Ru(0001) surface [6, 7], illustrating an intrinsic nanoscale complexity and the importance of the oxide-metal interface.

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