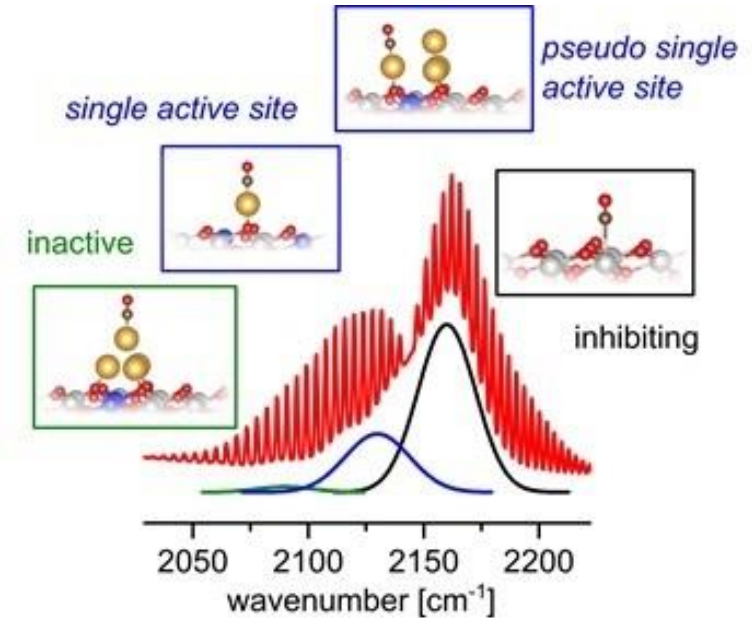
Direct detection of active centers in gold catalysts

Combination of operando IR spectroscopy with theory



While gold is generally considered inert, nanoparticulate gold has high catalytic activity for reactions at low temperatures and even allows oxidation of carbon monoxide below room temperature. Since its discovery, gold catalysis has developed into an independent and intensively researched field of catalysis. It is therefore all the more astonishing that fundamental aspects of the functioning of such supported gold catalysts have not yet been clarified.

For example, for gold catalysts based on cerium dioxide carriers, metallic gold on the one hand and cationic gold on the other hand have been suggested in the literature as active centres; the nuclearity of the active centre is also controversially discussed. To clarify these questions, the physicochemists Dr. Christian Schilling, Marc Ziemba and Prof. Dr. Christian Hess from the Eduard-Zintl-Institute of the TU Darmstadt in cooperation with Dr. Verónica Ganduglia-Pirovano from CSIC in Madrid recently gained direct access to the detection and structural identification of active centers in gold/cerondioxide catalysts by combining operando infrared spectroscopy, i.e. the application of vibrational spectroscopy under catalyst operating conditions, with modern DFT calculations.

In the analysis of gold/cermic oxide catalysts during the oxidation of carbon monoxide, isolated Au+ centres are identified as active centres for the reaction, which together with oxygen from the cerium dioxide carrier (Olattice) and the reactant carbon monoxide form Olattice-Au+-CO species (see figure). The generation and characterization of such isolated active sites is an exciting field of research as they allow to minimize the consumption of precious metals in catalysts. Interestingly, as shown by operando infrared spectroscopy, the isolated Au+ centers on gold/cermic oxide catalysts can be formed under reaction conditions after adsorption of carbon monoxide in a dynamic process, independent of the catalyst pretreatment. The results therefore underline on the one hand the

structural dynamics of catalysts, and on the other hand the need for operando methods which can visualize the changes of catalysts under reaction conditions.

Further information:

Christian Schilling, Marc Ziemba, Christian Hess, M. Verónica Ganduglia-Pirovano, J. Catal. 383 (2020), 264-272. https://doi.org/10.1016/j.jcat.2020.01.022