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Report on the PhD thesis of David Simonne entitled:

Catalytic properties at the nanoscale probed by surface x-ray diffraction and coherent diffraction imaging

The PhD thesis of David Simonne deals with the timely topic of operando investigations of the ammonia oxidation of Pt nanoparticle ensembles, single crystal surfaces and single Pt nanoparticles. Ammonia oxidation is an important topic for ammonia combustion, which is produced from clean hydrogen. Ammonia is an important greenhouse gas, which needs to be removed from the exhaust. For the investigation, different types of surface sensitive x-ray diffraction methods are employed, together with Bragg coherent diffraction imaging and x-ray photoemission spectroscopy. The experiments are supported by online mass spectrometry, demonstrating the catalytic activity. Before his work, the following questions were open:

- How does the shape of Pt nanoparticles supported by alumina evolve under ammonia oxidation conditions?
- How does the strain of individual Pt nanoparticles evolve under ammonia reaction conditions?
- How can the nanoparticle results be compared to Pt single crystal surfaces?

The thesis is very well structured. In chapter 3 of his thesis, David Simonne discusses the structural evolution of supported Pt nanoparticles under ammonia oxidation conditions (3.2: collective behavior, 3.3: single nanoparticles). In chapter 4 the results from the Pt (111) and (100) surface are presented. As main result, he finds that Pt nanoparticles undergo a reshaping when the reaction conditions are changed. At the same time, the Bragg CDI experiments reveal that the strain state of the single nanoparticles is changed under reaction conditions, which can have impact on the reaction rate and mechanism itself. Concerning the interaction of ammonia / oxygen mixtures with Pt single crystal surfaces, he observed distinct differences: the Pt (100) surface interacts much stronger with the gas mixture, as compared to the (111) surface, e.g. by the formation of surface reconstructions.

David Simonne has presented a very impressive scientific work, allowing to correlate structural changes on the surface of Pt nanoparticles with those on Pt single crystal surfaces under ammonia reaction conditions. The work was conducted with very high scientific care. One shortcoming, that might be corrected, is the lack of information about the nanoparticle sizes for the ensemble experiments, as well as in many figures of the Bragg CDI results.
The discussion of the results in the literature context is very well conducted and most important literature results of the vast amount of available literature are included in the

discussion. The written presentation of the results is of high quality and very convincing.
I recommend to authorize David Simonne to defend his PhD thesis.

Andreas Stierle