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Time-resolved Bragg coherent diffraction imaging of Pd nanoparticles during in situ acetylene hydrogenation

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The study of nanoparticle (NP) assisted chemical reactions is important for the development of efficient catalyst materials for a wide range of environmental and energy applications [1]. Such studies are primarily focused on the role of surface sites - whether on particle facets or vertices. The rate and efficiency of heterogeneous catalysis is dependent on these distinct adsorption energies and turnover rates [2]. In addition to facet-dependent catalytic activity, lattice strain is known to influence the reactivity of metal surfaces [3]. One technique that can be used to probe lattice displacement with pm precision is Bragg Coherent Diffraction Imaging (BCDI) [4]. From the lattice displacement, the local strain of metal NPs can be examined in situ during catalyst-enhanced reactions. In this study, we probe the hydrogenation of acetylene on Pd nanoparticles (NP) [5], where the partial hydrogenation to ethylene is desired. This relies on fine control of the sample temperature, gas pressure, and the reactant ratios [6].

In order to probe the kinetics of heterogeneous catalysis a dynamic probe of strain evolution is found using real-time, and stroboscopic BCDI at the ID01-EBS beamline of ESRF [7]. This is applied to Pd NP in situ during acetylene adsorption, hydrogen absorption, and under acetylene hydrogenation. With this study it is possible to track the evolution of the average lattice expansion of the particle as a function of time. By distinguishing the changes in local lattice parameter under each of these conditions, we can understand the role of various Pd facets in the partial hydrogenation of alkynes [5]. With this knowledge, we performed stroboscopic BCDI on Pd NP under reversible reaction conditions to observe the mechanism with a time resolution of 0.3 s. The evolution of the lattice parameter on the sub second timescale shows a clear facet dependence which we believe is linked to the preferential adsorption of acetylene. We observe that the top (111) facet exhibits a lattice contraction relative to the bulk of the Pd NP within a few seconds upon introduction of acetylene gas. Through DFT simulations we will unravel whether the rate-limiting step is pressure stabilisation or the surface reaction with adsorbed hydrogen as has been previously proposed [6].

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