Abstract Submitted
for the MAR14 Meeting of
The American Physical Society

Reaction-driven restructuring of Pt and Pd catalysts: In operando X-ray absorption spectroscopy study ANNIKA ELSEN, ULRICH JUNG, Department of Chemistry, University of Illinois at Urbana-Champaign, Urbana, IL 61801, YUANYUAN LI, ANATOLY FRENKEL, Department of Physics, Yeshiva University, New York, NY, 10016, RALPH NUZZO, Department of Chemistry, University of Illinois at Urbana-Champaign, Urbana, IL 61801 — The catalyzed hydrogenation of ethylene on supported metal catalysts has been intensively investigated, mainly because this reaction lies at the heart of many industrial processes. Most previous studies have been performed using surface science techniques in UHV. Therefor little is known about the nature of the active state of the catalyst at ambient pressure where the kinetics is very different. We employed operando X-ray absorption spectroscopy (XAS) to correlate the structural changes of SiO_2-supported Pt and Pd catalysts with their activity for ethylene hydrogenation. The XAS experiments were performed at the beamlines X19A and X18B, NSLS, BNL. For both catalysts, strong and largely reversible transformations of the metal bonding were identified at about the maximum ethane conversion. The changes were different for Pt/SiO_2 and Pd/SiO_2 due to the ability of the latter to form bulk hydride, while the former can only adsorb hydrogen on the surface. As a result, Pt/SiO_2 undergoes disordering of the surface, leading to a strong reduction of the Pt-Pt coordination number under H_2-deficient conditions, while the main effect for Pd/SiO_2 is the hydrogen uptake with concomitant increase in Pd-Pd bond length. The correlation between these different kinds of order transitions and differences in rates for these catalysts will be discussed.

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Date submitted: 11 Nov 2013 Electronic form version 1.4