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The origin of catalytic activity of supported noble-metal nanoparticles S.N. RASHKEEV, A.R. LUPINI, S. J. PENNYCOOK, S.T. PANTELIDES, Oak Ridge National Laboratory — Supported Au nanoparticles <5 nm show a sharp rise in the low-temperature catalytic oxidation of CO while the reverse occurs for Pt nanoparticles. Subsequent experimental and theoretical investigations focused on Au nanoparticles and reached conflicting conclusions, attributing the enhanced Au activity to particular nanoscale features such as perimeter sites and low-coordination atoms, or to a particular bilayer structures, independent of particle size. Here we report atomically-resolved Z-contrast images of on TiO_2- supported Au nanoparticles and theoretical results on an ensemble of rutile- and anatase- supported Au and Pt nanoparticles. We show that high catalytic activity requires that (i) reaction barriers are small, (ii) reaction barriers are smaller than desorption energies of reacting molecules. As nanoparticle size is reduced, attached molecules induce reconstruction of Au clusters, resulting in looser Au-Au bonding and higher desorption energies and smaller reaction barriers for the attached molecules. Pt clusters get tighter by attached molecules, resulting in larger reaction barriers. The bilayer gold structures (M. S. Chen and D. W. Goodman, Science **306**, 252 (2004)) are extremely active because local reconstruction significantly reduces coordination of Au atoms. This work was supported in part by DOE Grant DE-FC02-01CH11085 and by DOE Division of Chemical Sciences under contract No. DE-AC05-00OR22725 with ORNL.

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