The origin of catalytic activity of supported noble-metal nanoparticles
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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) reac-
tion 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 desorp-
tion 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 ex-
tremely active because local reconstruction significantly reduces coordination of Au
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