Abstract Submitted
for the MAR13 Meeting of
The American Physical Society

Rationale for the high reactivity of the interfacial sites in methanol reaction on Au/TiO2(110) SAMPYO HONG, TALAT RAHMAN, University of Central Florida — We have performed density functional theory calculations of methanol decomposition on gold nanoparticle supported on a partially reduced TiO2(110) surface. Our calculations show that the adsorption geometry of 13 atom gold nanoparticle strongly depends on the reduction level of the TiO2(110) surface such that a 30% reduced TiO2(110) surface prefers a hemispherical shape while a 10% reduced TiO2(110) surface prefers a flat shape. This hemispherical geometry of gold nanoparticle has a highest density of interfacial sites among the investigated geometries (flat, spherical, hemispherical ones), which may be a reason for the known high reactivity of interfacial sites towards various reactions on supported gold nanoparticles. We have found that methanol decomposition reaction occurring in the interfacial sites is much facile than that occurring in the non-interfacial sites of TiO2(110) surface in agreement with experiment [1]. We have found that the high activity of the interfacial sites is in fact, a result of charge transfer induced Coulomb interaction among the gold, reactant, and reducible TiO_2 atoms through the formation of ionic O-Au bond between gold and methoxy in the active sites, which turns the participating perimeter gold atom cationic. A direct result of such charge transfer induced repulsion is tilting of the methoxy axis, which leads to facile reaction of methoxy through C-H scission with the bridge oxygen atoms that are readily available from the reducible support. Work supported by DOE Grant No. DE-FG02-07ER15842. [1] S. A. Tenney, B. Cagg, M. Levine, W. He, K. Manandhar, and D. A. Chen, *Surf. Sci.* 606, 1233 (2012).

Sampyo Hong
University of Central Florida

Date submitted: 28 Nov 2012 Electronic form version 1.4