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Au nanoparticles: catalytic activity and CNT support stabilization. Insights from the theory.

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The structure, electronic properties, and catalytic activity toward oxygen activation of gold nanoclusters with size between 10 and 42 atoms were investigated with first principle methods. Nanoparticle symmetry, bond lengths, and surface charge distribution were analyzed and compared to those of macroscopic gold surfaces. Irregular charge distribution was found on the surfaces of nanoparticles consisting of fewer than 30 gold atoms. Nanoparticles with more than 30 atoms were characterized with core-shell charge separation, e.g, positively charged core and negatively charged surface. The charge distribution on those nanoparticles significantly differs from the charge distribution on macroscopic gold surface. The structure and electronic properties of the gold nanoparticles were related to their catalytic activity toward the aerobic oxidation of organic molecules. It was found that oxygen is activated by partially negatively charged surface gold atoms. Nanoparticles with sizes between 10 and 30 gold atoms could only activate oxygen over the negatively charged surface active sites, whereas larger nanoparticles could activate oxygen over the whole surface.

The nature of interaction between sub-nanometer size gold nanoparticles and single wall carbon nanotubes was further investigated. Systematical calculations of the ground state geometry, binding energy, and electron density spatial distribution were performed for large variety of nanoparticle-nanotube complexes. Investigated nanotubes varied in diameter size (0.5 nm - 1.5 nm), chirality, i.e., armchair tubes and zigzag tubes, and electronic properties, i.e., metallic tubes and semiconductor tubes. The calculation results revealed that the nanoparticle-nanotube binding interaction depends on and could be controlled by the surface curvature. Thus, an effective mechanism was proposed for the fixation of small gold clusters with supreme catalytic activity on support surfaces. The obtained results were compared with the interaction of gold nanoparticles with the planar graphene surface. The theoretical predictions are in good agreement and provide fundamental understanding to experimental observations reported in the literature for metal nanoparticles size dependence on the support curvature.