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Towards Catalysis by Gold Clusters: reaction cycles and poisons ELIZABETH C. BERET, LUCA M. GHIRINGHELLI, MATTHIAS SCHEFFLER, Fritz-Haber-Institut, D-14195, Berlin — Nanosized gold particles are good catalysts in a variety of oxidation reactions. These reactions, for which oxidation of CO to CO_2 serves as a paradigm, imply a transition in the total spin and therefore do not occur spontaneously in the gas phase. In the catalytic process, the catalyst clusters are exposed to an atmosphere of gas-phase O_2 and CO reactants at finite temperature and pressure. We have thus modeled free gold clusters in contact with an atmosphere composed of O_2 and CO by means of DFT calculations (PBE functional), and accounted for both temperature and pressure effects employing *ab initio* atomistic thermodynamics. On the basis of this analysis, we could recognize the thermodynamic driving force of the catalytic CO oxidation process and single out the possible (p, T)-dependent reaction cycles and those paths leading to stable structures that poison the catalytic process. This as a useful (exploratory) theoretical step, before taking chemical reaction kinetics into consideration. In the proposed reaction paths, the total spin is conserved in each elementary step, and it is the adsorption of an incoming O_2 molecule that drives the catalyst cluster from the singlet to the triplet spin state, and *vice versa*.

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