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Hydrogen Promoted Oxygen Activation by Free Gold Cluster Cations[1] ROBERT N. BARNETT, BOKWON YOON, UZI LANDMAN, School of Physics, Georgia Institute of Technology, SANDRA M. LANG, THORSTEN M. BERNHARDT, Institute for Surface Chemistry and Catalysis, University of Ulm, Germany — In this contribution we present experiments and first-principles density functional theory calculations on gas-phase reaction of small gold clusters, aiming at elucidation of the role of hydrogen in the activation of molecular oxygen for the selective oxidation of hydrocarbons. Positively charged gold clusters. Au_4^+ and Au_6^+. were chosen because electronic factors and experimental data suggest them to be most suitable for promoting the oxidation of unsaturated hydrocarbons. Our investigations show that, although small gas phase gold cluster cations are inert toward molecular oxygen, the pre-adsorption of molecular hydrogen cooperatively activates the adsorption of O_2 on Au_4^+ and Au_6^+. Temperature and reaction time dependent investigations in an octopole ion trap under multi-collision conditions reveal that hydrogen promotes the activation and dissociation of molecular oxygen on the gold clusters at temperatures as low as 200 K. The detailed mechanism of the hydrogen induced oxygen activation, involving an intermediate hydro-peroxy-complex is revealed by the DFT calculations.

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Robert Barnett
Georgia Institute of Technology

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