Despite the great advances in synthesis and structural determination of atomically precise, thiolate-protected metal nanoclusters, our understanding of the driving forces for their colloidal stabilization is very limited. Currently there is a lack of models able to describe the thermodynamic stability of these ‘magic-number’ colloidal nanoclusters as a function of their atomic-level structural characteristics. In addition, the catalytic properties of metal nanoparticles are strongly dependent on their structural characteristics, such as their size and shape. In this talk, I will address how computational modeling can give insights into both the nanoparticle synthesis1-4 and catalysis5-7, and accelerate the discovery of stable and active nanocatalysts. Specifically, this talk will focus on Au nanostructures and catalytic reactions of environmental interest, such as the CO2 reduction.