Supporting Information for Publication

Integrated Computational and Experimental Structure Refinement for Nanoparticles

Min Yu, Andrew B. Yankovich, Amy Kaczmarowski, Dane Morgan, and Paul M. Voyles**

Department of Materials Science and Engineering, University of Wisconsin-Madison, Madison, Wisconsin 53706, USA

Mackay Icosahedral Structure of 309-Atom Au Nanocluster

The Mackay icosahedron model is shown in Figure S1 with atomic coordinates listed in the supporting files. Atomic coordinates of GA simulated structure matching with model STEM image are also listed in the supporting files and show good agreement of the individual atomic coordinates.

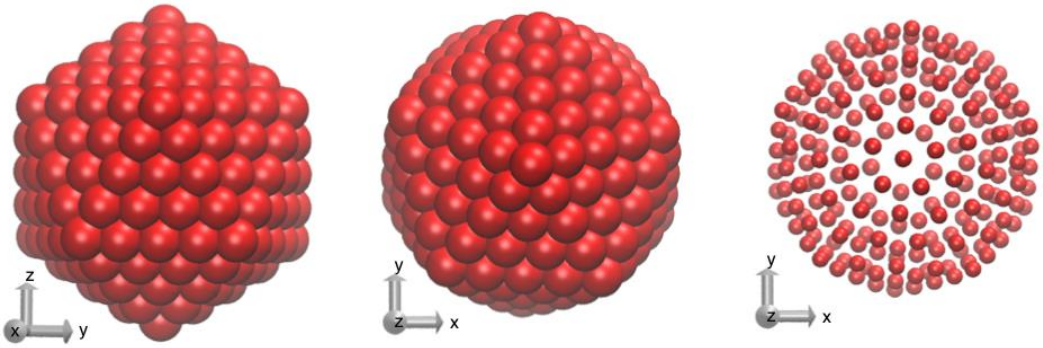

Figure S1: The model structure of Mackay Icosahedron in different orientations.

Ino-Decahedral Structure of 309-Atom Au Nanocluster

The Ino-decahedron model is shown in Figure S2 with atomic coordinates listed in the supporting files. Atomic coordinates of GA simulated structure matching with model STEM image are also listed in the supporting files and show good agreement of the individual atomic coordinates.

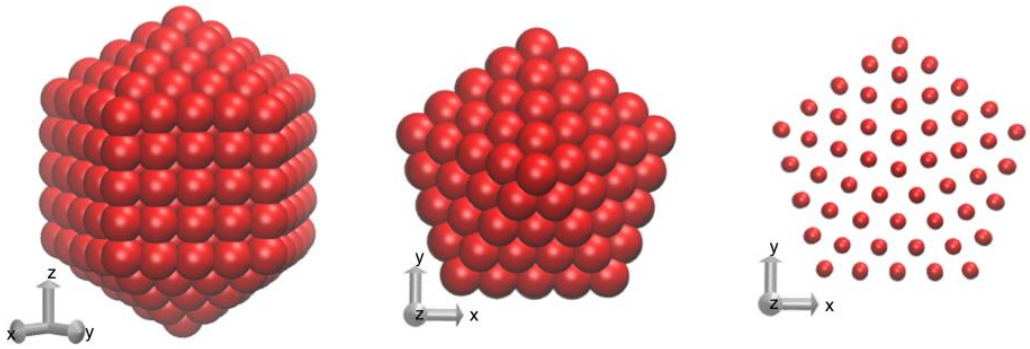

Figure S2: The model structure of Ino-decahedron in various orientations.

Au nanoparticle

The metastable GA optimized structure (Figure S3, atomic coordinates in the supporting files) shows rough surface facets through simultaneous optimization of energy and comparison of the simulated and experimental STEM image. Starting with this GA optimized structure, we perform a further restricted energy-only optimization with a new mutation scheme to simultaneously shift up or down a number of neighboring atomic columns as described in the second section of supporting information. This leads to a configuration (Figure S4) with smoother surfaces, 13.1 meV/atom lower in energy and an increase of 1.35 in the $\alpha\chi^2$ term. Overall, this smooth surface configuration has a worse

fitness function. To reproduce the experimentally observed nanoparticle structure, it is better to apply the integrated GA approach to simultaneously optimize STEM image and nanoparticle energy, which leads to the metastable structure with rough surfaces shown in Figure S3.

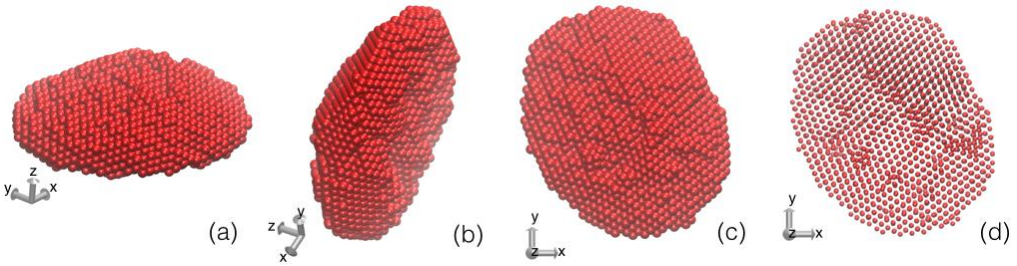

Figure S3: GA optimized structure of Au nanoparticle in different orientations.

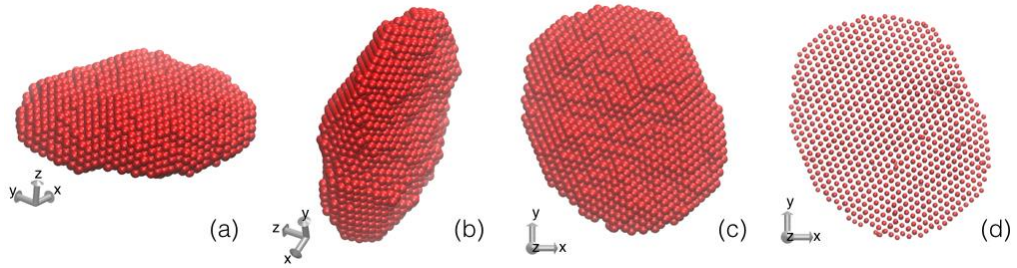

Figure S4: Restricted energy-only optimized structure of Au nanoparticle in different orientations.