

**Supporting Information:**  
**Influence of Cluster Sources on the Growth  
Mechanisms and Chemical Composition of  
Bimetallic Nanoparticles**

Murilo Moreira,<sup>†,‡,¶</sup> Levi C. Felix,<sup>†,¶</sup> Emmanuel Cottancin,<sup>‡</sup> Michel Pellarin,<sup>‡</sup>  
Daniel Ugarte,<sup>†</sup> Matthias Hillenkamp,<sup>†,‡</sup> Douglas S. Galvao,<sup>†</sup> and Varlei  
Rodrigues<sup>\*,†</sup>

<sup>†</sup>*Gleb Wataghin Institute of Physics, State University of Campinas, Campinas, SP,  
13083-970, Brazil*

<sup>‡</sup>*Institute of Light and Matter, University Lyon, Université Claude Bernard Lyon 1,  
CNRS, UMR5306, Villeurbanne F-69622, France*

<sup>¶</sup>*These authors contributed equally to this work*

E-mail: varlei@ifi.unicamp.br

## Supporting Information Available

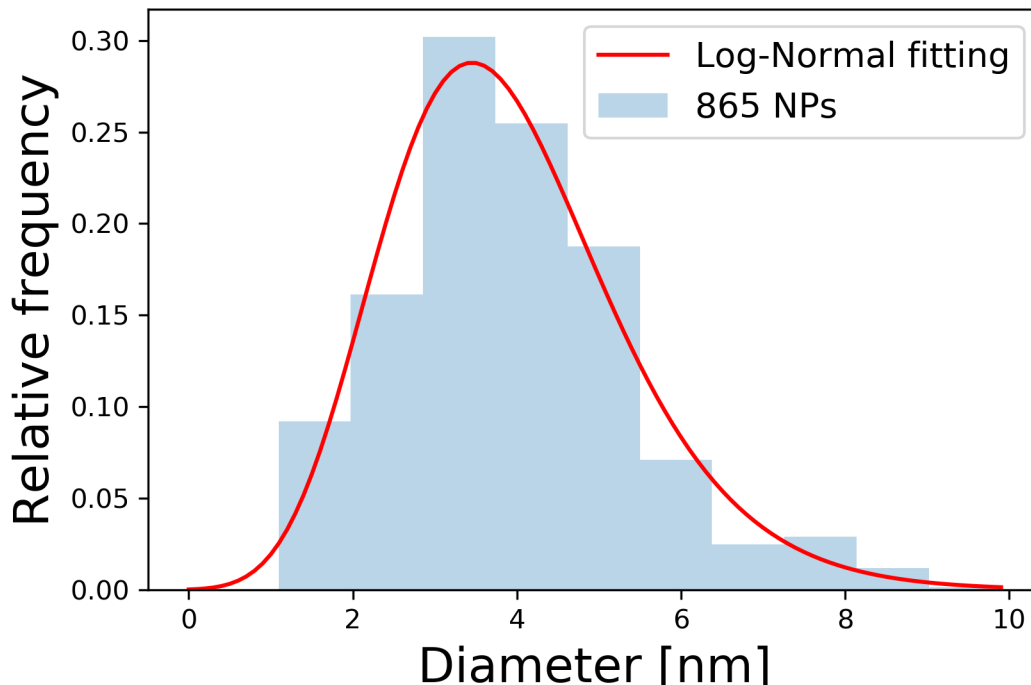


Figure S1: Histogram of AuAg NPs diameter distribution measured by TEM. The log-normal curve is adjusted on the histogram with a peak 4 nm and FWHM of 3 nm.

### Simulations of thermal evaporation

To study the evaporation dynamics of Au and Ag in a bimetallic nanoparticle, we build a  $40.9 \text{ \AA} \times 40.9 \text{ \AA} \times 40.9 \text{ \AA}$  cubic nanoalloy containing 4000 Ag atoms (see Figure S2 (a) at 100 K). Then, we considered three cases where Au ones replaced 25%, 50%, and 75% of the atoms. A temperature ramp using a Nosé-Hoover thermostat was applied, where the temperature of the system is increased from 100 K to 5000 K, linearly on average, as shown in Figure S2 (b). As the nanoalloy is heated, it assumes a spherical shape (snapshots shown in Figure S2 (a)). We observed the individual evaporation of Ag and Au atoms for all three relative percentages of each element, as can be seen in Figure S2 (c). From temperatures slightly higher than 3000 K, the nanoparticles of all compositions begin to

evaporate. We chose this value to be the initial temperature in the evaporative cooling simulations described in the main manuscript. We also carried out the same procedure for a  $12.27 \text{ \AA} \times 12.27 \text{ \AA} \times 12.27 \text{ \AA}$  cubic nanoalloy containing 100 atoms in total to determine how the evaporation of Au and Ag change for smaller sizes, as shown in Figure S2 (d). In addition, for the case where there is 50% of Au, we also simulated the thermal evaporation process for nanoparticles with 500 and 1000 atoms to evaluate the size dependence of Ag and Au dissociation from the original particle (Figure S2 (e)). The time evolution of the chemical composition of nanoparticles was analyzed using the Visual Molecular Dynamics (VMD) software through TCL scripting.

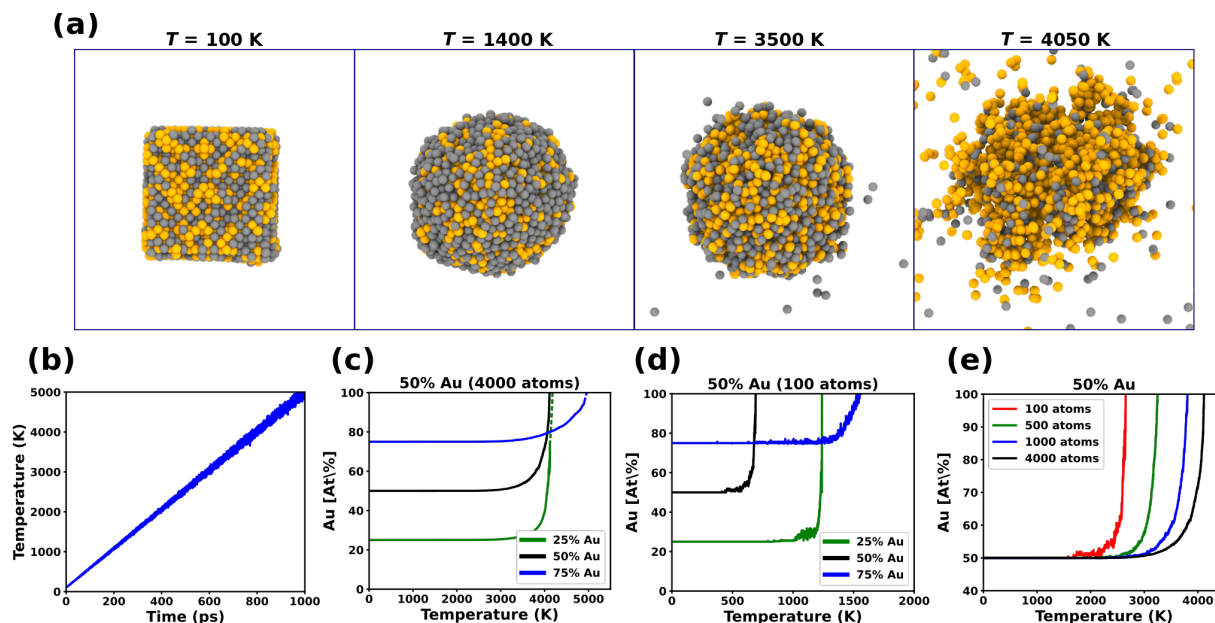


Figure S2: Thermal evaporation of an Au-Ag nanoparticle. (a) Structural evolution for successive indicated temperatures of a cubic nanoalloy containing a total of 4000 atoms. The relative amount of gold and silver is such that we have the following three cases: 25%, 50%, and 75% in Au. (b) Nosé-Hoover thermostat set the time evolution of the temperature of the system with an approximately linear profile. Au's composition as a function of temperature for nanoparticles with (c) 4000 atoms and (d) 100 atoms. (e) Size dependence of the gold composition for nanoparticles initially with 50% Au.

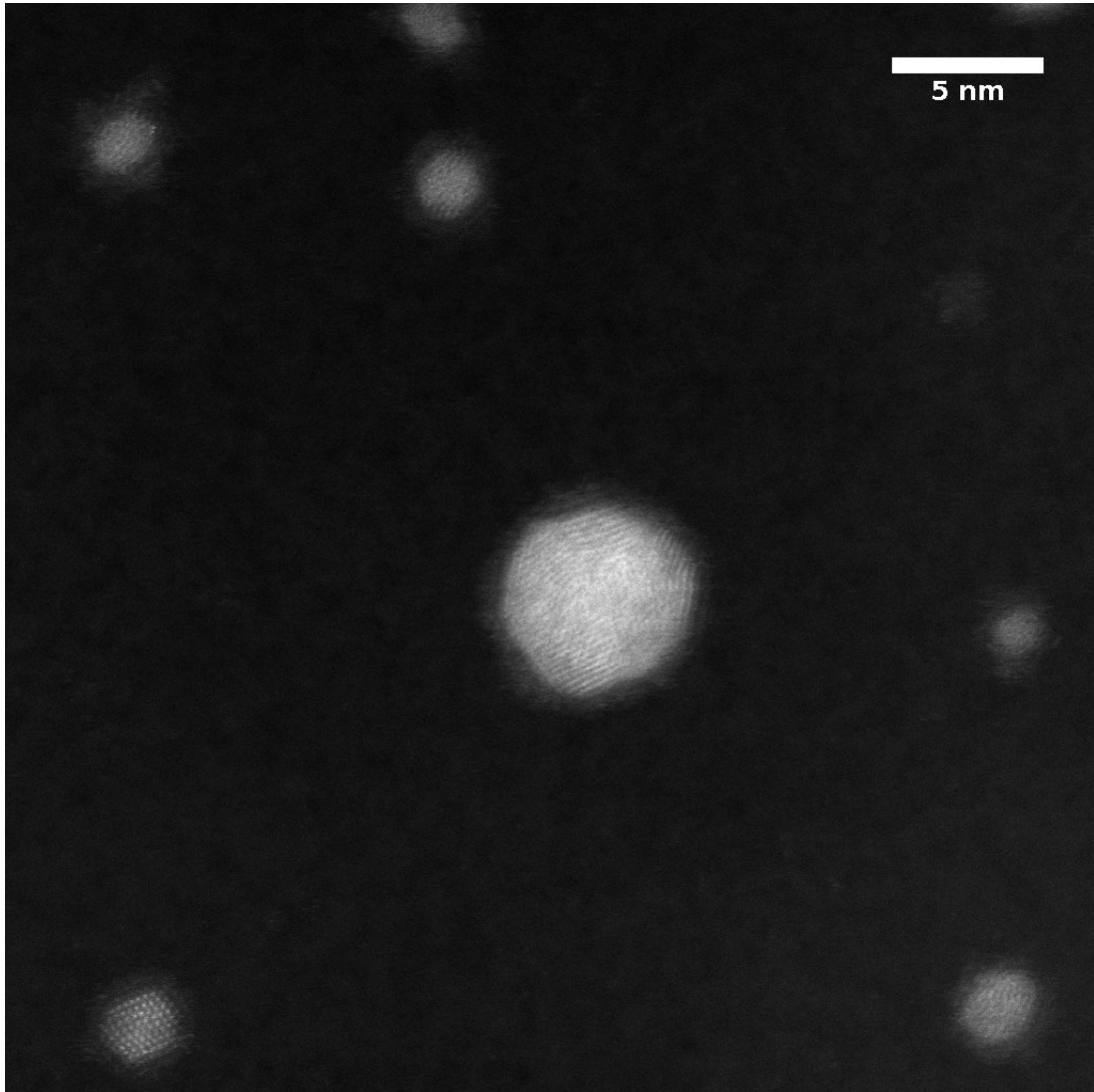


Figure S3: HAADF STEM image of AuAg BNPs produced by HCMSS.