



# Gold nanodumbbell-seeded growth of silver nanobars and nanobipyramids



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## ABSTRACT

Gold nanodumbbells (NDs) are prepared by the reduction of gold ions in the presence of gold nanorods. Gold NDs are then employed for the synthesis of gold–silver core–shell nanoparticles (Au@Ag NPs). The quasi-ellipsoidal NPs could be found at room temperature, but Au@Ag bar and triangular bipyramid (TBP) NPs were obtained at 75 °C. Our results show that the long ends of gold NDs are in the position of the bar center and closely paralleled the shorter edge of TBP. Mechanisms in the growth of silver on gold NDs are proposed for the formations of these Au@Ag NPs.

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## 1. Introduction

Noble metal nanoparticles with a high density of electrons usually possess the characteristics of chemical and physical properties [1]. Most of noble metal nanoparticles could be easily prepared by chemical reduction methods, and they could play the important role as the catalysts in catalysis [2–4]. On the other hand, localized surface plasmon resonance (SPR) occurs strongly on these metal nanoparticle surfaces [5,6]. Because both gold and silver nanoparticles exhibit strong SPR bands in the ultraviolet (UV) and visible regions, many researchers have frequently used them as the substrates for the surface enhanced Raman scattering (SERS) detection of the adsorbed analyte molecules [7,8]. For example, gold nanorods (NRs) and nanowires could exhibit their particular SPR bands in the visible and near-infrared (NIR) regime, respectively. The wavelength and intensity of the SPR band of gold NRs could be modified by changing of their shapes and the coating of the silver metal on their surfaces [9–13]. The latter has the advantage of a higher enhanced sensitivity of SERS than that of gold surfaces [14].

The morphological modification of gold NRs has been achieved in the literatures such as dog-bones, dumbbells, and peanuts [15–19]. The usual approach is to manipulate the anisotropic overgrowth of gold NRs that were usually used as seeds in the presence of a growth solution. At the same time, different parameters of the reaction condition employed in the process of growth could be fine-tuned to transform gold NRs into the new forms [19]. Reaction temperatures, pHs and concentrations of reactants (for example, surfactants, ascorbic acid, and metal ions such as Au and Ag) are

the major factors in the secondary growth. Among reported modification methods, some researchers employ ‘one-pot reaction’, in which gold NRs and reactants remained in the original growth solution, and others first collected NRs after centrifugation and then re-dispersed in a fresh solution before adding all the other new reagents [17]. In the former, all the excess reagents used in the synthesis of NRs and the reaction products still remain in the original growth solution, the control of the reaction in the former is more complicated than that in the latter and thus could have the unexpected influence on the control of the anisotropic overgrowth [19].

In this Letter, we report a simple and effective method to synthesize gold nanodumbbells (NDs) from gold NRs via the secondary growth. Our studies show that gold NDs with longer aspect ratios could be easily prepared by the addition of gold ions and ascorbic acid in the presence of gold NRs. Further, silver ions are subsequently coated on the surface of gold NDs in various reaction conditions and the shape of the resultant Au–Ag core–shell nanoparticles (i.e., Au@Ag NPs) strongly depends on the reaction temperatures. Two peculiar shapes of nanoparticles appeared in bars and triangular bipyramids (TBP) could be obtained at the higher temperatures. Finally, our studies show that the long ends of gold NDs are in the position of the bar center and closely paralleled the shorter edge of TBP, respectively.

## 2. Materials and methods

### 2.1. Chemicals

Silver nitrate and sodium borohydride were obtained from Sigma–Aldrich and Alfa Aesar, respectively. All other reagents were

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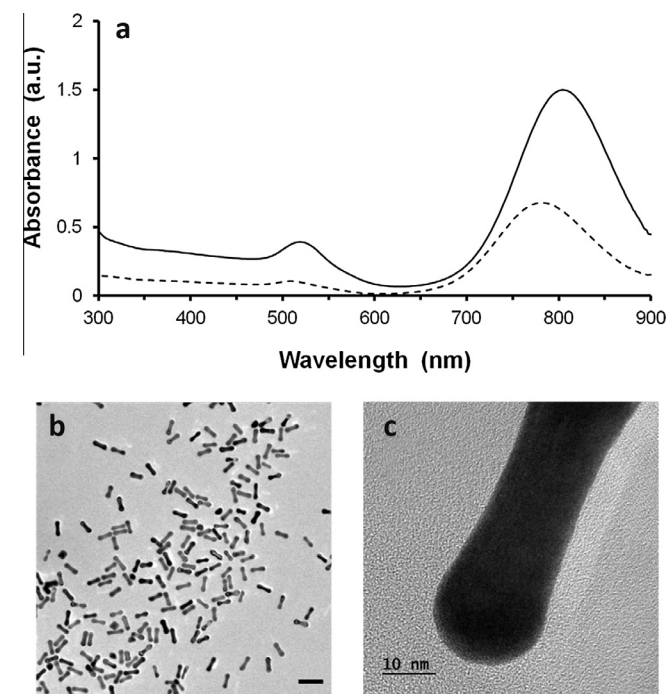
from Acros. All chemicals were used as supplied without purification.

## 2.2. Characterization

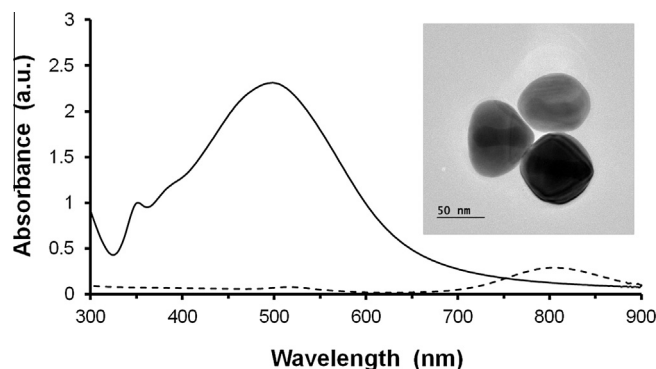
UV/visible (UV–Vis) absorption spectra were recorded on a Hitachi U-3900 spectrophotometer. The transmission electron microscopy (TEM) images were obtained on JEOL JEM-2100F microscopes operating at 200 kV. The morphologies of Au@Ag nanoparticles were characterized using Zeiss Sigma scanning electron microscopy (SEM) at 2.5 kV. A drop of the nanoparticle solution was placed on a carbon-coated copper grid and silicon wafer for TEM and SEM experiments, respectively.

## 2.3. Synthesis of Au nanodumbbells

In a typical synthesis of Au NDs, gold NRs were first prepared by a modified seed-mediated growth method published by El-Sayed and coworkers [7]. 5 mL of 0.25 mM HAuCl<sub>4</sub> and 0.1 M cetyltrimethylammonium bromide (CTAB) was mixed with 300  $\mu$ L ice-cold 10 mM NaBH<sub>4</sub>. The resulting mixture was then stirred at room temperature. After 3 h, the gold particles were formed and used as seeds for the synthesis of gold NRs. 5 mL of 0.5 mM HAuCl<sub>4</sub> and 0.1 M CTAB was mixed with 50  $\mu$ L of 10 mM AgNO<sub>3</sub>, followed by the addition of ascorbic acid (AA) (32  $\mu$ L, 0.1 M) and gold seed solution above (10  $\mu$ L). After being stirred at room temperature for 17 h, the reaction solution was centrifuged at 8000 RPM for 20 min. The supernatant was discarded and the NR-containing solution was diluted with 1 mL of 5 mM CTAB. The same procedure of centrifugation and dilution was repeated to give a 10 mL NR-containing solution of 5 mM CTAB. After that, 31.1  $\mu$ L of 79 mM AA was added to the above 5 mL NR-containing solution of 5 mM CTAB. HAuCl<sub>4</sub> (138  $\mu$ L, 25.4 mM) was subsequently added after 10 min of the reaction. After 1 h, Au NDs were formed in the reaction mixture and then subjected for collecting absorption spectra and microscopic images.



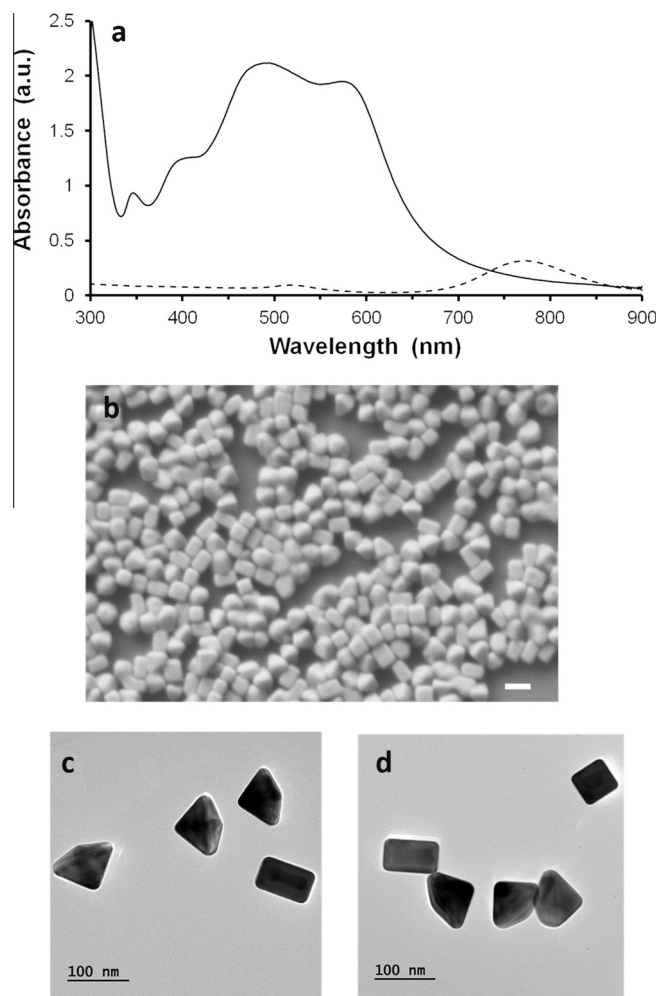
**Figure 1.** (a) Absorption spectra of gold NRs in CATB before (dash line) and after (solid line) the addition of AA and HAuCl<sub>4</sub>. TEM images of Au NDs at low (b) and high (c) magnifications. The scale bar in panel (b) is 100 nm.



**Figure 2.** Absorption spectra of gold NDs in CATB before (dash line) and after (solid line) the addition of AgNO<sub>3</sub>, AA and NaOH at room temperature. The inset shows the TEM image of Au–Ag core-shell nanoparticles (i.e., Au@Ag NPs) obtained in the reaction.

## 2.4. Synthesis of Au–Ag core-shell nanoparticles

The as-prepared gold NDs described in Section 2.3 were used as seeds for the synthesis of Au–Ag core-shell NPs. Typically, the ND-containing reaction solution prepared above was centrifuged twice and diluted to give a 25 mL ND-containing solution of 10 mM



**Figure 3.** (a) Absorption spectra of the gold NDs in CATB before (dash line) and after (solid line) the addition of AA and HAuCl<sub>4</sub> at 75 °C for 5 h. SEM (b) and TEM images (c and d) of Au@Ag NPs in panel (a) after heating. The scale bar in panel (b) is 100 nm.

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