Supporting information

1 Morphology of the source gold NPs prepared using LAL

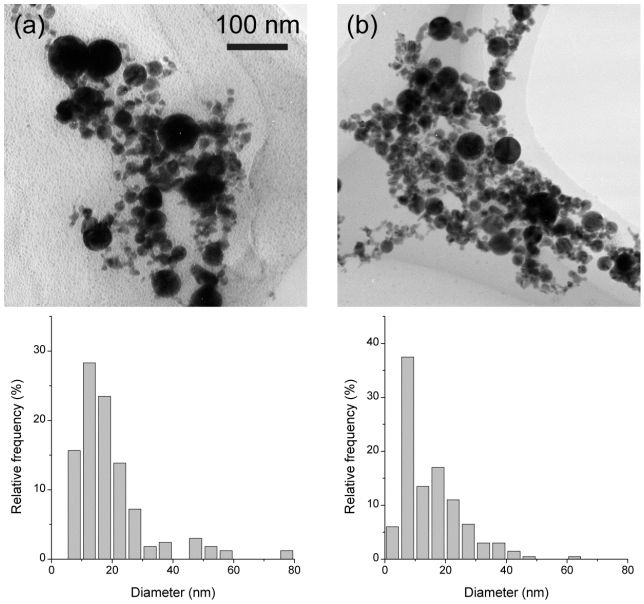

Fig. S1 TEM images and size distribution of the source gold NPs prepared using LAL of a gold plate in (a) pure water and (b) 0.005 mM citrate. Focused 1064 nm laser at 36 J/cm^2 was used. The average diameter of the NPs is 18 nm. The respective average diameters and the standard deviations of these NPs are (a) 18, 11 nm, (b) 15, 10 nm.

2 Stabilization of gold NPs by citrate

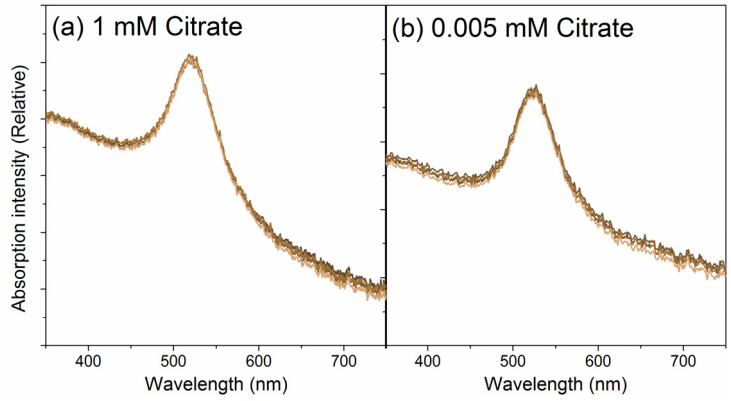

Fig. S2 Temporal changes in the UV–Vis spectra of the source gold NPs in (a) 1 mM and (b) 0.005 mM citrate solution without LI observed upto 30 min. Solutions were stirred during measurements. The measurements interval was 5 min.

3 NMR analysis of the citrate decomposition

The influence of LI on citrate in colloidal solutions of gold NPs was investigated by means of ^{1}H NMR (ECA-600 (600 MHz); JEOL) using a model system to obtain a sufficient amount of citrate for NMR measurements. LI was conducted for 30 min for 10 3-mL colloidal solutions containing 0.1 mM citrate. Those LI conditions were set to prevent the formation of SMPs because the efficiency of LI for Au NPs and citrate molecules on the NPs decreases after SMP formation. These colloidal solutions were gathered, evaporated, and then dispersed in D_2O before NMR measurements.

4 Estimation of the concentration of NPs

By means of an atomic absorption spectrometry, we estimated the concentration of gold atoms in a colloidal solution showing 0.72 of the absorption intensity at 400 nm in the UV-Vis spectra at $1.6*10^{-5}$ g/mL. Using this relation, the concentration of gold NPs (C_{NP}) in a colloidal solution of which the absorption intensity at 400 nm is A_{400} was estimated by following equation.

$$C_{NP}(number/mL) = 1.6\times10^{-5}\frac{A_{400}}{0.72}\frac{3}{4\pi(d_{NP}/2)^3\sigma}$$

$$C_{NP}(mM) = \frac{C_{NP}(number/mL)}{6.0*10^{17}}$$

Here, d_{NP} is the average diameter of the NPs, σ is the density of gold (19.32 g/cm^3).
In this study, we assumed that d_{NP} = 15 nm for all colloidal solutions