

**Supporting Information for**

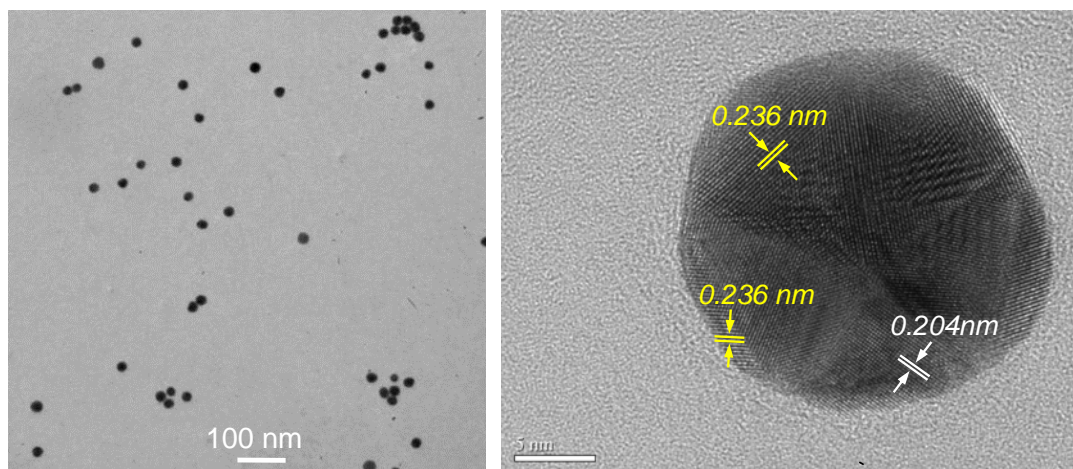
**High-Purity Gold Nanocrystal Dimers: Scalable Synthesis  
and Size-Dependent Plasmonic and Raman Enhancement**

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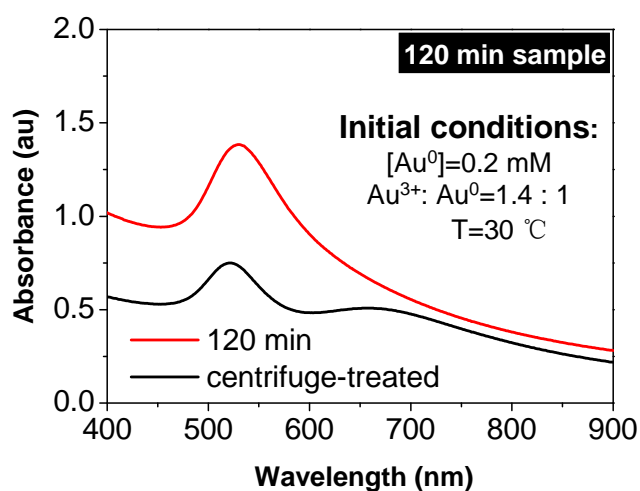
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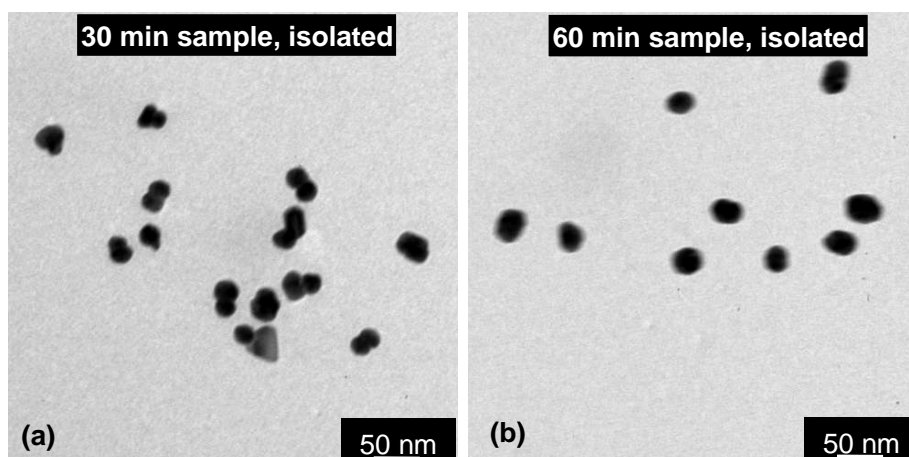
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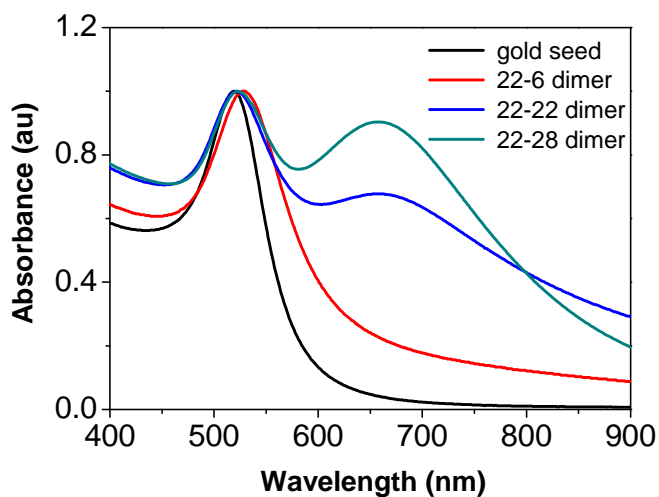
**Fig. S1** (left): TEM image of dot-shaped seeds of gold nanocrystals by the citrate reduction approach. (right): HRTEM image of a representative seed nanocrystal.



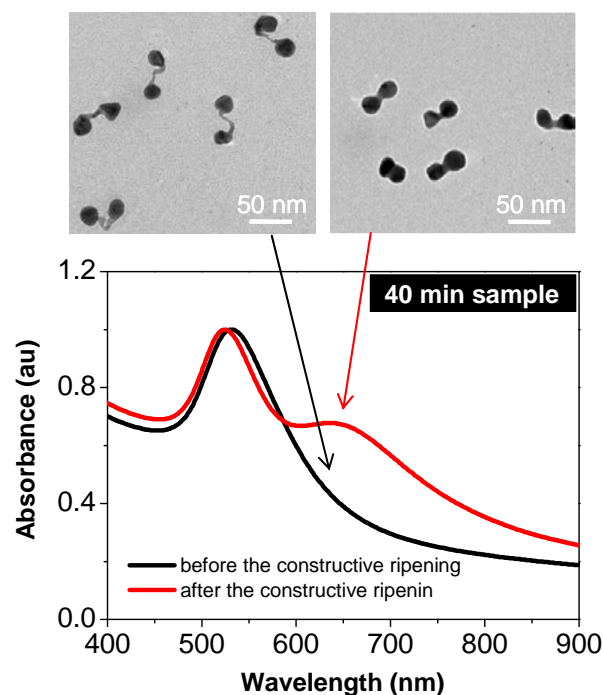
**Fig. S2** UV-Vis absorption spectra of the isolated sample taken at 120 min for a reaction with Au<sup>3+</sup>: Au<sup>0</sup> ratio being 1.4:1. For this specific reaction, the reduction rate was slower than that of the typical reaction. It took about 120 minutes to complete, and the sample shown here was the final product. As mentioned in the text, the reaction time for the typical reaction was about 60 minutes to consume nearly all precursor (with less than 10% precursor left, see Figure 2 (top)). The precursor concentration in this reaction was two times lower than that of the typical reaction. This would result in a slower reaction rate as well as slower intra-particle ripening rate due to lower Cl<sup>-</sup> ion concentration.



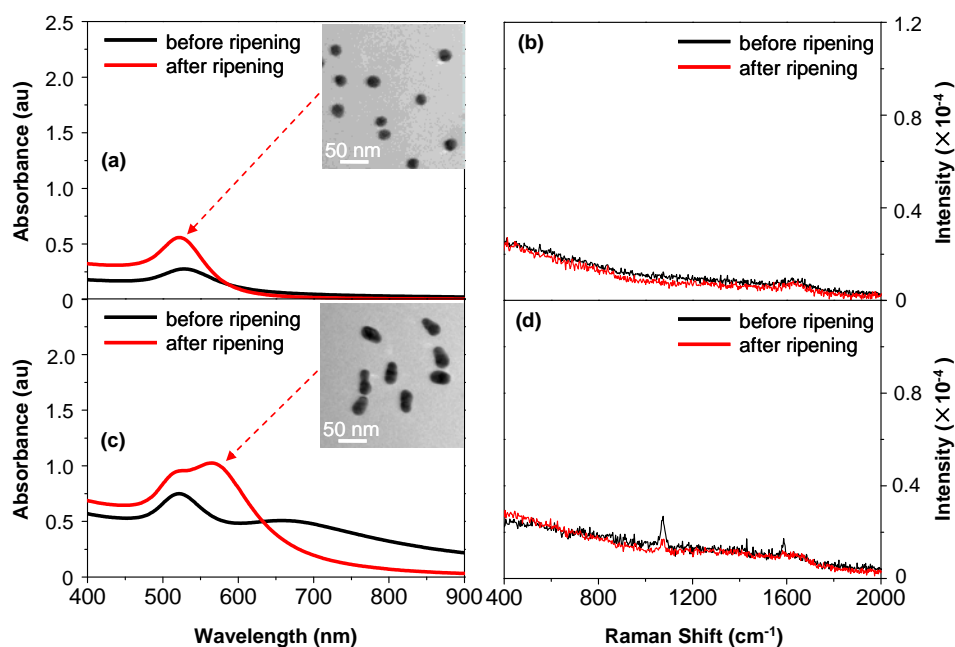
**Fig. S3** TEM images of the isolated samples taken at different time intervals for the typical reaction ( $Au^{3+}:Au^0$  ratio of 2.8:1) but without addition of NaOH. The nanostructures in the 30 min sample were mostly extremely tightly fused dimers and short rods, and the 60 min sample was mostly composed of large dots.



**Fig. S4** UV-Vis absorption spectra recorded for three types of dimers and the seeds. Different from Figure 7, the UV-Vis spectra in this plot are normalized at the main peak position. The absorption increase between 600 and 900 nm for “22-6 dimer” sample is clearly evidenced in this plot.



**Fig. S5** UV-Vis absorption spectra of the dimers for the homo-dimers with different intra-particle distances.



**Fig. S6** Morphology-dependent optical properties of smaller gold nanostructures. The TEM pictures in each plot were for the nanostructures upon “destructive intra-particle ripening”.