Electronic Supplementary Information

Facile synthesis of hydrangea flower-like hierarchical gold nanostructures with tunable surface topographies for single-particle surface-enhanced Raman scattering

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Stability. The stability of as-synthesized hydrangea flower-like hierarchical gold nanostructures were characterized by monitoring both the absorption spectra and morphologies of the fresh products and the ones stored at 4 °C after one and three months, respectively. The SEM and optical characterization results are shown in Fig. S1a-d, from which no distinct changes of the optical and morphological features can be observed. This demonstrates that the as-prepared hydrangea flower-like gold nanostructures are stable for at least three months at 4 °C. We also stored the as-prepared Au HFs at room temperature (Fig. S1e) and in a 40 °C water bath (Fig. S1f) for 1 month, the SEM images show that the particles were stable without obvious changes of morphology.

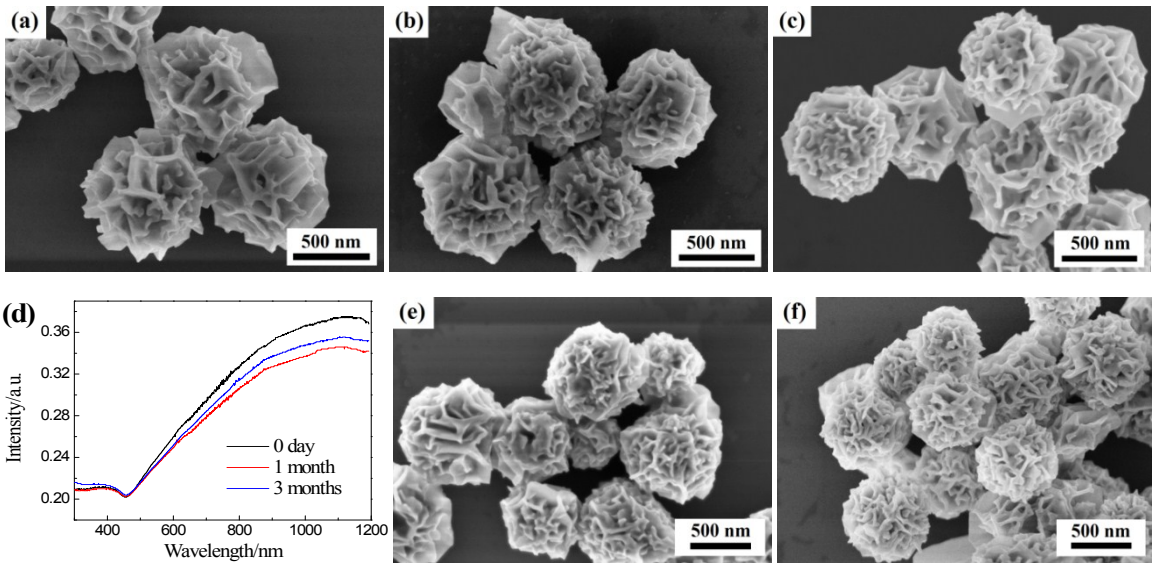

Fig. S1 Stability of the Au HFs. (a)-(c) SEM images of the Au HFs stored at 4 °C for 0 day (i.e. fresh particles), 1 month, and 3 months, respectively. (b) Absorption spectra of the Au HFs stored at 4 °C for 0 day, 1 month, and 3 months. (e)-(f) SEM images of the Au HFs stored at room temperature and 40 °C water bath for 1 month, respectively.

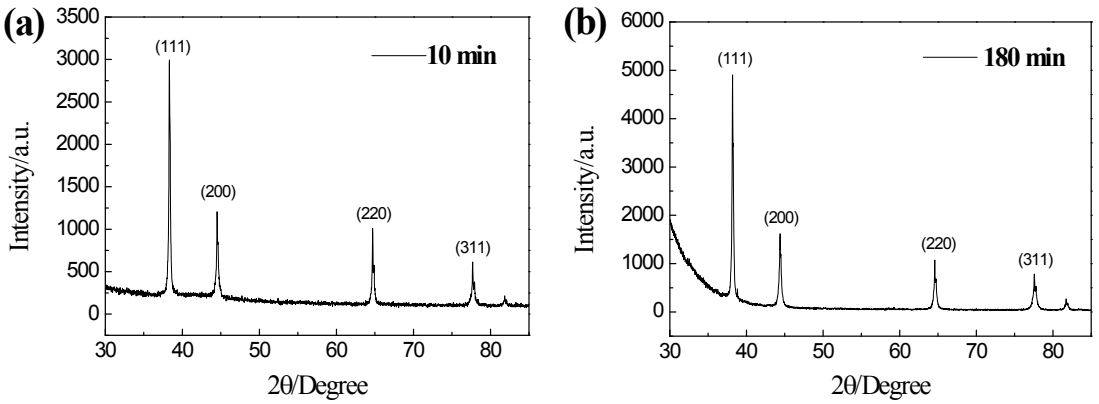

Fig. S2 XRD patterns of gold nanocrystals after synthesis for (a)10 min and (b) 180 min.

Table S1 XRD results

	$I_{(111)}$:$I_{(200)}$	$I_{(111)}$:$I_{(220)}$	$I_{(111)}$:$I_{(311)}$
Au nanocrystal (10 min)	2.76	3.04	5.26
Au nanocrystal (180 min)	3.00	4.57	6.10

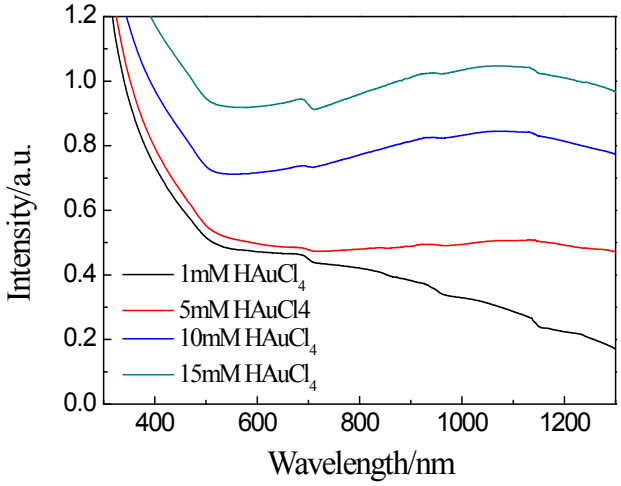

Fig. S3 SPR characterizations of Au HFs produced by changing the gold precursor concentration, 1 mM, 5 mM, 10 mM, and 15 mM.

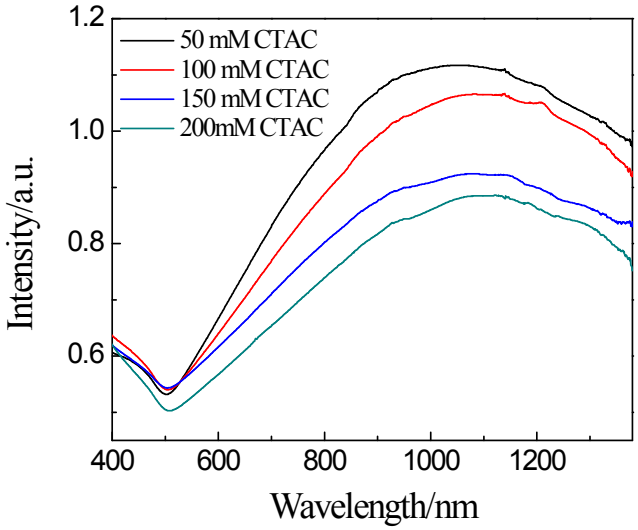

Fig. S4 SPR characterizations of Au HFs produced by changing the CTAC concentration, 50 mM, 100 mM, 150 mM, and 200 mM.

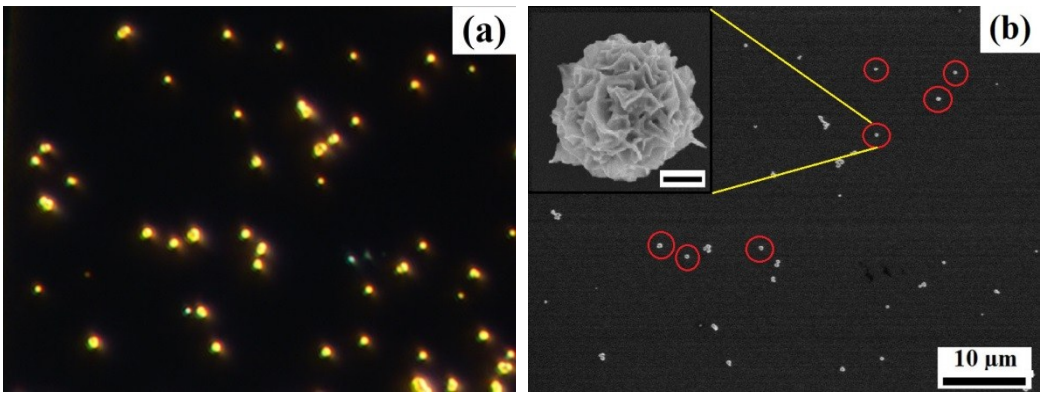

Fig. S5 (a) A dark-field optical microscopic image of HF4 on ITO slide. (b) The corresponding in-situ SEM image. The scale bars in the inset represent 200 nm.

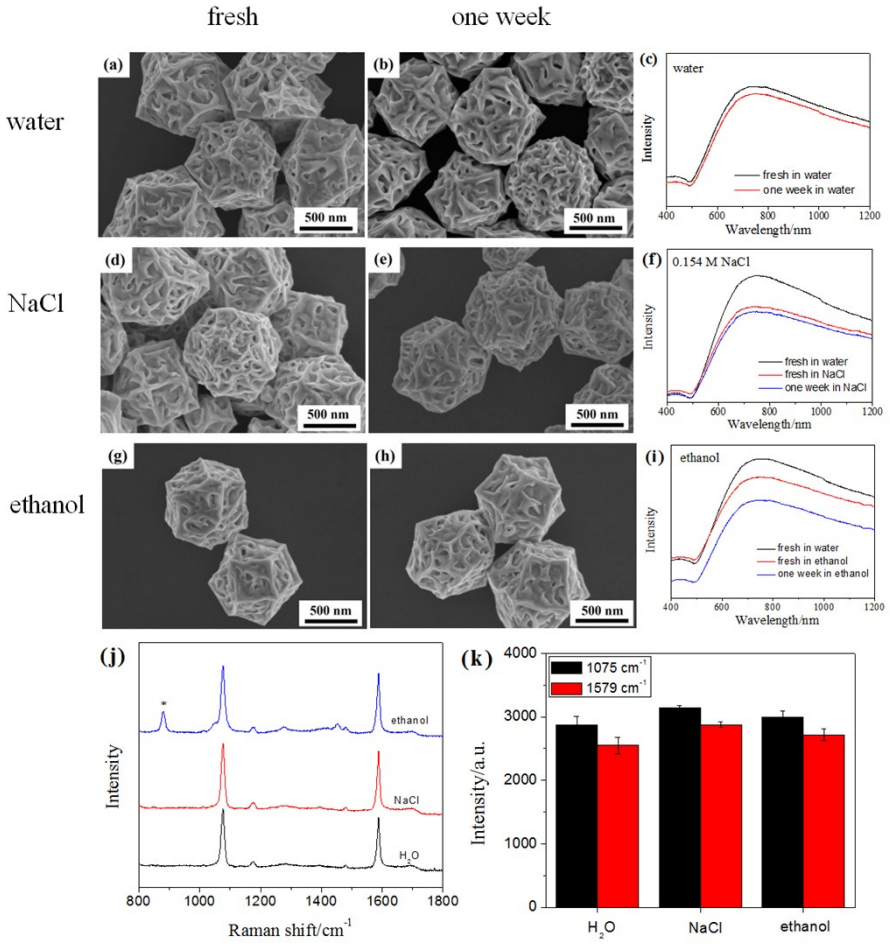

Fig. S6 Stability of the 20 °C synthesized gold nanoparticles dispersed in three different dispersants, (a)-(c) deionized water, (d)-(f) water containing 0.154 M NaCl, and (g)-(i) ethanol. (j)-(k) SERS spectra of 4-MBA collected from the gold colloid in water, 0.154 M NaCl solution, and ethanol, respectively. The absorption spectra and SEM images of the fresh samples and the samples stored at 4 °C for one week were tested. The asterisk marked peak in (j) belongs to ethanol.

Note: Three aliquots of Au nanoparticles were synthesized at 20 °C, followed by centrifugal purification. Subsequently, the three precipitates were redispersed in 1.5 mL deionized water, ethanol and 0.154 M NaCl aqueous solution, respectively. Then the absorption spectra and SEM images of the three fresh samples were recorded immediately. After being stored at 4 °C for one week, the three samples were tested again. The characterization results were shown in Fig. S6. When the medium was changed from water to NaCl aqueous solution or ethanol, only slight changes can be found on the absorption spectra. One week later, for each sample almost no obvious morphology changes or SPR peak shifts can be seen from the SEM images and absorption spectra. Additionally, it should be noted that no visible aggregates can be observed in the three colloids. The SERS spectra of 4-MBA in the three gold colloids dispersed in water, ethanol and 0.154 M NaCl aqueous solution respectively were measured and the results were shown in Fig. S6 (j)-(k). The strongest signal was obtained on the gold colloid in NaCl solution and the weakest signal was collected from the water dispersed gold nanoparticles. However, the difference of intensity between the strongest and weakest SERS signals is very small (less than 9%), which means the synthesized nanoparticles have good stability of SERS properties in different dispersants.