

Supplementary Information

Nanoarray-based Biomolecular Detection Using Individual Au-Nanoparticles with Minimized LSPR Variations

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Supplementary Information 1:

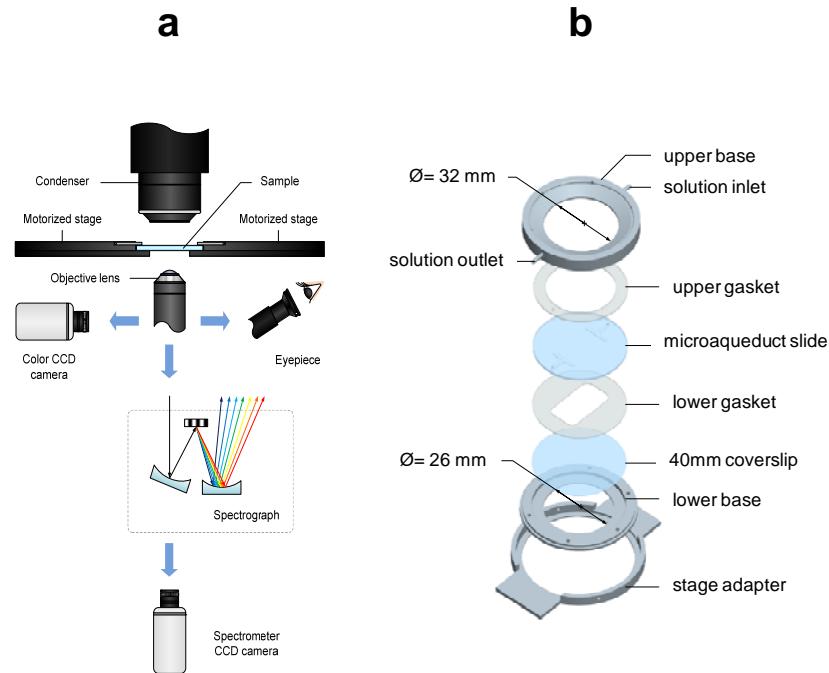


Figure S1: Schematic diagram of the darkfield system (a) and the homemade flow cell (b).

Supplementary Information 2: Spectral resolution of the system

The imaging spectrograph employed two gratings of 300 lines/mm and 1200 lines/mm with spectral resolutions of ~ 0.945 and ~ 0.162 nm, respectively (calculated based on our PIXIS 100F CCD camera and a central wavelength of 750 nm). Although superior resolution (almost one order of magnitude better) could be obtained by using the 1200 lines/mm grating, the wavelength coverage using this grating was only 87 nm as opposed to 506 nm for the 300 lines/mm grating (Figure S2). Considering the broad peak-wavelength distributions resulting from polydispersion of the synthesized nanoparticles (Figure 3b), it was necessary to pre-scan the user-defined area with the 300 lines/mm grating to choose a suitable central wavelength for the 1200 lines/mm grating. In addition, higher line number gratings (e.g., 2400 lines/mm) could further improve the spectral resolution, which was useful for LSPR-shift measurements. However, because the typical full width at half-maximum (FWHM) of a single AuNR was $\sim 30\text{-}60$ nm, gratings with line numbers higher than 1200 are not recommended. In order to explore the reproducibility of the system for single AuNR detection, 15 replicates of the same AuNR were collected on the two gratings (Figure S3). The resulting standard deviation (σ) of the peak wavelengths was 0.22 nm for the 1200 lines/mm grating, and 1.02 nm for the 300 lines/mm grating.

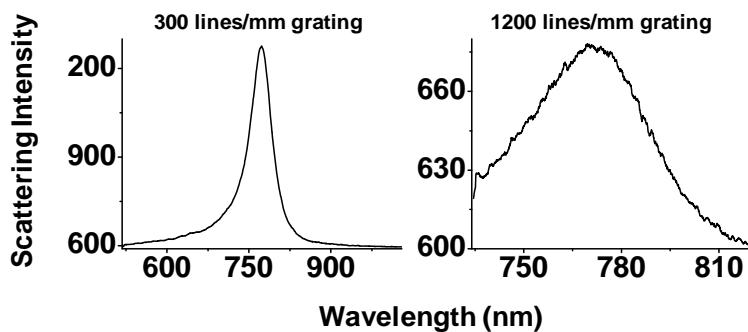


Figure S2: Typical AuNR scattering spectra obtained from the two gratings.

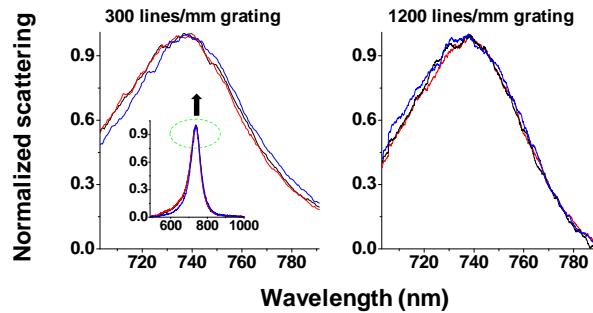


Figure S3: LSPR scattering spectra of an AuNR obtained on 300 and 1200 lines/mm gratings. Three of the 15 spectra obtained for the same Au-nanorod are shown.

Supplementary Information 3:

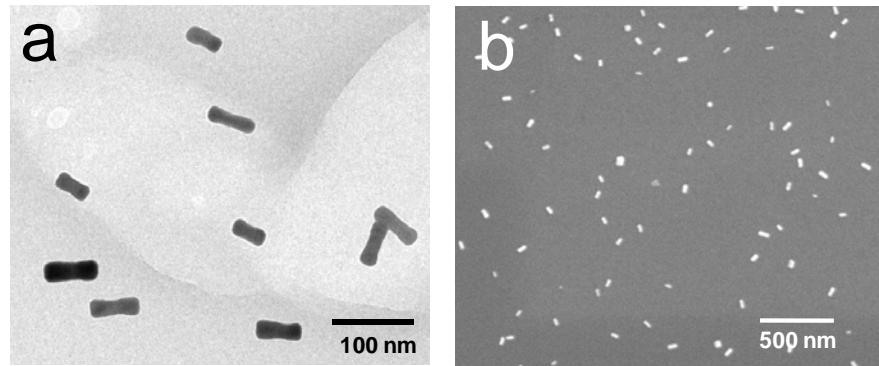


Figure S4: TEM (a) and SEM (b) of AuNRs used in this work.

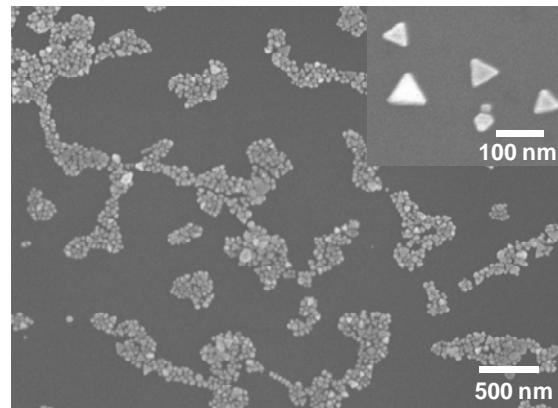


Figure S5 SEM of triangular Au nanoplates used in this work.

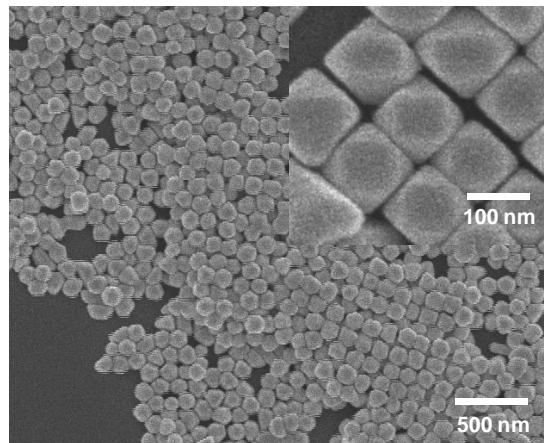


Figure S5 SEM of truncated Au nanoctahedrons used in this work.

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Table S1 LSPR responses for triangular Au nanoplates and truncated Au nanoctahedrons before and after normalization*

particle number	Triangular Au nanoplates				truncated Au nanoctahedron			
	peak wavelength		LSPR response		peak wavelength		LSPR response	
	$\lambda_{\max(1)}$	$\lambda_{\max(2)}$	$\Delta\lambda_{\max}$	ΔN	$\lambda_{\max(1)}$	$\lambda_{\max(2)}$	$\Delta\lambda_{\max}$	ΔN
1	636.49	641.16	4.67	0.03091	638.16	642.24	4.08	0.02656
2	590.47	592.91	2.44	0.03725	634.68	638.92	4.24	0.02876
3	649.73	654.97	5.24	0.02983	640.86	645.74	4.88	0.03065
4	650.5	655.35	4.85	0.02745	625.14	628.33	3.19	0.0247
5	635.45	639.49	4.04	0.02718	633.87	638.53	4.66	0.03184
6	666.28	672.31	6.03	0.02921	641.38	646.82	5.44	0.03385
7	600.19	602.93	2.74	0.03284	637.38	641.24	3.86	0.0254
8	666.4	672.36	5.96	0.02885	635.84	639.93	4.09	0.02738
9	701.73	710.17	8.44	0.03092	637.77	641.86	4.09	0.02675
10	649.73	654.88	5.15	0.02933	644.33	649.19	4.86	0.02936
11	705.72	713.7	7.98	0.02852	640.86	645.49	4.63	0.02912
12	649.83	654.78	4.95	0.02819	637.38	642.33	4.95	0.03236
13	645.49	650.35	4.86	0.02899	628.77	633.26	4.49	0.03279
14	644.33	649.58	5.25	0.03165	639.46	644.11	4.65	0.02972
15	647.03	651.97	4.94	0.02897	633.62	638.2	4.58	0.03141
16	635.45	639.63	4.18	0.0281	641.22	645.56	4.34	0.02723
17	666.66	672.64	5.98	0.02888	619.34	622.6	3.26	0.02747
18	631.97	635.95	3.98	0.02797	635.5	639.59	4.09	0.02749
19	643.56	648.89	5.33	0.03239	631.84	635.24	3.4	0.02403
20	634.68	639.03	4.35	0.02949	628.22	632.5	4.28	0.03153
21	660.51	666.89	6.38	0.0325	646.88	652.37	5.49	0.03216
22	652.42	657.74	5.32	0.02945	637.12	641.89	4.77	0.03131
23	597.69	600.74	3.05	0.03852	636.53	640.61	4.08	0.02708
24	655.12	660.97	5.85	0.03145	632.17	636.29	4.12	0.02886
25	640.09	644.72	4.63	0.02938	641.68	646.7	5.02	0.03121
26	642.79	647.19	4.4	0.02711	635.2	639.88	4.68	0.03146
27	637.14	641.51	4.37	0.02875	647.28	652.76	5.48	0.03197
28	653.28	658.76	5.48	0.03006	645.12	649.95	4.83	0.02894
29	596.37	598.93	2.56	0.03354	632.8	636.4	3.6	0.0251
30	645.92	650.83	4.91	0.02915	634.67	638.83	4.16	0.02823
RSD (%)	\	\	27.16	8.853	\	\	13.68	9.105

* $\lambda_{\text{max}(1)}$: LSPR peak wavelength of AuNPs in water.
 $\lambda_{\text{max}(2)}$: LSPR peak wavelength of AuNPs in ethanol.
 $\Delta\lambda_{\text{max}}$: $\lambda_{\text{max}(2)} - \lambda_{\text{max}(1)}$.
 ΔN : LSPR responses normalized according to equation (2).

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Table S2: LSPR wavelength shifts and corresponding normalized responses for the 30 particles shown in Figure 4*

Particle number	$(\lambda_{\max(1)} + \lambda_{\max(2)})/2$ (nm)	$\Delta\lambda_{\max}$ (nm)	ΔN
1	708	3.77	0.01361
2	804	6.51	0.01442
3	838	7.55	0.01471
4	753	4.96	0.01383
5	697	3.47	0.01350
6	773	5.87	0.01486
7	689	3.27	0.01348
8	722	4.08	0.01349
9	739	4.63	0.01389
10	716	4.17	0.01430
11	763	5.29	0.01404
12	781	5.41	0.01321
13	694	3.88	0.01542
14	683	3.46	0.01494
15	756	4.98	0.01367
16	793	5.94	0.01377
17	706	3.86	0.01412
18	711	3.89	0.01377
19	726	4.28	0.01382
20	733	4.45	0.01380
21	709	3.82	0.01370
22	719	4.15	0.01397
23	722	4.37	0.01445
24	735	4.44	0.01362
25	661	3.03	0.01581
26	738	4.66	0.01406
27	744	4.77	0.01393
28	807	6.23	0.01364
29	715	4.02	0.01388
30	753	5.01	0.01397

* $\lambda_{\max(1)}$: LSPR peak wavelength before thrombin binding.

$\lambda_{\max(2)}$: LSPR peak wavelength after thrombin binding.

$\Delta\lambda_{\max}$: $\lambda_{\max(2)} - \lambda_{\max(1)}$.

ΔN : LSPR responses normalized according to equation (2).