## Supporting Information

## Concave gold bipyramids bounded with multiple high-index facets:

## improved Raman and catalytic activities

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Tab. S1 Average lengths and widths of AuBPs (LWP = 749 nm) and corresponding regrown CAuBPs.

Samula	Volume of seeds	Length	Width
Sample	(µL)	(nm)	(nm)
AuBPs	/	$70\pm3$	24±1
Concave AuBPs-1	50	560±17	99±6
Concave AuBPs-2	100	452±14	80±3
Concave AuBPs-3	200	319±9	64±2
Concave AuBPs-4	400	229±8	52±3
Concave AuBPs-5	500	210±6	44±1
Concave AuBPs-6	600	197±4	43±1
Concave AuBPs-7	700	119±3	42±1

Sample	Volume of seeds (µL)	Length (nm)	Width (nm)
AuBPs	/	$108\pm3.2$	26±1.6
Concave AuBPs-1	50	448±3	81±4.5
Concave AuBPs-2	100	432±15	83±5
Concave AuBPs-3	200	353±6.5	66±5.5
Concave AuBPs-4	400	256±10	49±4.5

Tab.S2 Average lengths and widths of AuBPs (LWP=917 nm) and corresponding regrown CAuBPs

Tab. S3 Average lengths and widths of AuBPs (LWP=1045 nm) and corresponding regrown CAuBPs

Samula	Volume of seeds	Length	Width
Sample	(µL)	(nm)	(nm)
AuBPs	/	$148\pm6.5$	37±2.3
Concave AuBPs-1	50	518±14	105±5
Concave AuBPs-2	100	491±5	93±4
Concave AuBPs-3	200	424±13	76±3
Concave AuBPs-4	400	324±16	61±2

Tab. S4 Atomic ratio of Ag/Au on the surfaces of the samples at different overgrowth time.

Name	AuBPs	After 1h reaction	After 2h reaction	After 4h reaction
Au atomic	16.1	33.0	26.5	19.8
Ag atomic	83.9	77.0	73.5	80.2
Ag/Au ratio	0.191	0.45	0.361	0.247

Tab. S5 Atomic ratio of Ag/Au on the surfaces of concave AuBPs for single-partilce SERS activities.

Name	Concave	Concave	Concave	Concave
	AuBPs-1	AuBPs-2	AuBPs-3	AuBPs-4
Au atomic	65.68	68.51	71.71	73.86
Ag atomic	34.32	31.49	28.29	26.14
Ag/Au ratio	0.52	0.46	0.39	0.35



Fig. S1 TEM image of AuBPs (LPW = 749 nm).



Fig. S2 SEM images of AuBPs with a LPW of 749 nm (a) and corresponding CAuBPs regrown from different amount of AuBPs as seeds (b-h): (b) 50  $\mu$ L. (c) 100  $\mu$ L. (d) 200  $\mu$ L. (e) 400  $\mu$ L. (f) 500  $\mu$ L. (g) 600  $\mu$ L. (h) 700  $\mu$ L. (i)The corresponding extinction spectra of the AuBPs and CAuBPs.



Fig. S3 SEM images of AuBPs with a LPW of 917 nm (a) and corresponding CAuBPs regrown from different amount of AuBPs as seeds (b-e): (b) 50  $\mu$ L. (c) 100  $\mu$ L. (d) 200  $\mu$ L. (e) 400  $\mu$ L. (f) The corresponding extinction spectra of the AuBPs and CAuBPs.



Fig. S4 SEM images of AuBPs with a LPW of 1045 nm (a) and corresponding CAuBPs regrown from different amount of AuBPs as seeds (b-e): (b) 50  $\mu$ L. (c) 100  $\mu$ L. (d) 200  $\mu$ L. (e) 400  $\mu$ L. (f) The corresponding extinction spectra of the AuBPs and CAuBPs.



Fig. S5 XRD pattern of CAuBPs.



Fig. S6 XPS spectra of CAuBPs.



Fig. S7 SEM images of as-synthesized nanoparticles with different amount of AgNO<sub>3</sub>. (a) 0  $\mu$ L , (b) 2  $\mu$ L, (c) 10  $\mu$ L, (d) 20  $\mu$ L, (e) 40  $\mu$ L, (f) 80  $\mu$ L.



Fig. S8 XPS spectra of the samples overgrowth with different  $Ag^+$  concentration.a) AuBPs as seeds. b) overgrowth with 10 µl AgNO<sub>3</sub>. c) overgrowth with 20 µl AgNO<sub>3</sub>. d) overgrowth with 40 µl AgNO<sub>3</sub>.



Fig. S9 SEM images of as-synthesized nanoparticles with different surfactants. (a) CTAC. (b) CTAB+CTBAB. (c) CTAC+CTBAB.



Fig. S10 a)The simulated and experimental extinction spectra of AuBPs and Concave AuBPs with similar sizes. b) TEM images of AuBPs (left, length =  $282 \pm 5$  nm, width =  $52 \pm 2$ ) and Concave AuBPs (right, length =  $281 \pm 6$  nm, width =  $51 \pm 3$ ) for experimental extinction spectra.



Fig. S11 (a) TEM images of AuBPs (length =  $282 \pm 5$  nm, width =  $52 \pm 2$  nm) and CAuBPs (length =  $285 \pm 13$  nm, width =  $59 \pm 3$ ). (b) The corresponding extinction spectra.



Fig. S12 Optical, SEM and corresponding overlap images of different concave AuBPs at the length of  $285 \pm 13$  nm,  $410 \pm 10$  nm,  $577 \pm 13$  nm and  $748 \pm 14$  nm, respectively. Scale bar = 500 nm.