

## Synthesis of AuAg/Ag/Au open nanoshells with optimized magnetic plasmon resonance and broken symmetry for enhancing second-harmonic generation†

Tao Zhou,<sup>a</sup> Si-Jing Ding,<sup>\*a</sup> Zhi-Yong Wu,<sup>c</sup> Da-Jie Yang,<sup>d</sup> Li-Na Zhou,<sup>a</sup> Zhi-Rui Zhao,<sup>a</sup> Liang Ma,<sup>\*b</sup> Wei Wang,<sup>c</sup> Song Ma,<sup>c</sup> Si-Man Wang,<sup>c</sup> Jia-Nan Zou,<sup>c</sup> Li Zhou,<sup>c</sup> Qu-Quan Wang,<sup>\*c</sup>

<sup>a</sup>School of Mathematics and Physics, China University of Geosciences (Wuhan), Wuhan 430074, China.

E-mail: dingsijing@cug.edu.cn

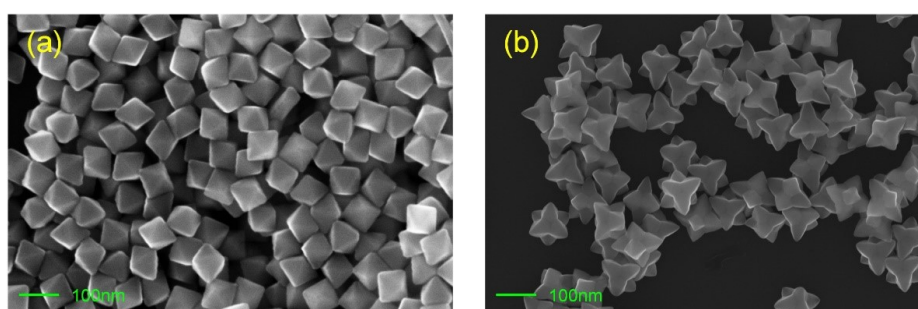
<sup>b</sup>Hubei Key Laboratory of Optical Information and Pattern Recognition, Wuhan Institute of Technology, Wuhan 430205, China.

E-mail: maliang@wit.edu.cn

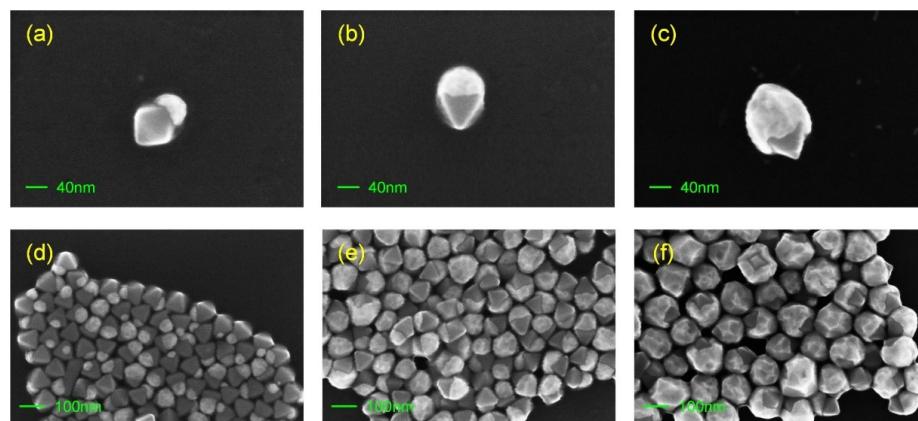
<sup>c</sup>Department of Physics, Key Laboratory of Artificial Micro- and Nano-structures of the Ministry of Education, Wuhan University, Wuhan 430072, China.

E-mail: qqwang@whu.edu.cn

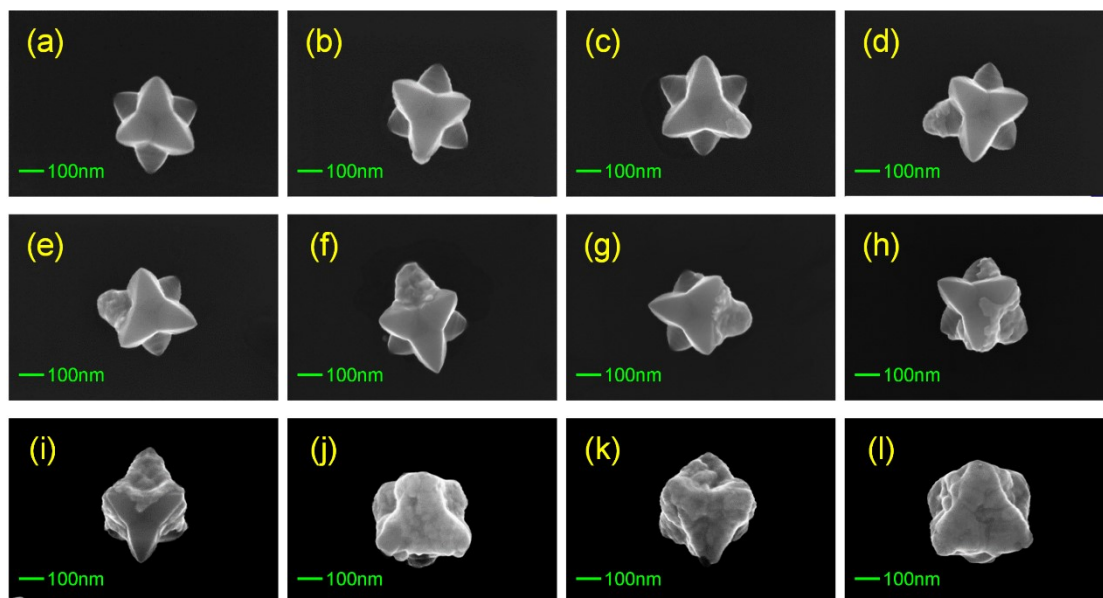
<sup>d</sup>Mathematics and Physics Department, North China Electric Power University, Beijing 102206, China.



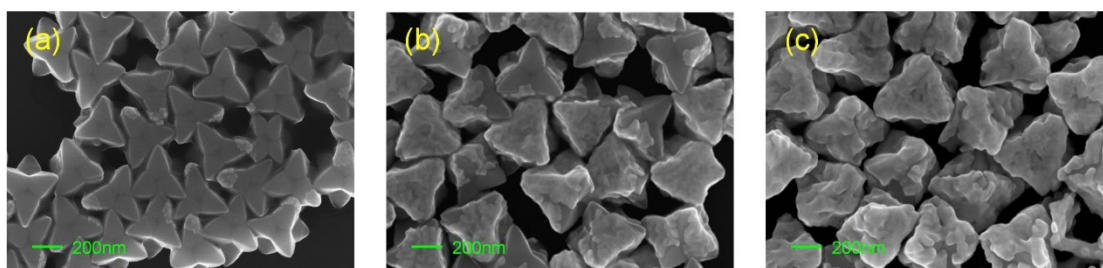
**Figure S1.** SEM images of PbS templates. (a) PbS nanooctahedron with an average side length of  $66.5 \pm 2.5$  nm. (b) PbS nanostar with an average side length of  $316.2 \pm 8.8$  nm.



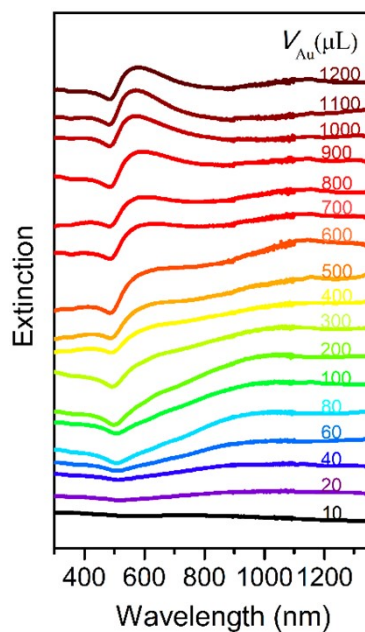
**Figure S2.** SEM images of Au nanocups grown on PbS nanooctahedron. The corresponding amount of Au ( $V_{Au1}$ ) is 60  $\mu$ L (a, d), 200  $\mu$ L (b, e) and 800  $\mu$ L (c, f).



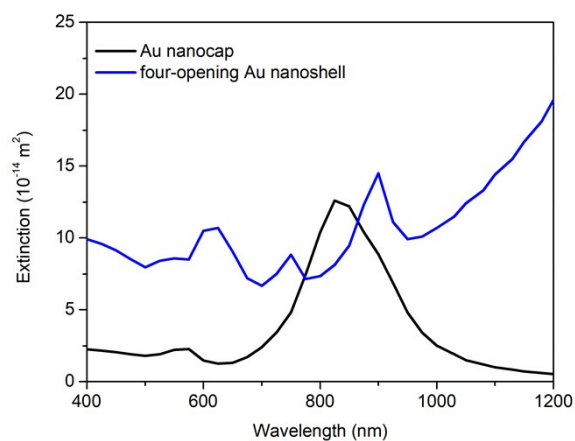
**Figure S3.** SEM images of Au nanoshell grown on a single PbS nanostar. The corresponding amount of Au ( $V_{Au1}$ ) is 0  $\mu\text{L}$  (a), 10  $\mu\text{L}$  (b), 40  $\mu\text{L}$  (c), 60  $\mu\text{L}$  (d), 80  $\mu\text{L}$  (e), 100  $\mu\text{L}$  (f), 200  $\mu\text{L}$  (g), 400  $\mu\text{L}$  (h), 500  $\mu\text{L}$  (i), 800  $\mu\text{L}$  (j), 900  $\mu\text{L}$  (k), and 1200  $\mu\text{L}$  (l).



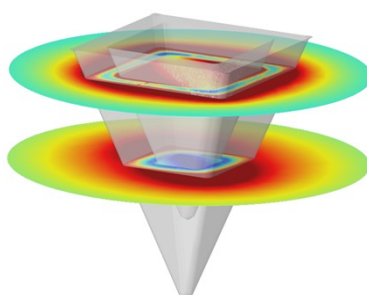
**Figure S4.** Large-scale SEM images of Au nanoshells grown on PbS nanostars overgrowth with Au open nanoshells. The corresponding amount of Au ( $V_{Au1}$ ) is 20  $\mu\text{L}$  (a), 500  $\mu\text{L}$  (b) and 900  $\mu\text{L}$  (c).



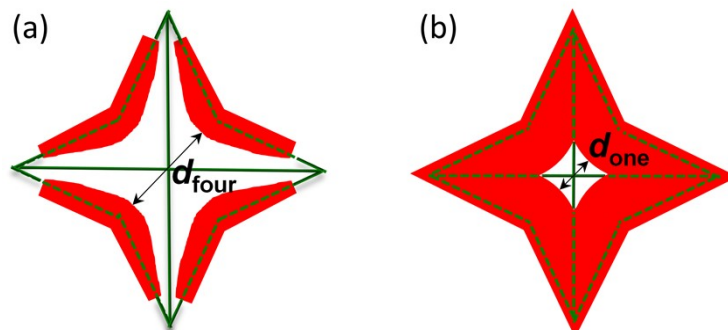
**Figure S5.** Extinction spectra of Au open nanoshells recorded after dissolving off PbS templates.



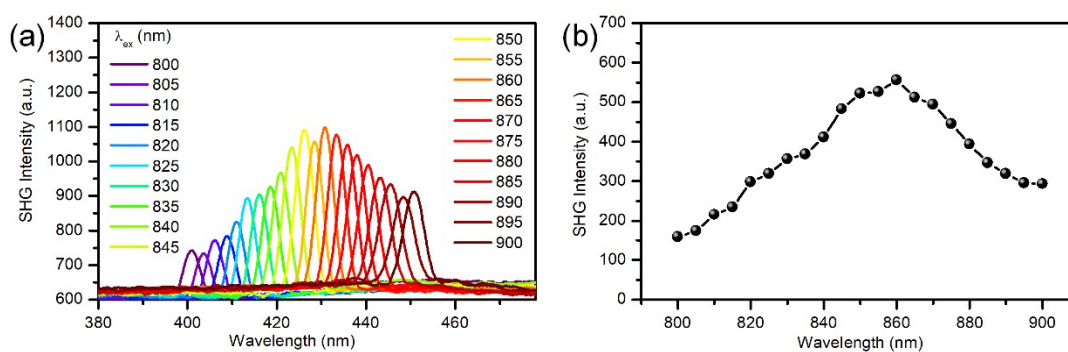
**Figure S6.** Calculated extinction cross sections of the Au nanocap and four-opening Au nanoshell.



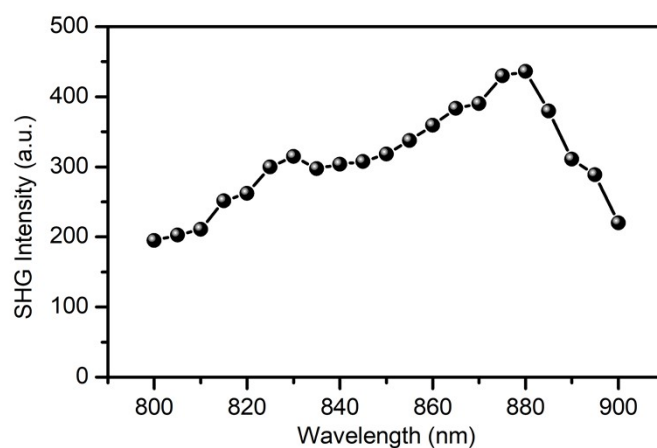
**Figure S7.** The magnetic field intensity around the nanocap. The magnetic field is mainly at the outer boundary of the nanocap.



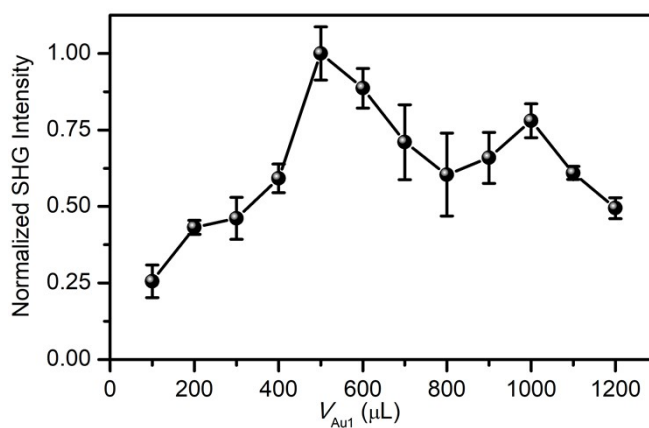
**Figure S8.** Bottom-view illustrations of the four-opening ( $V_{Au1} = 500 \mu\text{L}$ ,  $d_{\text{four}} = 185.6 \pm 26.8 \text{ nm}$ ) and one-opening ( $V_{Au1} = 900 \mu\text{L}$ ,  $d_{\text{one}} = 69.3 \pm 12.0 \text{ nm}$ ) Au nanoshells.



**Figure S9.** Morphology-dependent SHG of the Au open nanoshells. (a) SHG spectra of the Au open nanoshells ( $V_{Au1} = 1000 \mu\text{L}$ ) with varied excitation wavelength. (b) SHG excitation spectra with  $V_{Au1} = 1000 \mu\text{L}$ .



**Figure S10.** SHG excitation spectra of the Au nanocaps with  $V_{Au1} = 80 \mu\text{L}$ . The law of the SHG excitation spectra of the Au nanocaps are similar to those of the four-opening and one-opening Au nanoshells.



**Figure S11.** Normalized SHG intensity as a function of  $V_{Au1}$ . The excitation wavelength is 880 nm.

**Table S1.** The Au and Ag atomic content of Au and Ag/Au open nanoshells in the EDX measurement.

Samples	Atomic content	
	Au (At%)	Ag (At%)
Au open nanoshells	100	0
Ag/Au open nanoshells ( $V_{Ag} = 300 \mu\text{L}$ )	78.85	21.15