Supporting Information

Two-dimensional Gold Trisoctahedron Nanoparticle Superlattice Sheets: Self-assembly, Characterization and Immunosensing Applications

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1.Characterisation of TOH particles

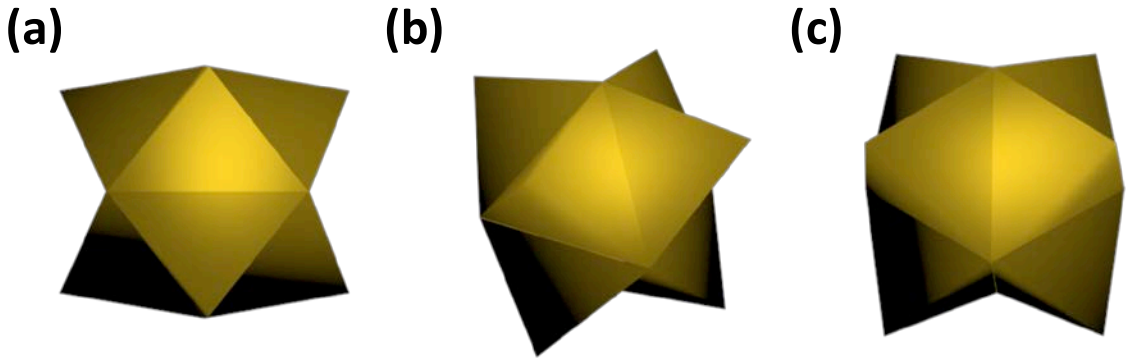

Fig.S1. Schematic drawing of TOH nanoparticles at different viewing directions (a-c)

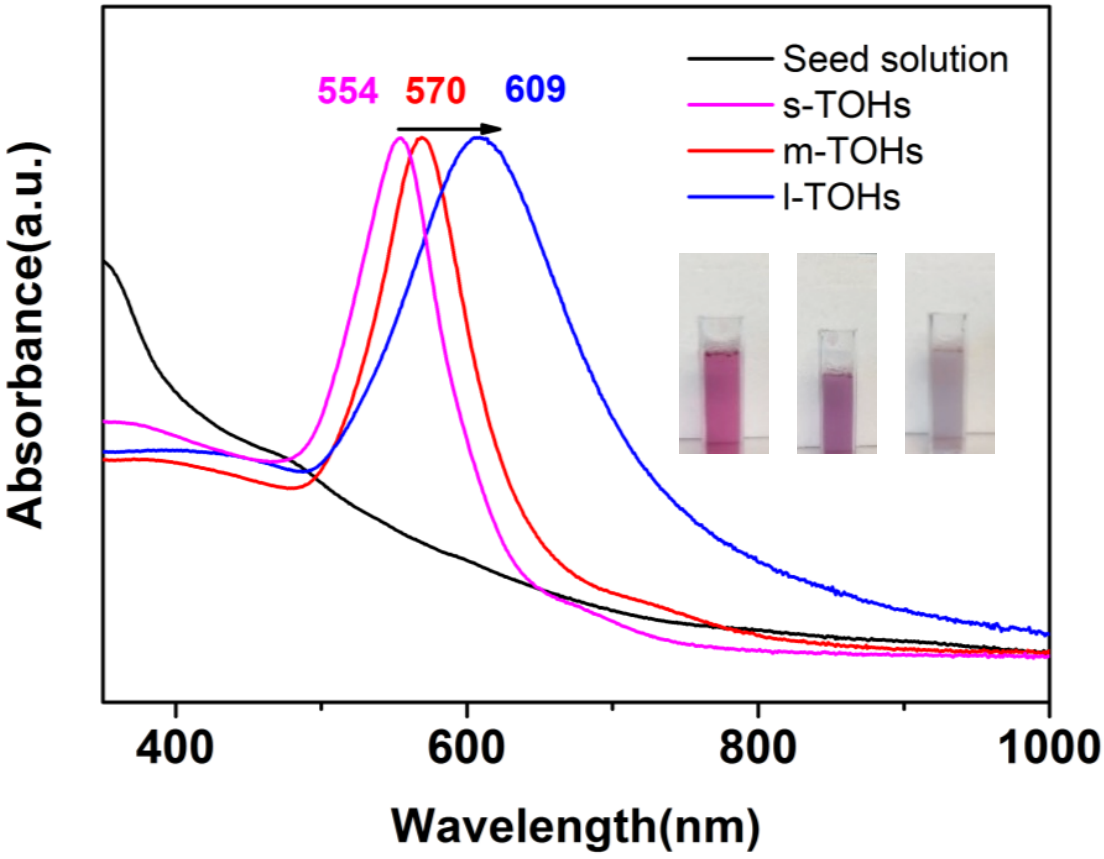

Fig. S2. UV-vis optical spectra of the as-synthesized Au TOH seeds (black line), small (red line), medium (pink) and large (blue line) Au TOH solutions. Insets are photos of small, medium and large Au TOH solutions.

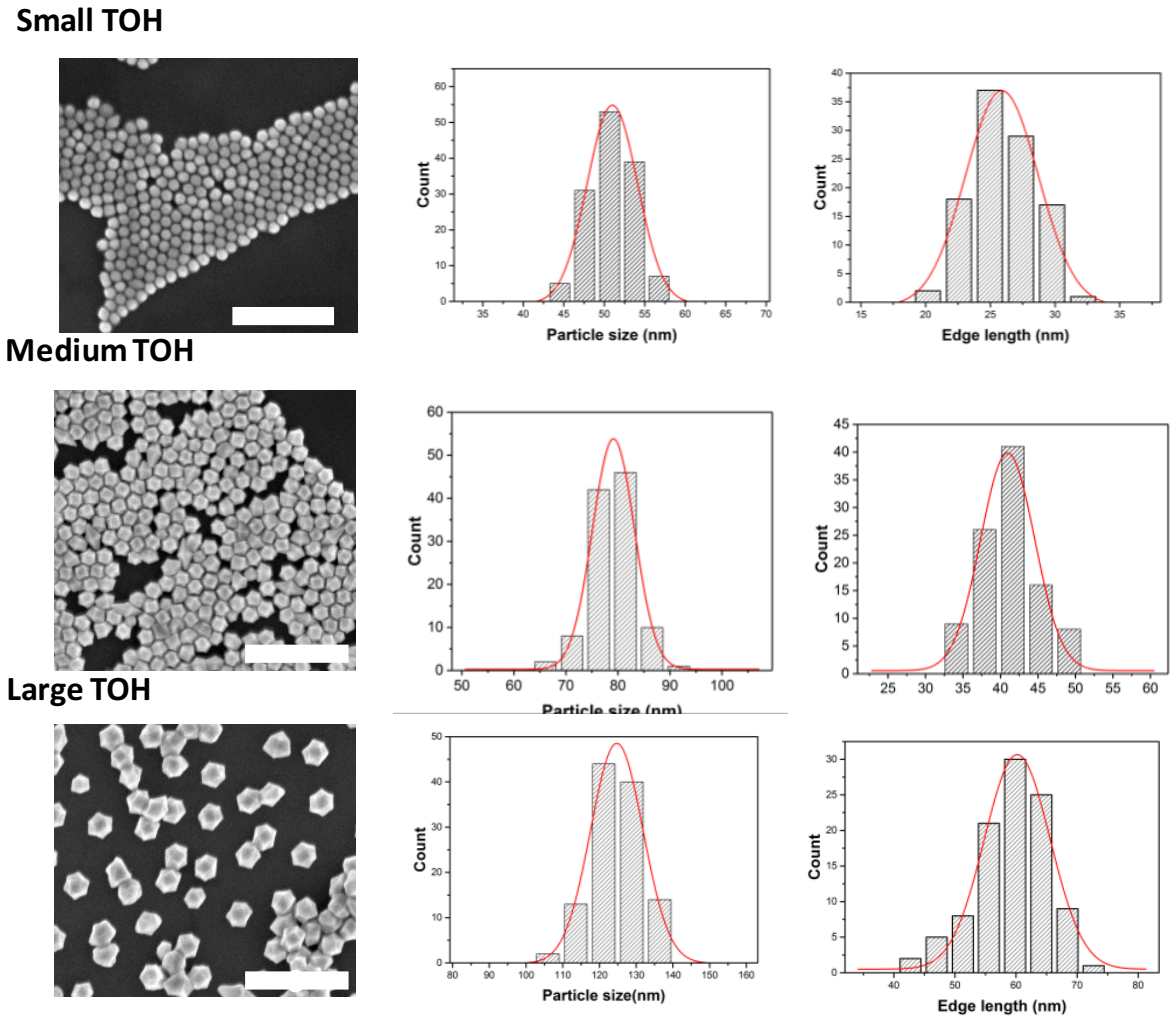

Fig. S3. SEM image of TOH particle and corresponding histogram of average particle diameters and apparent edge lengths for three sizes Au TOH. Red lines are the Gaussian fits of the size distributions. Scale bar is 500nm.

The number percentage of TOH with three different sizes was obtained by counting nanoparticles number on SEM images, which is over 96% for small TOH, 95% for medium TOH and 92% for large TOH.

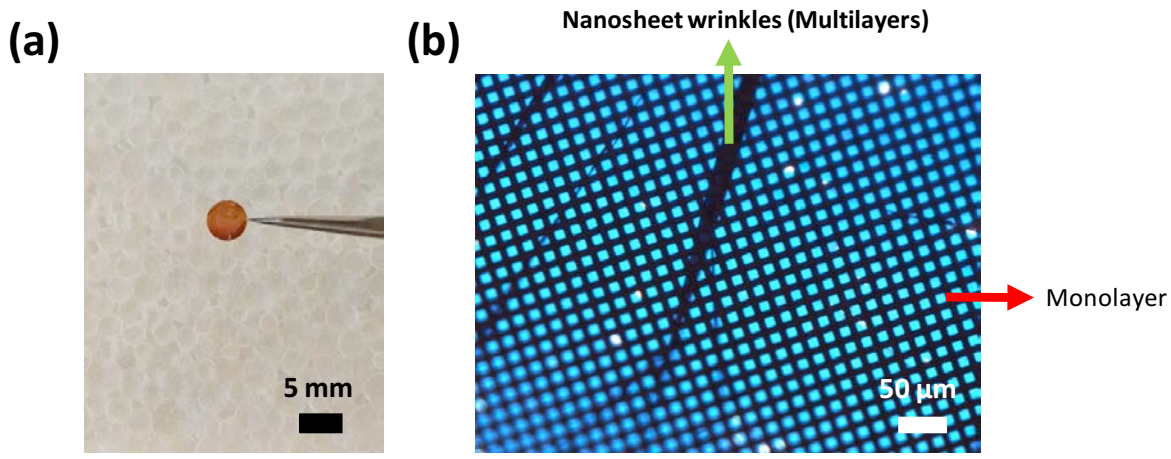

Fig. S4 (a) A photo of as fabricated free-standing TOH superlattice nanosheet covering on top of a holey copper grid. (b) Optical miscopy image of TOH nanosheet in transmission mode.

2. Maintaining structural integrity after plasma treatment

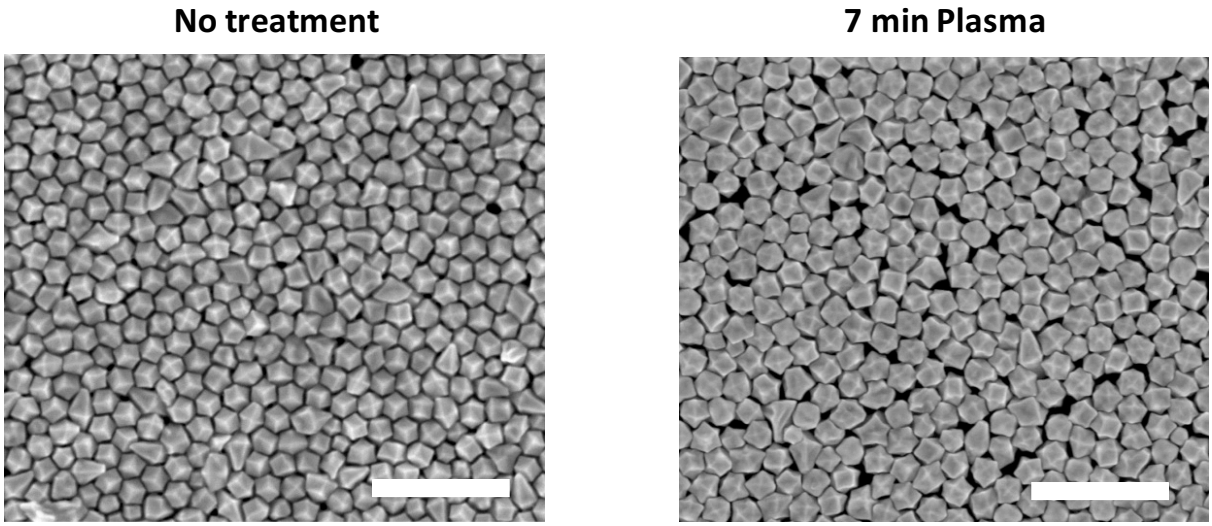

Fig. S5. SEM image of l-TOH (12k PS) superlattices before and after treatments. Scale bar is 500nm.

3. Comparison of TOH superlattice SERS with nanosphere superlattices

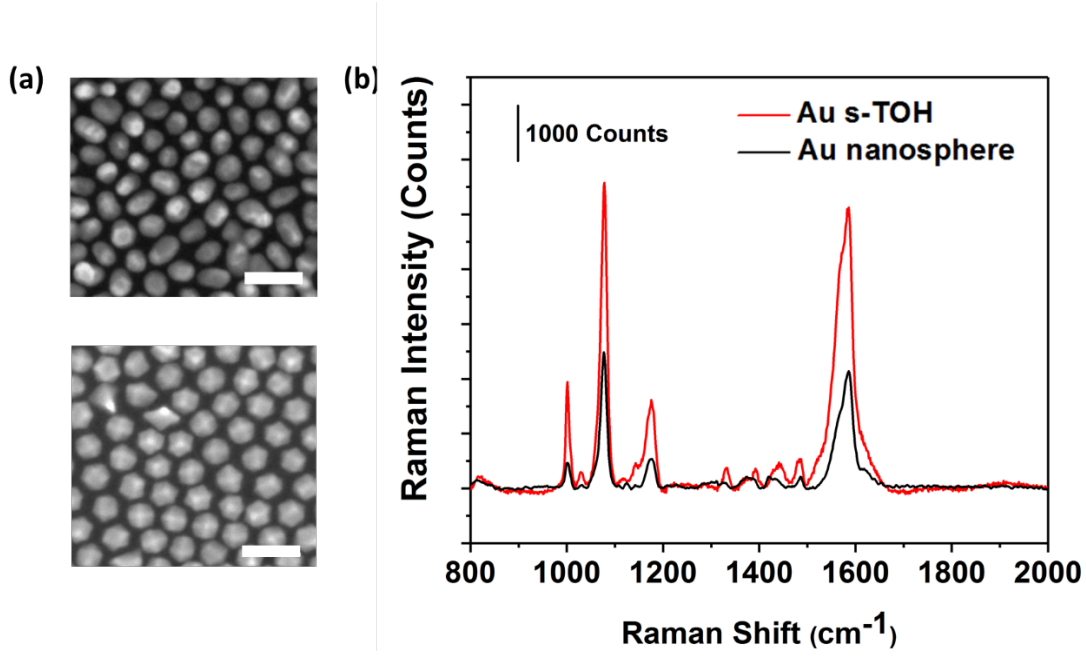

Fig. S6. (a) SEM image of gold nanosphere superlattices (top) and s-TOH superlattices (bottom). Scale bar is 100 nm. (b) SERS spectra of 4-ATP from gold nanosphere and s-TOH nanosheet.

4. Comparsion of TOH superlattice SERS performmance with TOH powder

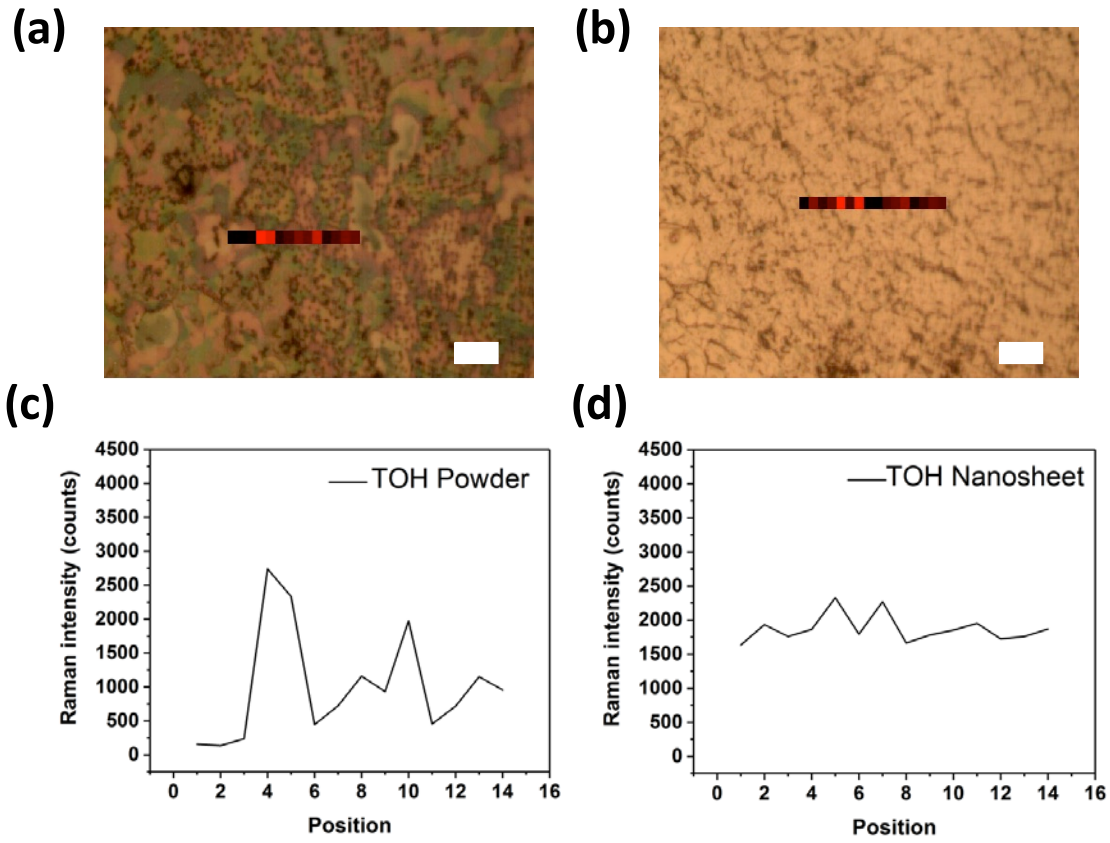

Fig. S7. (a) The optical microscopy image of TOH powder. (b) The optical microscopy image of TOH superlattices. The insets red color region are the Raman mapping region. Scale bar is 20 μm. (c) and (d) are the Raman signals at 1078 cm^{-1} collected from (a) and (b).

5. Calculation of SERS Enhancement Factor (EF)

The EF can be calculated by the classic equation[1]:

$$EF = \frac{I_{SERS} N_{BULK}}{I_{BULK} N_{SERS}},$$

where I_{BULK} and I_{SERS} are the peak intensity of bulk-4-ATP and 4-ATP on superlattice nanosheest at 1078 cm^{-1}; N_{BULK} and N_{SERS} are the number of 4-ATP molecules in a bulk sample and the superlattice nanosheet within the laser spot area. N_{SERS} can be calculated by $N_{SERS} =$

$\frac{N_d A_{laser} A_N}{\sigma}$. Here, N_d is the number density of particles in unit area, A_{laser} and A_N are the areas of focal laser spot and the nanosheet respectively. σ is the footprint size of 4ATP molecule, for which we used 0.20 nm^2 according to the literature[2]. For analysis, the 4-ATP Raman peak of 1078 cm^{-1} was chosen for all EF calculation.

6. Verstality of TOH superalttices immunosensing in freestading system.

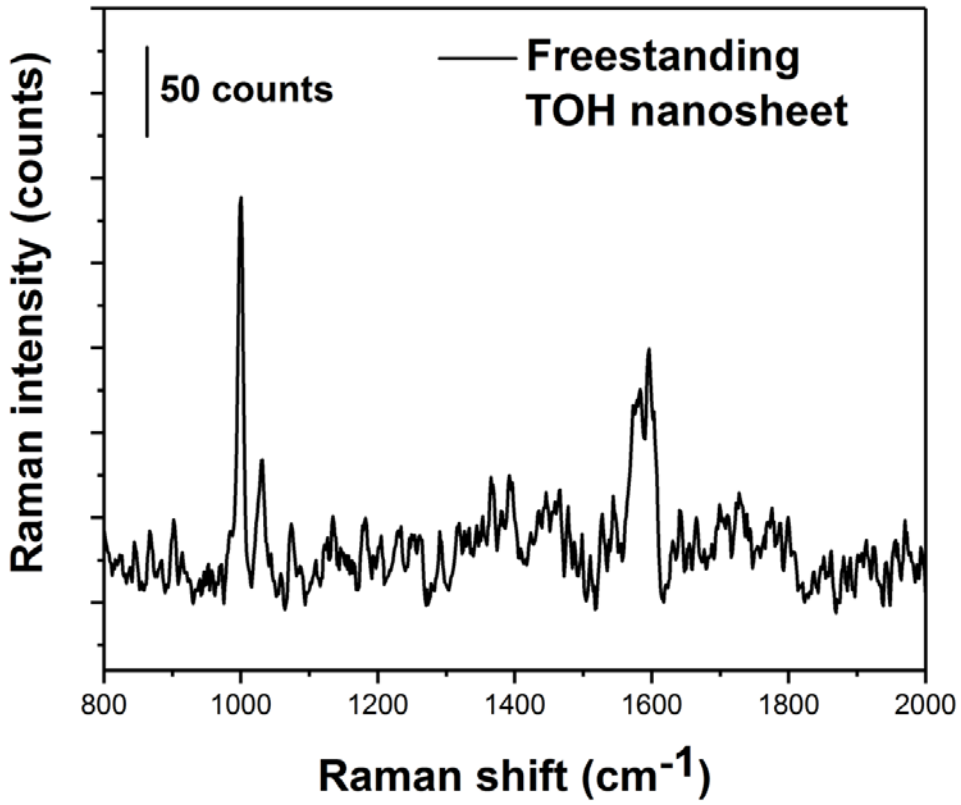

Fig.S8. Immunosensing result of freestanding TOH nanosheet by using 1 ng/ml rabbit IgG.

References

1. Y. Chen, K. J. Si, D. Sikdar, Y. Tang, M. Premaratne and W. Cheng, *Advanced Optical Materials*, 2015, **3**, 919-924.
2. K. Kim and J. K. Yoon, *The Journal of Physical Chemistry B*, 2005, **109**, 20731-20736.