Supporting Information

## Highly Sensitive and On-Site NO<sub>2</sub> SERS Sensors Operated under Ambient Conditions

## **Details of the methods**

## Optical properties of the 3D stacked Ag NWs

Extinction spectra were calculated as  $1 - R_{3D \text{ plasmonic nanostructures}}$  after measuring the diffuse reflectance from the 3D plasmonic substrate using UV–vis near-infrared (NIR) spectroscopy (Cary 5000, Agilent).<sup>1</sup>

**Gas-sensing measurements.** The gas-sensing test was conducted by exposing the SERS substrate to the NO<sub>2</sub> gas diluted with N<sub>2</sub> in a flow chamber for 3 min. The concentrations of the analyte gases were adjusted by modulating the flow rate ratio of both gases (N<sub>2</sub> and NO<sub>2</sub> gas). During the purge process, N<sub>2</sub> was supplied into the chamber at 5000 standard cubic centimeters per minute (sccm).



**Fig. S1.** Scanning electron microscopy (SEM) images of (a) the 3D stacked Ag nanowires (NWs) and 3D bimetallic (b) 5 nm, (c) 10 nm, (d) 15 nm, (e) 20 nm, (f) 25 nm thick Aucoated Ag NW structures.



**Fig. S2.** Energy dispersive X-ray spectroscopy (EDS) of the 3D bimetallic 15 nm thick Aucoated Ag NWs structures. (a) An SEM image of 3D Au-Ag bimetallic NW structures. (b) Elemental analysis of the sample shows an Ag of 88.10% and an Au of 11.90%. The EDS data clearly confirms the 3D Au-Ag bimetallic NW structures.



Fig. S3. Diameter distribution of the 3D Ag NWs (black) and Au-Ag NWs (red pattern).



Fig. S4. Extinction spectra of the 3D Ag NW structures as a function of the Au deposition layer thickness.



Fig. S5. SERS intensity changes at  $810 \text{ cm}^{-1}$  as a function of the NO<sub>2</sub> concentration.



Fig. S6. Transient  $NO_2$  gas response between 0.3 ppm and 3.0 ppm using the 3D hybrid Ag

NW structures at RT.

References

1. M. Park, H. Jung, Y. Jeong and K. H. Jeong, ACS Nano, 2017, 11, 438-443.