Supporting Information

Table S1. Relationship between SERS intensity of "bacteria (S. *aureus*, SA, 5×10^7 cfu/ml)" and inter-particle gap (W)/diameter (D) ratio of Ag nanoparticles.

Ag/NSP	**1/99**	**7/93**	**15/85 (Ag/MMT)** [d]	**30/70**	**50/50**
Inter-particle gap (W, nm)	49.8±11.3	31.3±8.0	17.4±4.8 (16.1±4.6)	8.5±2.8	6.3±2.2
Mean diameter (D, nm) [a]	3.8±1.8	5.0±3.2	9.3±4.5 (11.7±3.9)	17.0±5.0	35.0±21.0
W/D	13.1±3.0	6.3±1.6	1.9±0.5 (1.4±0.2)	0.5±0.2	0.2±0.1
SERS intensity $\times10^{-3}$ [b]	7.7±2.1	54.3±8.5	130.1±10.1 (65.1±20.3)	213.3±12.3	238.7±46.8
SERS intensity per Ag particle$\times10^{-3}$ (I_{Stokes})	2.0±0.5	6.0±0.6	9.3±0.7 (4.3±1.4)	14.2±0.8	17.1±3.3
Normalized SERS signal ($I_{Stokes}/I^{\infty}_{Stokes}$) [c]	1.0±0.3	3.0±0.3	4.7±0.4 (2.2±0.7)	7.1±0.4	8.6±1.7

[a] measured by TEM

[b] integrated area of SERS intensity between 700 and 770 cm^{-1}

[c] I_{Stokes} is the average Raman signal per Ag nanoparticles and I^{∞}_{Stokes} is I_{Stokes} for a large inter-particle gap (Ag 1/NSP 99)

[d] Ag/ montmorillonite (MMT, few layer silicate platelets, inter-particle gap in z-direction:~20 nm)

Table S2. Relationship between SERS intensity of "small molecules (adenine from DNA, 5×10^{-5} M)" and inter-particle gap (W)/diameter (D) ratio of Ag nanoparticles.

Ag/NSP	**1/99**	**7/93**	**15/85 (Ag/MMT)** [d]	**30/70**	**50/50**
Inter-particle gap (W, nm)	49.8±11.3	31.3±8.0	17.4±4.8 (16.1±4.6)	8.5±2.8	6.3±2.2
Mean diameter (D, nm) [a]	3.8±1.8	5.0±3.2	9.3±4.5 (11.7±3.9)	17.0±5.0	35.0±21.0
W/D	13.1±3.0	6.3±1.6	1.9±0.5 (1.4±0.2)	0.5±0.2	0.2±0.1
SERS intensity $\times10^{-3}$ [b]	10.5±11.4	53.6±22.3	173.0±32.0 (104.2±15.4)	301.9±19.1	323.9±32.5
SERS intensity per Ag particle$\times10^{-3}$ **(I_{Stokes})**	2.7±2.5	6.0±1.2	14.4±2.6 (6.6±1.1)	20.1±1.3	23.1±2.3
Normalized SERS signal ($I_{Stokes}/I^{\infty}_{Stokes}$) [c]	1.0±0.1	2.2±0.5	5.3±1.0 (2.4±0.4)	7.4±0.5	8.5±0.9

[a] measured by TEM

[b] integrated area of SERS intensity between 700 and 770 cm^{-1}

[c] I_{Stokes} is the average Raman signal per Ag nanoparticles and I^{∞}_{Stokes} is I_{Stokes} for a large inter-particle gap (Ag 1/NSP 99)

[d] Ag/ montmorillonite (MMT, few layer silicate platelets, inter-particle gap in z-direction:~20 nm)

Figure S1. The experiment of tilted sputtering of Au for investigating the locations of Ag nanoparticles on NSP: Ag nanoparticles were confirmed to grow on both sides of NSP. **(a)** Schematic diagram of Au nanoparticles sputtered from a tilted angle onto a silicate platelet where a Ag nanoparticle is attached on the top side of the platelet. **(b)** TEM image (corresponds to the schematic in a) shows a Ag nanoparticle (large particle) on the top side of the silicate platelet. As a result, Au nanoparticles could not reach the dashed-line region. **(c)** EDS spectra (line scan) show the distribution of Au and Ag from the Ag particle area shown in b. The distribution of Au is directional as a result of the blocking of the large Ag particle. **(d)** Schematic diagram of Au nanoparticles sputtered from a tilted angle onto a silicate platelet where a Ag nanoparticle is attached on the bottom side of the platelet. **(e)** TEM image (corresponds to the schematic in d) shows a Ag nanoparticle (large particle) attached on the bottom side of the same silicate platelet shown in b, where Au nanoparticles (small particles) were deposited randomly on top of the platelet. **(f)** EDS spectra (line scan) shows the distribution of Au and Ag from the Ag particle area shown in e. Au is evenly distributed in the area since the Ag particle is under the platelet and could not block the sputtering of Au. The Ag signal is stronger than the Au signal because the Ag particle is much larger than the Au particles in size. (Scale bar: 5 nm)

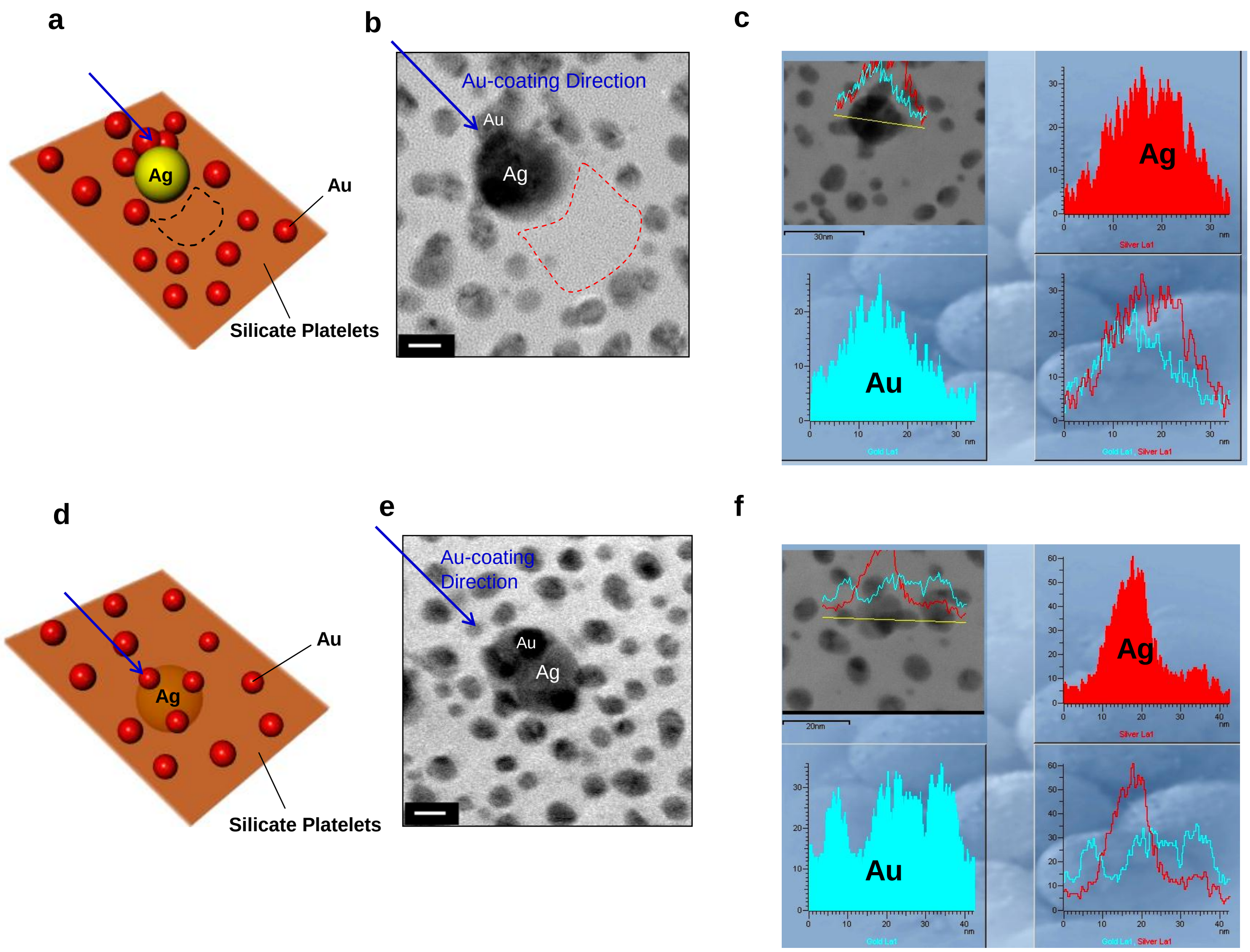

Figure S2. Relationship between inter-particle gap (W)/diameter (D) ratio and absorption wavelength of Ag/NSP nano-hybrid substrate.

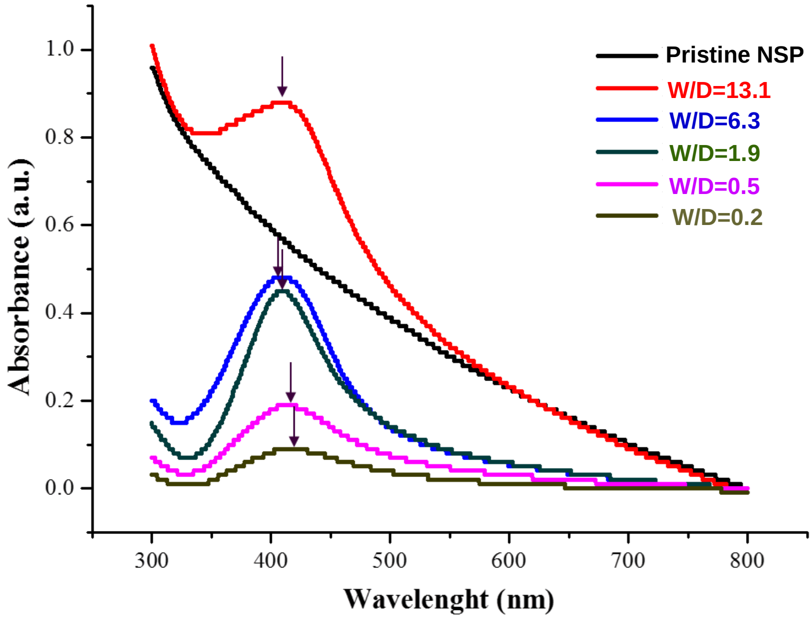

Figure S3. (a) The stability (UV/Vis) tests of Ag30/NSP70 for 4 hours; (b) The heating stability tests at 80°C for 8 hours. These results exhibit strong binding between Ag nanoparticles and NSP, because no significant change of UV/Vis absorption peak.

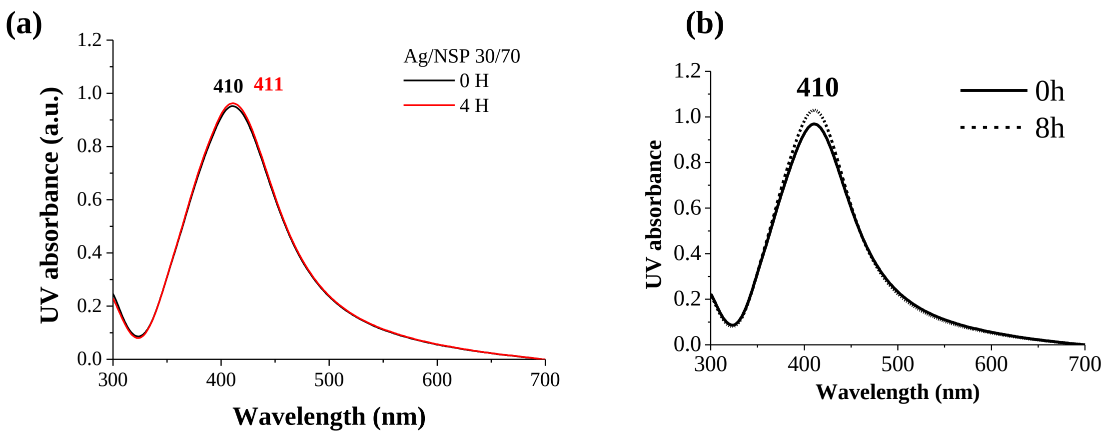

Figure S4. The thorough investigation as the contact between the nanoparticles and the bacteria were shown in the following. We prepared different ratios of bacteria and Ag-NSP to measure the SERS spectra (one unit of bacteria=10^8 cfu/ml; one unit of Ag-NSP=100 ppm). The detection limits of bacteria/Ag-NSP ratios should be set in the range of 1 to 30. The bacteria/Ag-NSP ratios in the 1/3 and 1/10 show poor SERS spectra due to the fewer bacteria and huger fluorescence interference from NSP. In contrast, the bacteria/Ag-NSP ratio in the 30/1 displays the lack of Ag-NSP to detect the huge numbers of bacteria.