Supplementary Information

3D Multilayerd Plasmonic Nanostructures with High Areal Density

for SERS

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Fig. S1 Evolution of the Ag nanostructures as a function of the RIE time. PDMS nanoprotrusions were generated by CF_4 RIE over different etching time periods of (a) 30 s, (b) 45 s, (c) 75 s, and (d) 90 s. A 50 nm thick Ag layer was evaporated onto the PDMS protrusions.

Ag nanostructure



Ag NPs / Al₂O₃ / Ag nanostructures



Fig. S2 The measured number densities of the Ag nanostructures were (a) 131 μ m⁻² and (b) 1400 μ m⁻².



Fig. S3 SEM images of the Ag nanoparticles deposited onto the flat 2D alumina/PDMS double layer. Deposition of (a) 3 nm, (b) 5 nm, (c) 7 nm, and (d) 9 nm thick (based on nominal thickness) Ag thin films on the smooth alumina/PDMS surface.



Fig. S4 Comparison of the SERS performance on (a) the 3D nanostructures and (b) the 2D nanostructures, depending on the size of the top Ag nanoparticles. The excitation laser was $\lambda = 632.8$ nm and the incident illumination power was 0.45 mW. The 3D plasmonic nanostructures exhibited superior SERS performances compared to the 2D plasmonic nanostructures, regardless of the size of the Ag nanoparticles, due to the enhanced optical response.



Fig. S5 Comparison of the SERS peak intensity at 998 cm⁻¹ in the presence of the probe molecule (benzenethiol) on the different SERS substrates, as a function of the top Ag nanoparticle size.



Fig. S6 SEM images of the Ag nanoparticles deposited onto the smooth Si substrate. Deposition of (a) 3 nm, (b) 5 nm, (c) 7 nm, and (d) 9 nm thick (based on nominal thickness) Ag NPs on the Si wafer. All scale bars indicate 100 nm.



Fig. S7 2D Raman mapping of a 2 μ M BT-treated 3D plasmonic nanostructure. The Raman mapping was based on the signal intensity at 998 cm⁻¹ Raman mode.



Fig. S8 SERS spectra of methylene blue at different concentrations.