

# Three generations of surface nanocomposites based on hexagonally ordered gold nanoparticle layers and their application for surface-enhanced Raman spectroscopy

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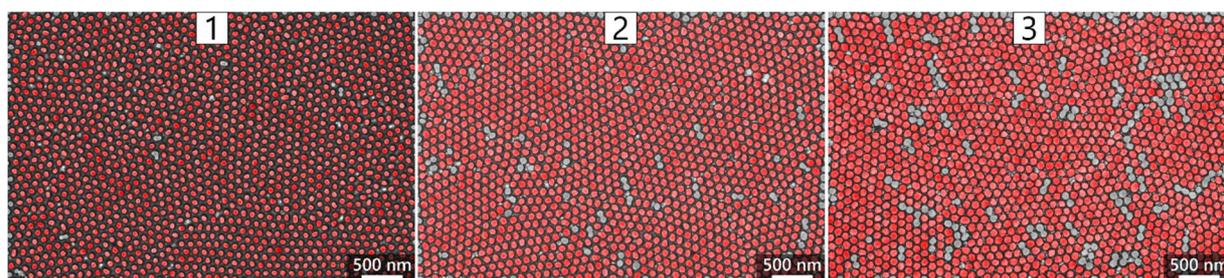
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## S1. Nanoparticle Size Distribution Determination

Wide-view field SEM images were used to estimate the parameters of AuNP layer distributions, as illustrated in Fig. S1. The calculation was performed over the selected nanoparticles (masked red) (>1300). The size is estimated as the mean of circle diameters with an area equal to the area of NPs. Merged AuNPs were excluded from the calculations and counted as defects. The calculations were performed by image processing software (Adobe Photoshop, Gwyddion). The results are presented in Table 1 (in the main paper).



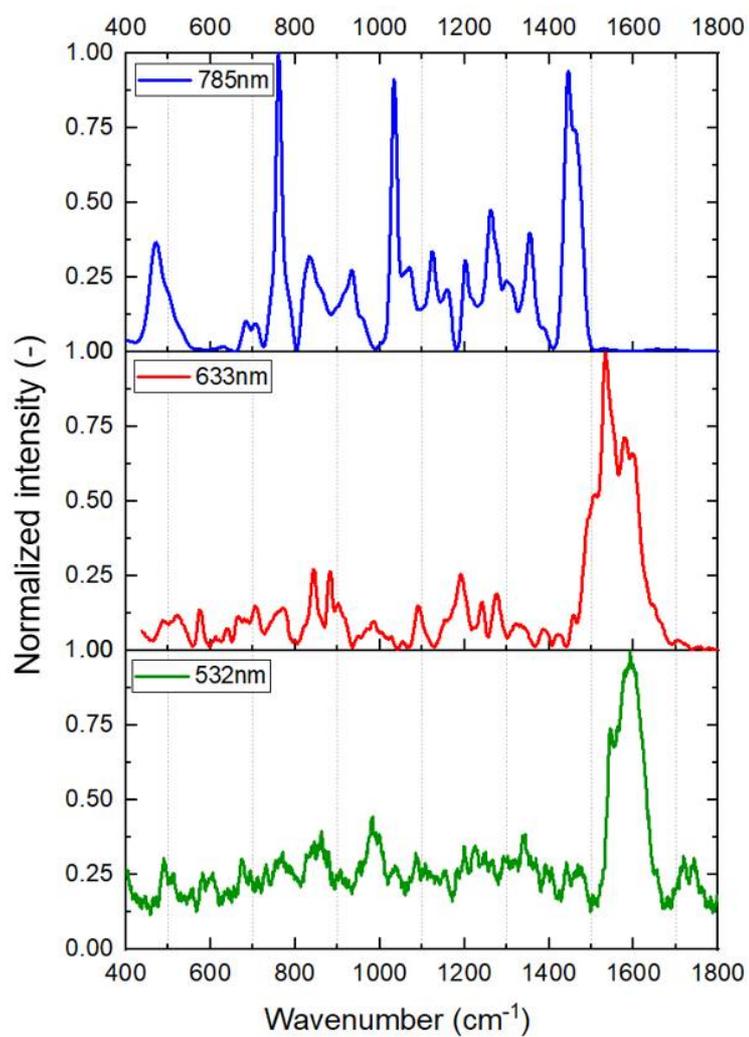
**Figure S1.** Wide view field SEM images made on the three different nanoparticle arrangements. The red mask marks the particles that were included in the particle size distribution determination. Unmasked particles were treated as defects.

## S2. Thermal Stability of the Different SERS Substrates

As mentioned in the main text, one of the main improvements of the third generation of nanocomposites is that the SiO<sub>2</sub> film also increased the thermal stability, allowing SERS measurements using a 633 nm laser.

The thermal stability of the nanocomposites depends on the used excitation wavelength and power. Closer to the plasmon absorption band of the nanoparticle arrangement, the thermoplasmonic effects are stronger (e.g. 532 and 633 nm). With the second generation of nanocomposites, this resulted in the degradation of the epoxy layer at layer powers of 0.5 mW at 633 nm, and 0.6 mW at 532 nm, as illustrated in Fig. 2S. However, 20 mW at 785 nm did not damage the substrate at all. All three spectra in Fig. 2S were obtained on a DNA monolayer covering the gold nanoparticles. The DNA-related peaks are only visible at 785 nm, while the wide peak around 1600 cm<sup>-1</sup> related to carbon proved the degradation of the epoxy substrate at 532 and 622 nm excitation wavelengths.

As demonstrated in Section 3.4 in the main paper, similar or even higher laser power at these excitation wavelengths was not damaging to the third generation of samples. This enables the fine-tuning of the absorption peak of the substrates to the used excitation wavelength at 633 nm.



**Figure S2.** SERS spectra obtained on a DNA monolayer obtained at different wavelengths and powers with the second generation of nanocomposite substrates. The power was 20 mW at 785 nm, 0.5 mW at 633 nm, and 0.6 mW at 532 nm.