

Supplementary materials

Plasmonic nanoparticle-based expansion microscopy with surface-enhanced Raman and dark-field spectroscopic imaging

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Characterization of image background:

The data presented in the paper has undergone no background subtraction nor smoothing. The background signal which we measure at every point on each maps comes mainly from the microscope slide and/or coverslip fluorescence emission in the NIR. Figure 1 shows one example Raman spectrum of the microscope slide @785 nm laser excitation 60 mW, 1 s integration time. This background was present on all spectra and changed very little. The conspicuous background peak below is at $\sim 1500\text{ cm}^{-1}$, which is very far away from the SERS fingerprints or the 590 cm^{-1} peak we used.

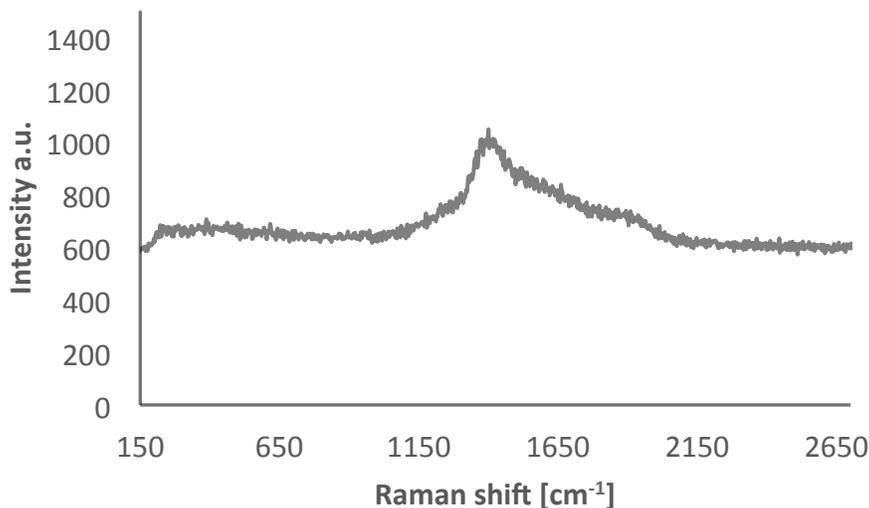


Fig. 1 Background in SERS measurements.

2. SERS intensity distribution before and after expansion

We have generated the intensity histograms from 5 un-expanded and 3 expanded samples as shown in Fig. 2. We observe an intensity reduction of approximately 2-3X instead of 16X. We believe this is due to remaining NP clusters within individual pixels after expansion. In other words, in order to see a 16X reduction in SERS intensity, all NP labels have to become farther apart (farther than the imaging pixel size) in a uniform fashion. We suspect this did not happen. In other words, some of the clustered NP labels after expansion were still within an imaging pixel size so their SERS intensities were still summed, resulting in the non-linear intensity decrease. As shown in the FDTD simulation, for the plasmon to decouple, however, the NPs only have to be more than $\sim 20\text{ nm}$ apart.

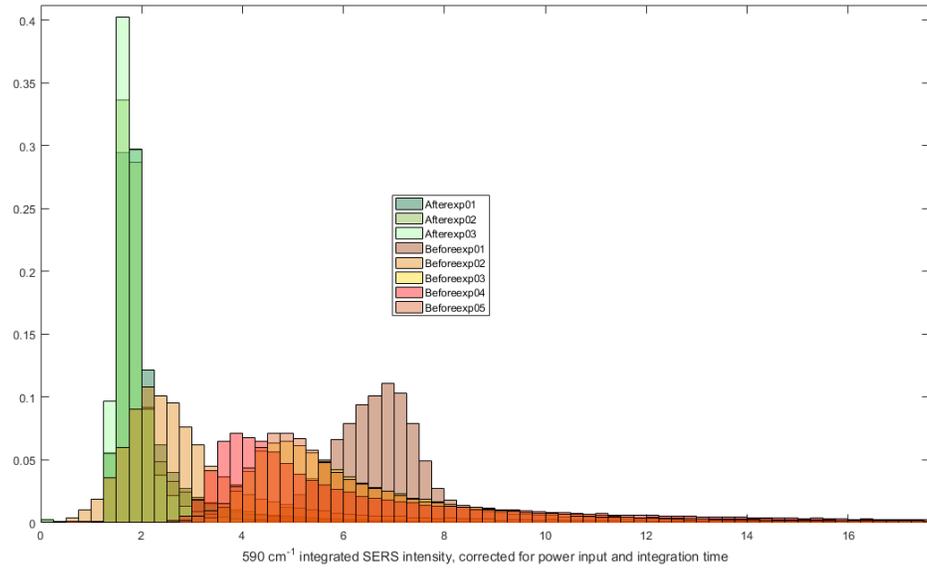


Fig. 2 SERS intensity histogram for before and after expansion specimens.