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Strong correlations between optical near-field microscopy and far-field Raman imaging on nanorough gold SERS substrates

It is largely considered that the Surface Enhanced Raman Scattering (SERS) effect is mainly due to the presence of localized electromagnetic fields (known as hot spots) on metallic surface under irradiation. The origin and the role of hot spots on a plasmonic material have been widely studied. However, since hot spots are due to nanoscale surface singularities, it remains a challenge to control the density of singularities on a surface by depositing nanoparticles from a solution or by depositing metallic grains by vapor deposition technique for instance. Although the thermal evaporation and the sputtering techniques are not commonly used to elaborate SERS surfaces [1,2], we recently optimized procedures to elaborate efficient nanorough gold SERS substrates. The modification of deposition parameters can induce variations of the surface topography thus leading to variations in the SERS enhancement factor. Raman mapping were recorded at $\lambda = 633$ nm on different nanorough gold substrates after immersion in a 10^{-6} M solution of thiophenol for 10 minutes. It is then possible to analyze the intensity distribution and the average Raman intensity on each substrate. By using near-field microscopy known as PhotoEmission Electron Microscopy (PEEM), it is possible to study densities of hot-spots and their energy distributions.

Here, we will show how near-field optical microscopy (PEEM) and far-field Raman scattering are strongly correlated when it comes to quantifying surface physical properties. These results allow to better understand the SERS phenomena on a plasmonic metal surface, in addition to being able to predict the relative variations of the Raman enhancement factors by the quantification of hot spots on a surface.

References

- [1] A. Merlen, V. Gadenne, al. "Surface Enhanced Raman Spectroscopy of Organic Molecules Deposited on Gold Sputtered Substrates". *Nanotechnology* 20 (21),**2009**, 215705.
- [2] M. Edely, N. Delorme, D. Siniscalco, J.-F Bardeau."Alternative Strategy Based on Scanning Probe Lithography for Patterning Complex Metallic Nanostructures on Rigid or Flexible Substrates". *Advanced Materials Technologies* **2018**, 3 (11), 1800134.

Figures

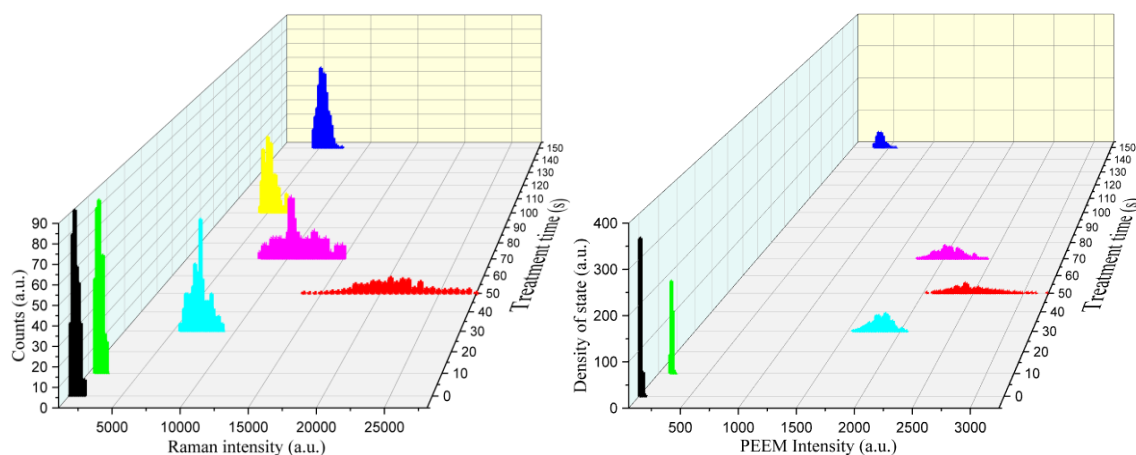


Figure 1. a) Raman intensity distributions function of treatment time b) PEEM Intensity function of treatment time