Self-Assembly of Au Nano-Islands with Tuneable Organized Disorder for Highly Sensitive SERS

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S1. Large Scale Uniformity of SERS Substrates with Uniform Disorder

Fig S1. (a-f) Low magnification SEM images for the Au NIs substrates with different Au aerosol exposure time from 1 s to 30 s. The micrographs show the high uniformity and reproducible disordered morphology over large scales. A transition from an incoming aerosol- to a surface coagulation-induced growth is observed at 5 s.
S2. Morphological and Correlated Disorder Characterization

**Fig. S2.** (a,b) Scanning electron microscopy images of the Au NIs substrates with 5 s and 10 s aerosol deposition time and (c,d) two-dimensional FFT images of the respective samples.
S3. Optical Properties After Exposure of Different Concentration of R6G

**Fig. S3.** Normalized reflection spectra before and after the drop-casting of different concentration of R6G onto the Au NIs substrate with 20 s of deposition time.
**S4. SERS EF with washing substrate**

**Fig. S4.** SERS and Raman spectra of R6G 10^{-4} M on the 20 s Au NIs substrate and a bare Si substrate, respectively. Note that the red line has been multiplied three times. In this case, after dropcasting, the analyte was let dry naturally and thereafter the substrates were gently rinsed with ethanol to remove any excess of dye and induce the presence of a monolayer.