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SUPPORTING INFORMATION

Engineering the synthesis of silica-gold nano-urchin particles using continuous synthesis

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Figure S1. TEM images of silica particles synthesized by two phase sol-gel reaction and subsequent seeded regrowth. (a) 25 nm silica seed particles synthesized by two phase method. (b) 80 nm rough surfaced particles prepared by two phase regrowth. (c) 80 nm smooth surfaced particles prepared by Stöber regrowth procedure.



Figure S2. TEM images of Au seeded-roughened silica nanoparticles synthesized under ultrasonic radiation using a sequential addition procedure, adding ascorbic acid as reducing agent. Nanostructures obtained after :a) 7 cycles, b) 15 cycles and c) 22 cycles.



Figure S3. TEM images of Au seeded-roughened silica nanoparticles synthesized under ultrasonic radiation using a sequential addition procedure, adding hydroxylammonium chloride as reducing agent. Nanostructures obtained after :a) 7 cycles, b) 15 cycles and c) Detail of nanostructures obtained at 15 cycles.



Figure S4. TEM images of Au seeded-roughened silica nanoparticles synthesized under ultrasonic radiation using a sequential addition procedure, adding sodium borohydride as reducing agent. SGNU obtained after :a) 7 cycles, b) 15 cycles and c) 22 cycles.



Figure S5. TEM images of Au seeded-roughened silica nanoparticles synthesized under magnetic stirring using a single addition procedure, adding sodium borohydride as reducing agent. Nanostructures obtained after :a) 5 minutes, b) 20 minutes and c) 40 minutes.



Figure S6. TEM images of Au seeded-roughened silica nanoparticles synthesized under ultrasound radiation using a single addition procedure, adding sodium borohydride as reducing agent. Nanostructures obtained after :a) 5 minutes, b) 20 minutes and c) 40 minutes.



Figure S7. TEM images of Au seeded-roughened silica nanoparticles synthesized under ultrasonic radiation using a continuous flow microreactor, adding sodium borohydride as reducing agent. Residence time = 12 minutes. SGNU obtained under different NaBH₄ | Gold precursor flow rates: a) 50 μ L/min | 50 μ L/min , b) 20 μ L/min | 50 μ L/min c) 5 μ L/min | 50 μ L/min | 50



Figure S8.- Optical absorption spectra from SGNU produced in sequential addition batch reactor (22 additions) and microfluidic continuous system (Residence time = 12 min, NaBH4:gold precursor solution flow ratios of 1 and 50 μ L/min, respectively)



Figure S9. Scheme for the microfluidic platform for the preparation of SGNUs and 3D SERS substrates. (a) Scheme for the microfluidic reactor used for the continuous synthesis of SGNUs. (b) Scheme of the emulsion generator to prepare 3D SGNU substrates. Right pictures show produced uniform emulsions containing SGNUs to be template into 3D spherical assemblies through evaporation.



Figure S10. Bulk Raman spectra of R6G. (a) Raman spectrum from the R6G doped silica nanoparticle susbstrate. (b) Bulk Raman spectrum from the pure R6G powder.



Figure S11. SEM images of assembled SGNUs in 2D form on silicon wafer (a) and 3D form (b). In (b), 3D sample was crushed to see internal structures.