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Colloidal lenses as universal Raman scattering enhancers

SUPPORTING INFORMATION

SI 1. Optical microscope images of SiO₂ spheres on TiO₂ thin films: example of sampling procedure

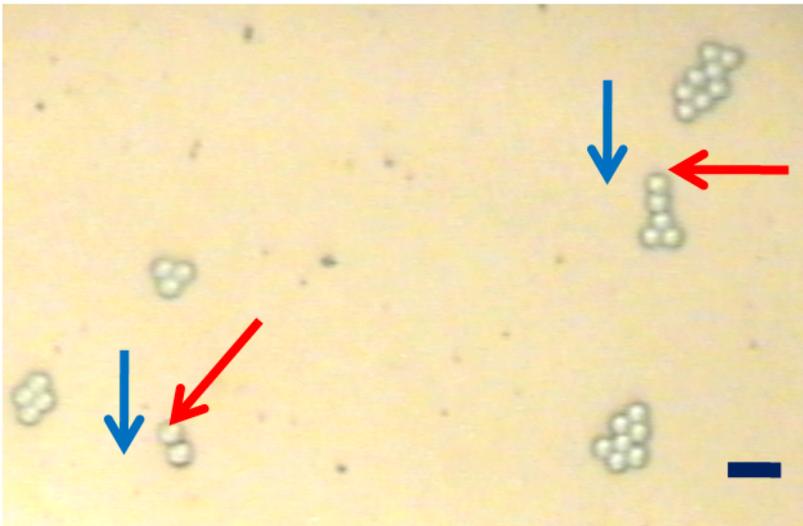


Figure S1. Example of SiO₂ microspheres drop-casted onto a single-crystal Si substrate coated with anatase (thickness: 20 nm) by ALD (*see* experimental section for details). Raman spectra were acquired on both individual spheres and planar (without spheres) regions. An example of sampling is displayed. Red arrows indicate two spheres used as μ -lenses. Blue arrows indicate reference planar regions. Scale bar: 5 μ m

SI 2. ERI factor of anatase layer and Si substrate as a function of N.A.

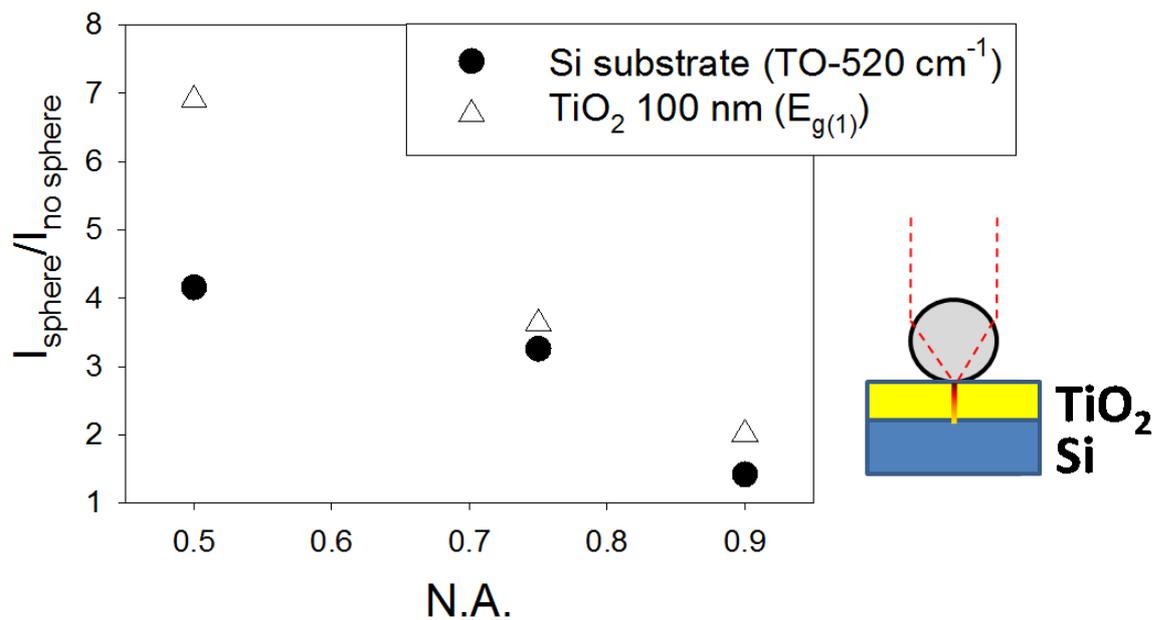


Figure S2. ERI factor (intensity ratio of a Raman mode between SiO_2 -coated and uncoated samples) for either anatase or Si main Raman modes as a function of N.A. of the microscope objective. The error bar is included in the experimental points. See the main text for details.

SI 3. Optical microscope images of SiO₂ spheres on Si substrates soaked by methylene blue (MB) solutions: example of sampling procedure.

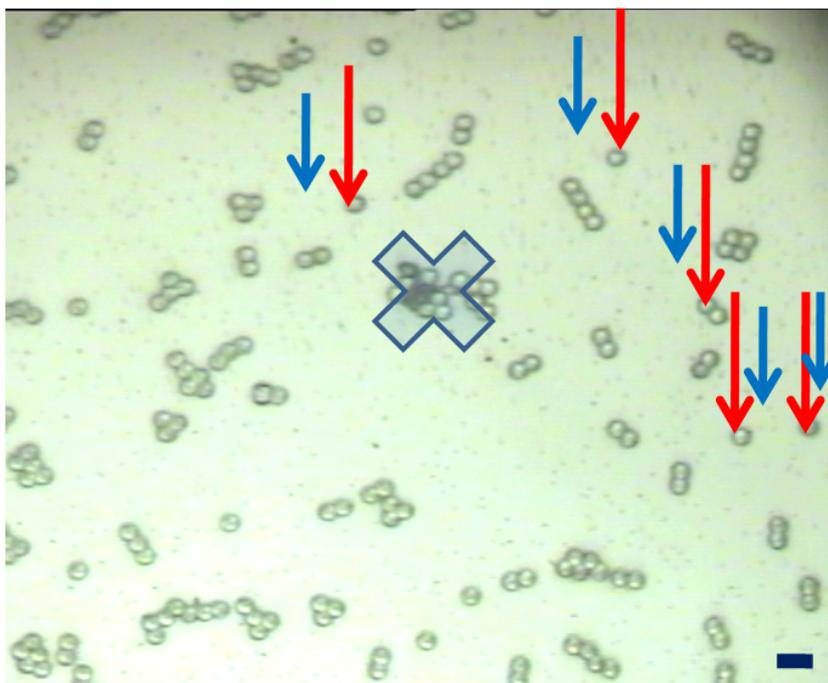


Figure S3. Example of SiO₂ microspheres drop-casted onto a single-crystal Si substrate which have been previously soaked in a 10⁻⁴ M MB solution (*see* experimental section for details). Raman spectra were acquired on both individual spheres and planar (without spheres) regions. An example of sampling is displayed. Red arrows indicate some spheres used as μ -lenses. Blue arrows indicate the corresponding reference planar regions. The light-blue cross indicates a region where MB is accumulated on the spheres. This type of region was not considered in acquisition of Raman spectra. Scale bar: 5 μ m

SI 4. Au nanoislands: SEM and optical characterization of the Raman active substrate

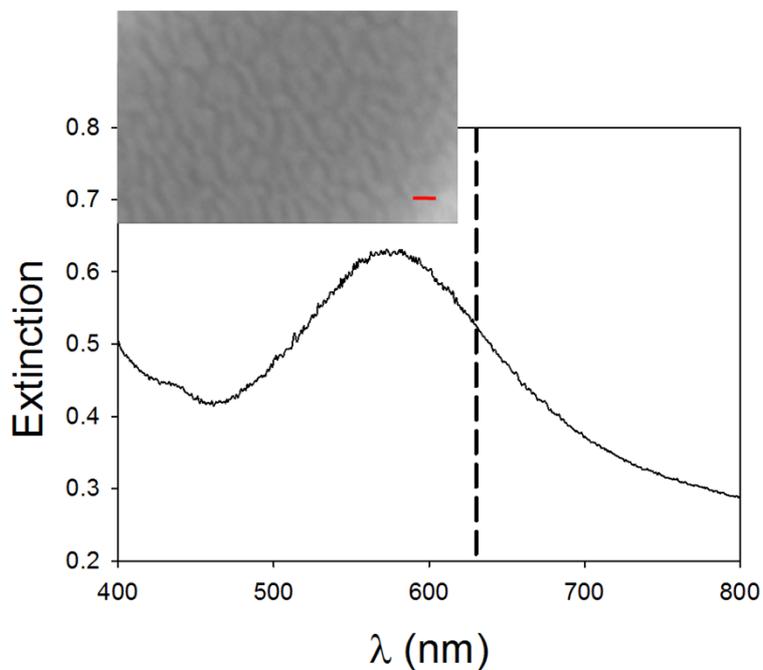


Figure S4. Inset: SEM image showing the morphology of a gold-coated sample. Scale bar: 20 nm. Main Figure: optical extinction showing the broad surface plasmon resonance band due to gold nanoislands. Reference: uncoated Si substrate. Dashed line indicates the position of the He-NE laser used for Raman experiments.

SI 5. Micromanipulation of colloidal μ -lenses

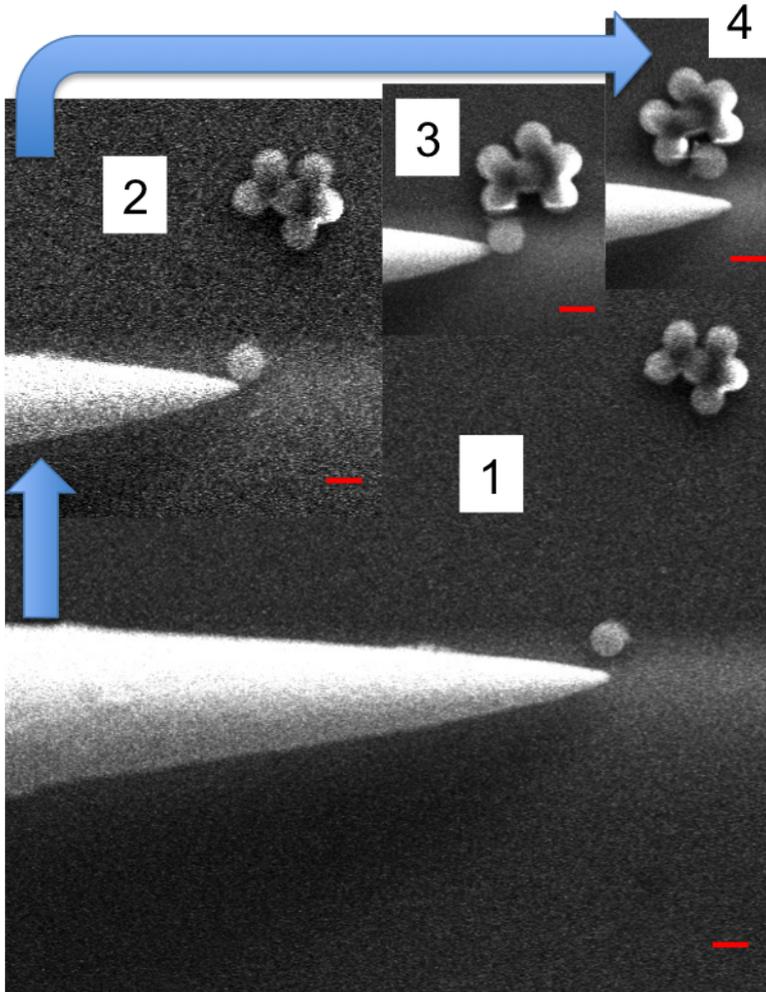


Figure S5. SEM images showing an example of controlled positioning of SiO₂ microspheres by means of a piezo-actuated micromanipulator. Scale bars: 2 μ m.