Supporting information: Selective TERS Detection and Imaging through Controlled Plasmonics

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DF and AFM images of SW620 cells incubated with c-RGD GNPs:



Figure S1. a, b) Typical dark-field images of SW620 cells after incubation with 80 nm cRGD-GNPs are shown. TERS tip is engaged onto cell membrane, with blue circle representing tip positioning; c, d, e, f) The corresponding trace and re-trace images obtained from the AFM scan during TERS microscopy on the different areas of the cell membrane are shown.



PLS-DA and TERS analysis of wild type and mutant proteins:

Figure S2. a). TERS map (using marker band 960cm^{-1}) of biotin GNPs on RC-streptavidin slide, 1x1 um (50x50 pixel); b) Sample/scores plot on LV1 in PLS-DA analysis; c) Scores map using PLS-DA calculated scores on LV1 as an indicator of the full-spectral similarity of each TERS spectrum to LV1; d) Selected TERS spectra compared with PLS-DA calculated LV1 (blue). The green curve corresponds to TERS spectrum circled in orange and red curve to TERS spectrum circled in red.



Figure S3. a). TERS map (using marker band 960cm⁻¹) of biotin GNPs on WTstreptavidin slide, 1x1 um (50x50 pixel). b) Sample/scores plot on LV1 in PLS-DA analysis; c) Scores map using PLS-DA calculated scores on LV1 as an indicator of the full-spectral similarity of each TERS spectrum to LV1; d) Selected TERS spectra compared with PLS-DA calculated LV1 (blue). The green curve corresponds to TERS spectrum circled in orange and red curve to TERS spectrum circled in red.





Figure S4. a). SERS spectra of RGD peptide and integrin proteins; b) Sample/scores plot along two principle components in PCA analysis of the SERS data without any constraints; c) sample/scores plot in MCR analysis of the SERS data with non-negativity requirement and constrained to 2 components; d) MCR calculated pure components from the SERS data, green corresponding to component 1 while red component 2.



Figure S5. a,e). TERS map (using marker band 1003cm⁻¹) of RGD GNPs on SW620 cells, 3x3 um, 31x31 pixel, step 97 nm/pixel; b,f) sample/scores plot on LV1 in PLS-DA analysis; c,g) Scores map using PLS-DA calculated scores on LV1 as an indicator of the full-spectral similarity of each TERS spectrum to LV1; d,h) Selected TERS spectra are compared with PLS-DA calculated LV1 (blue). The red curve corresponds to TERS spectrum circled in red, the orange spectrum from the pixel circled in orange, and green spectrum from the pixel circled in green in the images above.