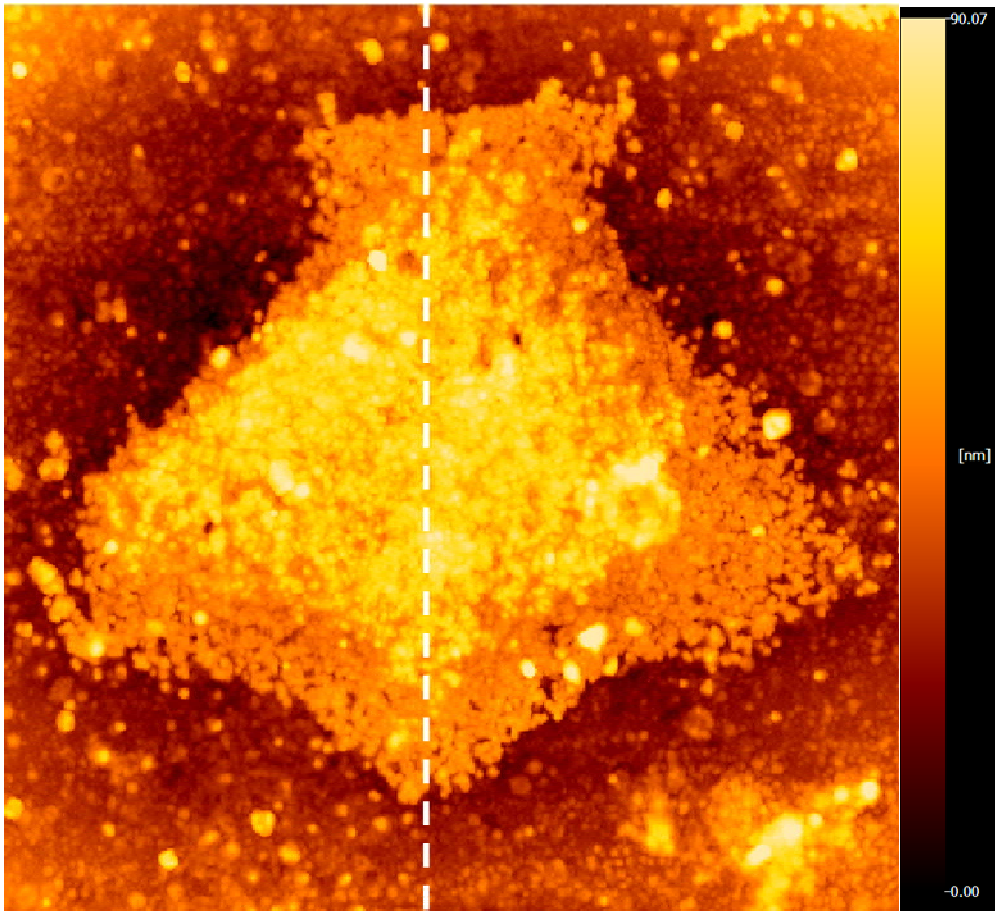
Electronic Supplementary Material for PCCP

SI: Cross-sectional view for layered nanostructure of gold nanoparticles

We have observed that the layered nanostructure of gold nanoparticles under investigation in this study consists of single layer and double layer of colloidal gold nanoparticles. The center part of the layered nanostructure is double layer and is surrounded by single layer of gold nanoparticles near the rim. The single layer is mostly close-pack and covers small area. Although surface view and top view of the layered nanostructure mentioned in the manuscript (fig. 1 a and b) indicate directly the layered nature of the gold nanostructure under this investigation, cross-sectional view of the same sample is shown herewith again to quantify the height value for individual layers. Fig. S1 (a) shows the same layered nanostructure of gold nanoparticles. Image size is 8 μm x 8 μm. Figure SI (b) shows the height value of the same sample along the dashed white vertical line shown in fig. SI (a). The dashed solid line included in the line scan (fig. SI (b)) represents the average height value crossing bare substrate, single layer, double layer, single layer and bare substrate sequentially.

(a)



(b)

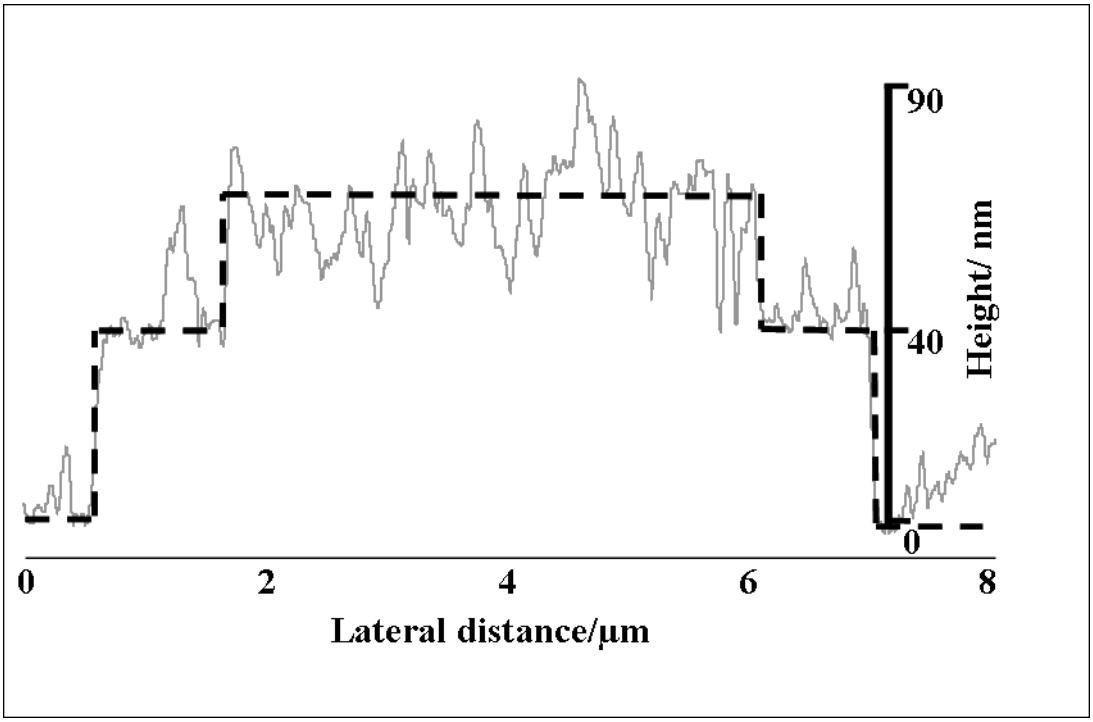


Figure Caption:
Figure SI: (a) AFM image of the layered nanostructure of gold nanoparticles and (b) Height value of the scan along the white dashed line shown in (a).

Electronic Supplementary Material for PCCP

SI: Near-field localization for double layer and single layer nanostructure of gold nanoparticels

Usually in conventional microscopic Raman measurements, the incident laser is always normal to the substrate. In that case, the electromagnetic field of the laser can couple fully with the nanoparticles. In this present study, the incident laser was projected with an angle of 30^0 to get another facility to carry out SPR measurements using dark-field condenser at the spatial position. A freehand schematic is shown in the manuscript (fig. 5 a). In this particular configuration, the electromagnetic field of the incident laser should split into vertical and horizontal components as shown in fig. SII. The individual component is capable to induce localized near-field distributions at the junctions of the gold nanostructure. From this perspective, the double layer nanostructure will be having more localized near-field distribution and thus should induce higher enhancement in SERS. Figure SII shows the schematic of layer nanostructure and their possible near-field distributions.

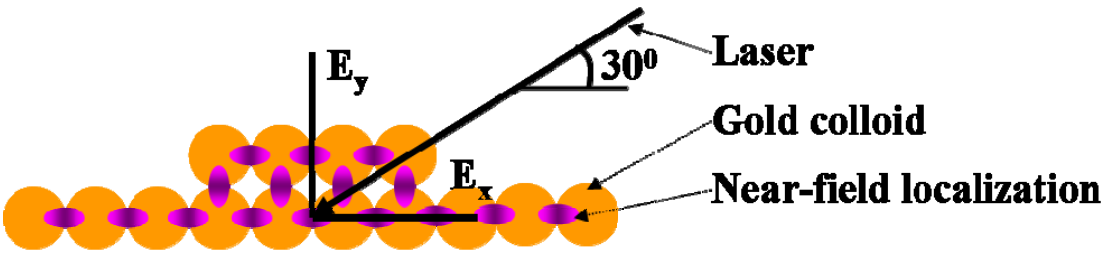


Figure Caption:
Figure SII: Schematic for electromagnetic near-field distribution in the single layer and double layer nanostructure of gold nanoparticles.