Supporting Information for

Tailoring the Core-Satellite Nanoassembly Architectures by Tuning Inter-nanoparticle Electrostatic Interactions

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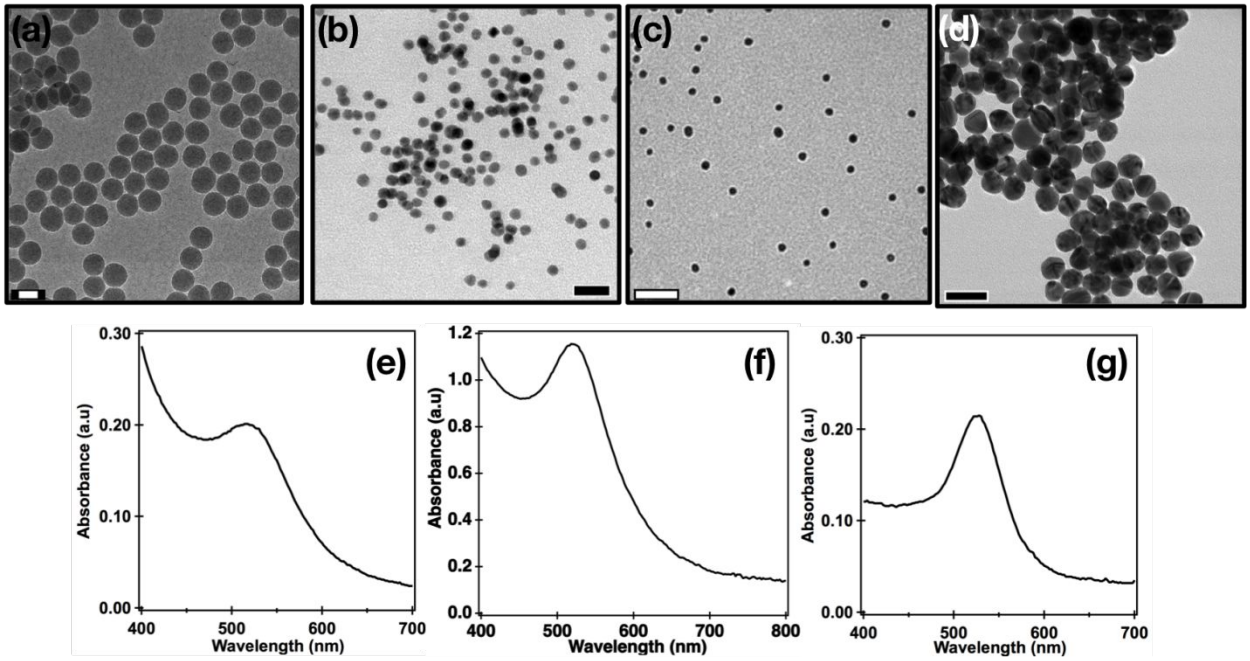

Figure S1: Characterization of the nanoparticles used for the nanoclusters formation. TEM micrographs (a) d= 60 nm SiNPs (scale bar: 100 nm), (b) d= 5 nm, (c) d=10 nm and (d) d= 15 nm gold nanoparticles. Scale bars: 20 nm. Ensemble absorption spectra of (e) d= 5 nm, (f) d=10 nm and (g) d= 15 nm gold nanoparticles.

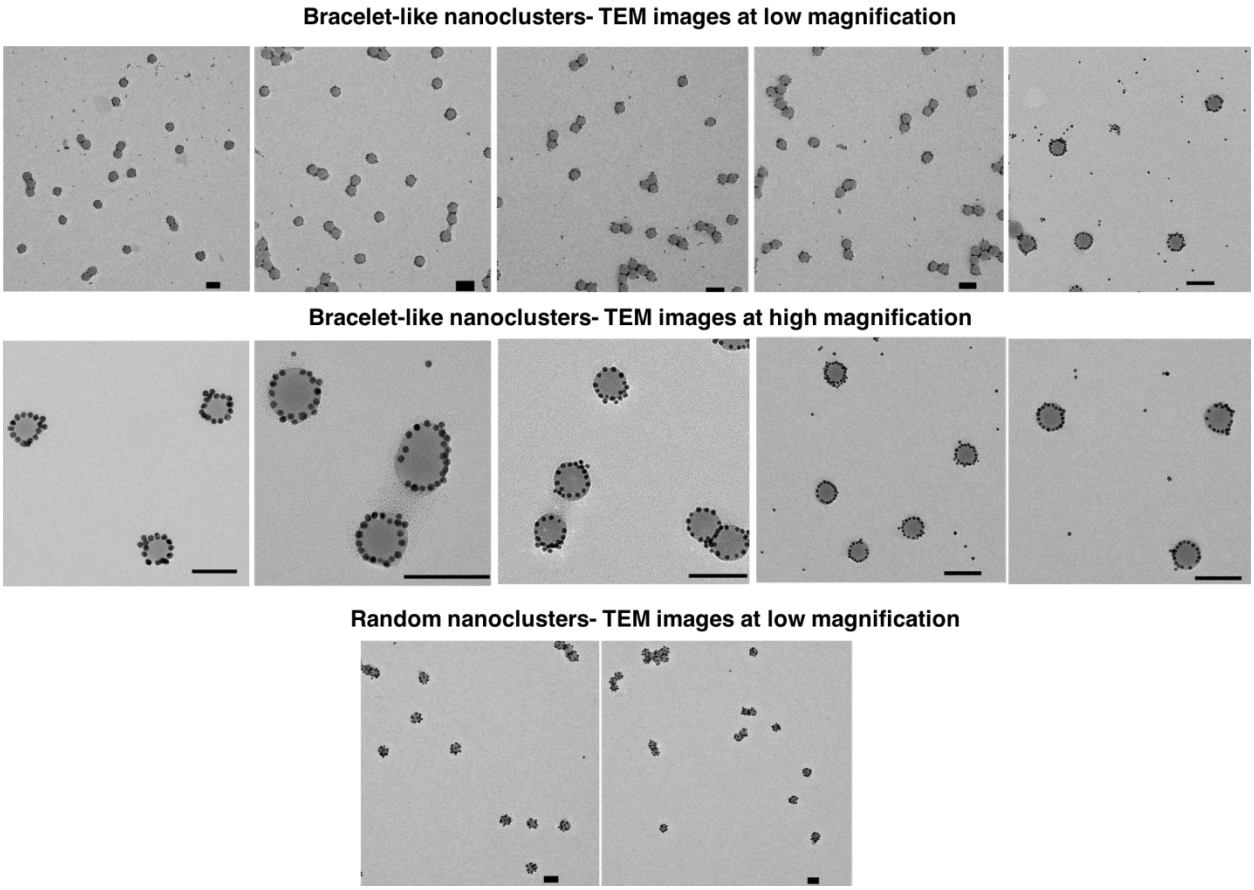

Figure S2: TEM images acquired at low and high magnification show the preferred formation of "bracelet"-like nanoclusters over other nanocluster architecture at pH 9. Low magnification TEM images also show the dominance of random- nanoclusters formed at pH 7, but no other nanocluster architectures. This also shows that the formation of "bracelet"-like nanoclusters is not drying artifact but it depends on the reaction condition, particularly the pH at which the assembly is carried out. Scale bar = 100 nm

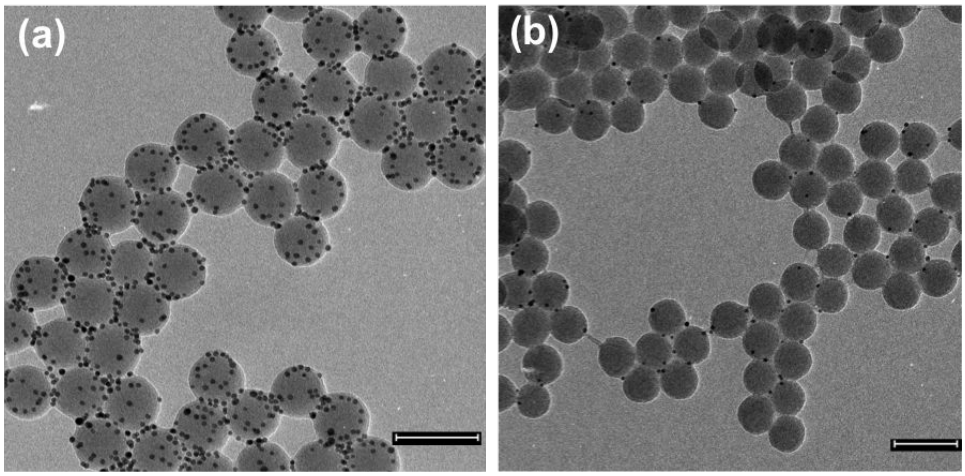

Figure S3: TEM micrographs of nanoclusters prepared with 5nm diameter AuNP satellites (a) at highly acidic (pH 3) condition and (b) at highly basic (pH 9) condition. Nanocluster architecture remains randomly-arranged under both experimental conditions. Scale bar: 100 nm

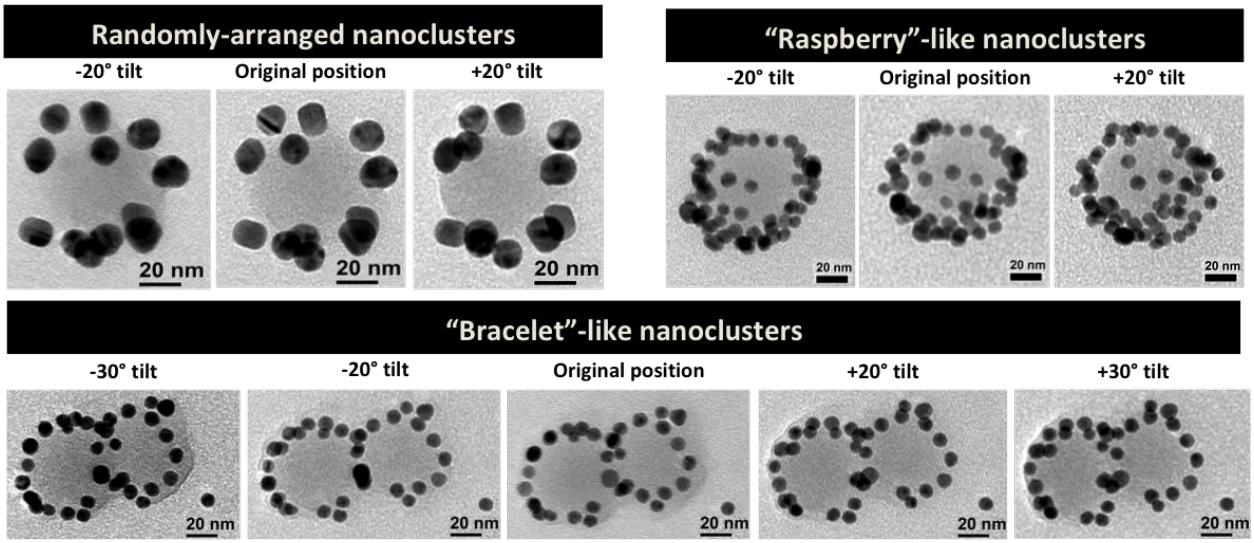

Figure S4: Three- dimensional architecture of nanoassemblies is evident by tilt-angle TEM images. 2-D TEM views of the same single nanoassemblies at -30°,-20°, 0°, +20°, +30° angles for a randomly arranged nanocluster from pH 7, and at -20°, 0°, +20 for a "bracelet"-like nanocluster from pH 9 clearly show the three-dimensional arrangement of satellite AuNPs on the core SiNP.

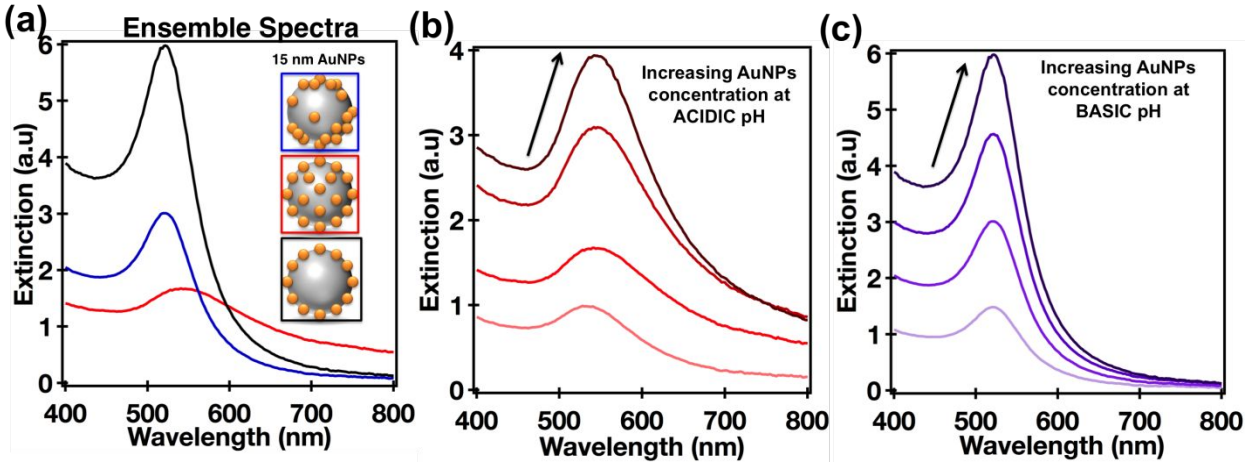

Figure S5: Optical response of nanoclusters is influenced by the pH and the concentration of the AuNP in the assembly reaction medium. (a) ensemble extinction spectra of nanoclusters with 15 nm AuNPs at different nanoclusters architectures obtained acidic neutral and basic pH conditions. Change in the $LSPR_{max}$

peak position of nanoclusters as a function of the concentration of 15 nm AuNPs at (b) acidic pH and (c) basic pH.

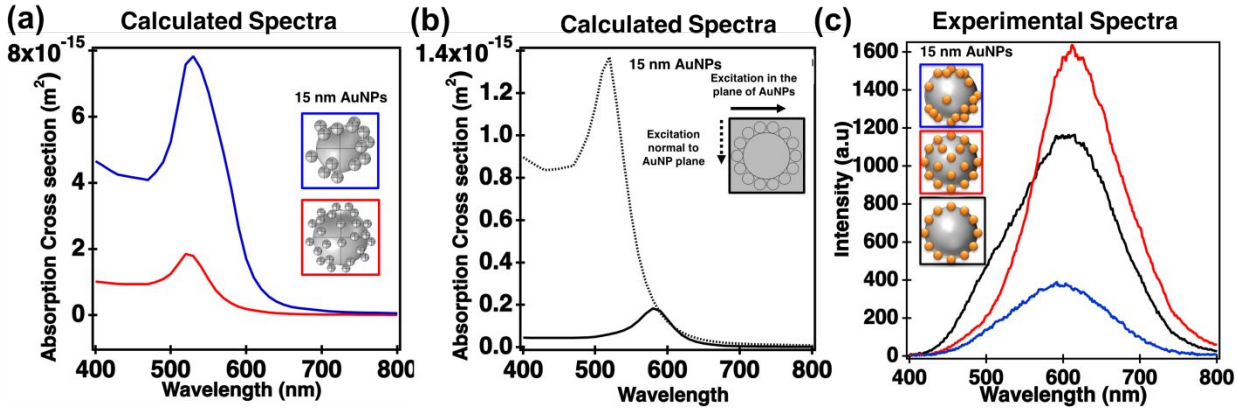

Figure S6: Core-satellite anoassembly architecture affects their optical response. COMSOL caluculated optical spectra of (a) Raspberry-like assemblies AuNPs ($LSPR_{max}$= 525 nm), and randomly- arranged clusters with 15 nm ($LSPR_{max}$= 530 nm), (b) Optical response of bracelet-like nanoclusters with 15 nm diameter AuNPs is dependent with respect to the incident light. Absorption cross-section versus light moving horizontally when the E-field polarized vertically in the plane of AuNPs ($LSPR_{max}$= 580 nm), and the E-field polarized normal to the plane of AuNPs ($LSPR_{max}$= 520 nm). The number and the arrangement of AuNPs of the model nanoclusters are matched to the representative clusters shown in figure 1. (c) Representative single-particle experimental dark-field scattering spectra of a bracelet-like ($LSPR_{max}$= 605 nm), a raspberry-like ($LSPR_{max}$= 610 nm) and a randomly-arranged nanocluster ($LSPR_{max}$= 598 nm) composed 15 nm diameter AuNPs.

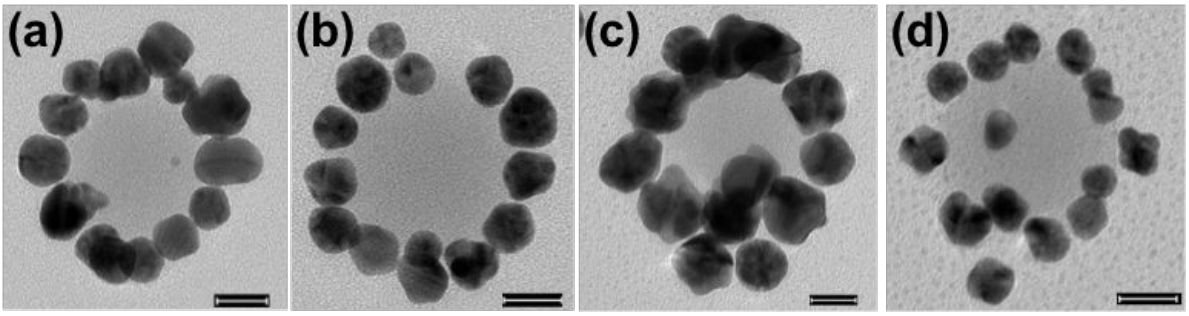

Figure S7: Morphological characteristics of nanoclusters architectures can be modified by the In-situ growth of satellite AuNPs. TEM micrographs showing the growth of (a-c) 10 nm diameter satellite AuNPs and (d) 5 nm diameter satellite AuNPs into larger satellites. The average size of individual satellite AuNPs and the physical contact between them increases to a greater extent as evident in (a-c). It is prominent when go from (a) to (c) where the amount of gold used for the in- situ growth becomes higher respectively. Overall size of 5 nm satellite AuNPs are also increased. Scale bar: 20 nm.

Table S1: Zeta potential measurements of gold nanoparticles and amine-terminated silica nanoparticles at varying pH conditions used for nanocluster formation.

pH	gold nanoparticles	amine-terminated silica nanoparticles
3.0	-9.00 ± 3.25 mV	30.00 ± 13.40 mV
7.0	-12.00 ± 4.15 mV	6.01 ± 4.52 mV
9.0	-50.00 ± 10.70 mV	-4.67 ± 12.90 mV