Optimal design of Au core-shell nanoparticles with embedded Raman reporters

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Recently, several research groups have reported on highly efficient multilayer SERS tags (also called nanomatryoshkas, NMs), in which Raman molecules are embedded in a nanometer-sized interior gap between the metallic core and the shell [1–5]. Such multilayered structures have great potential for biomedical applications due to several advantages: (1) Raman molecules are protected from desorption, subjected to a strongly enhanced electromagnetic (EM) field in the gap, and their SERS response does not depend on the environmental conditions and NM aggregation; (2) owing to bright and uniform spectral pattern, NMs provide a linear correlation between probe concentration and SERS intensity and allow for a real-time *in vivo* imaging and high throughput sensing with short integration times; (3) NM size and spectral properties can be designed for effective cellular uptake and tissue imaging with negligible background from sample autofluorescence; (4) NM probes can be multiplexed by incorporating different Raman molecules into two-layered or multilayered NMs. However, the synthesis and design of optimal gold NMs remain still challenging. Here, we report a detail analysis of EM enhancement in a (core/gap/shell) Au NMs as a function of NM structure, including the core size, the gap and shell thicknesses, and the gap refractive index. The optimization strategy is based on an efficient rigorous solution for internal EM fields in a layered sphere and explicit analytical solutions for the surface and volume averaged EM intensities within a particular layer and around the NM. The SERS enhancement is shown to be strongly dependent, in a resonance manner, on the core and gap size, whereas the shell thickness plays a minor role. The simulation data are exemplified by experimental measurements of SERS spectra for Au NMs with 1,4 benzenedithiol molecules embedded in a sub-nanometer gap.

References

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