Determination of the Optical Properties of Individual Gold Nanorods Through Numerical Modelling and Experiment

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Abstract

The optical scattering and absorption of gold nanorods (GNRs) depends on its size, shape, and surroundings. This dependence is due to both intrinsic and extrinsic effects. A good understanding of this dependence is needed for applications of GNRs in photo-thermal therapy, optical and opto-acoustic imaging, biosensing, and other photonic areas. Extrinsic effects are caused by the production of localized and surface plasmons and are well understood through Mie theory for spherical particles and Maxwell's equations for arbitrary objects. Intrinsic effects become prominent at the nano-scale, when the cluster size becomes comparable or smaller than the bulk mean free path of electrons between collisions and additional scattering of the conduction electrons from the cluster surface occur. The addition of this scattering will shorten the mean free path between collisions and increase the damping constant. The increased damping is due to radiative losses in the case of gold nanoparticles larger than 20 nm in diameter and surface scattering in the case of smaller nanoparticles. The surface scattering effect is quantified using a surface scattering parameter, A [1] and Leff=4V/S (V is the volume and S is the surface of the object) [2]. Surface scattering parameter A affects full width at half maximum of the surface plasmon resonance and usually used as a fitting parameter between theory and experiment. The surface scattering parameter for silica coated GNRs is not known. We will report on its measurement for silica coated GNRs using a combination of spatial modulation spectroscopy (SMS) [3], transmission electron microscopy (TEM), and numerical modeling using the finite element method (FEM). Individual silica coated gold GNRs were deposited on a TEM grid (made of silica) (Figure 1a) and SMS (Figure 1b,c) was used to quantitatively determine the absorption cross-section of three individual GNRs. TEM was used to image dimensions of the three GNRs (Figure 1d). The numerical model was built using COMSOL Multiphysics 3.5a to solve for the total electric field and to simulate the experimental setup of SMS technique (Figure 2). The incident electric field, Einc, propagated in the downward direction (Figure 2b) with a polarization in the left/right direction (along longitudinal axis of the nanorod). In order to obtain a stable solution, at least 10 linear elements per wavelength were used during meshing of the geometries with further refinement inside of the nanorod (Figure 3a). The theoretical calculations of the absorption cross-section of the three GNRs were made using the modified bulk dielectric function of gold with inclusion of the electron surface scattering effect [2]. The experimental data and numerical calculations were used to determine A for each GNR (Figure 4). The three values obtained were A1= 0.47, A2= 0.46 and A3 = 0.55, suggesting a universal value of A = 0.5 for silica coated GNRs. The dependence of the dielectric function of gold nanoparticles on size/shape

can be quantitatively determined by combining SMS and correlated TEM of measurements of size/shape with numerical modeling, assuming Leff is quantitatively known.

Reference

1. Uwe Kreibig and Michael Vollmer, Optical Properties of Metal Clusters (Springer, 1995)
2. Eduardo A. Coronado and George C. Schatz, "Surface plasmon broadening for arbitrary shape nanoparticles: A geometrical probability approach," Journal of Chemical Physics 119, 3926–3934 (2003)
3. Otto L. Muskens, et. al., "Quantitative absorption spectroscopy of a single gold nanorod", Journal of Physical Chemistry C 112, 8917–8921 (2008).

Figures used in the abstract

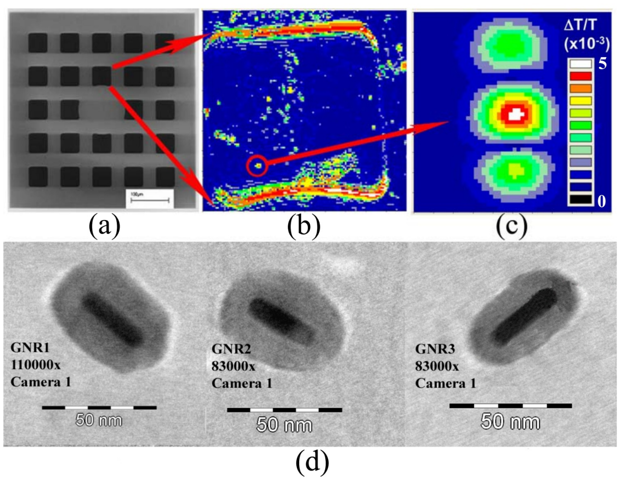


Figure 1: (a) TEM grid with 50e-6×50e-6 m windows, (b) an image of TEM window from SMS nanoscope, (c) a zoom-in view (1.5e-6×1.5e-6 m window) of a gold nanorod from the SMS nanoscope. Color bar is an arbitrary scale that corresponds to the light transmission change due to the absorption of the light by the imaged particle. (d) 3 TEM images of single gold nanorods coated with silica shells.

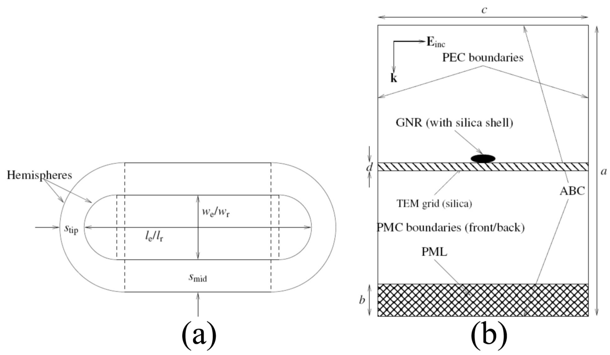


Figure 2: (a) Schematic of single nanorod, showing the various dimensions obtained from TEM imaging; (b) a 2D projection of the 3D geometry and boundary conditions of the computational model. The model was truncated at the top and bottom using an absorbing boundary condition (ABC), at the left and right with a perfect electric conductor (PEC), and at the front and back with a perfect magnetic conductor (PMC). The incident wave was polarized on the left-right direction, travelling downward. Therefore an additional absorbing layer, known as a perfectly matched layer (PML) was placed at the bottom of the domain. The medium inside the domain was air (n = 1), and the TEM grid was modeled as a silica layer (n = 1.46). In the computational model a = 2.4e-6 m, b = 0.4e-6 m, c = 1.4e-6 m, and d = 40 nm. The domain in the front-to-back direction had a size equal to c.

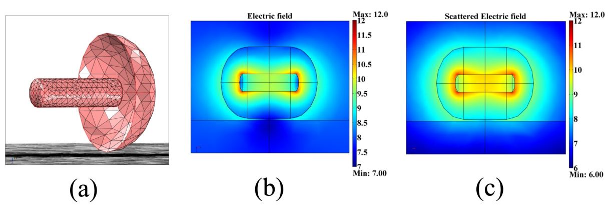
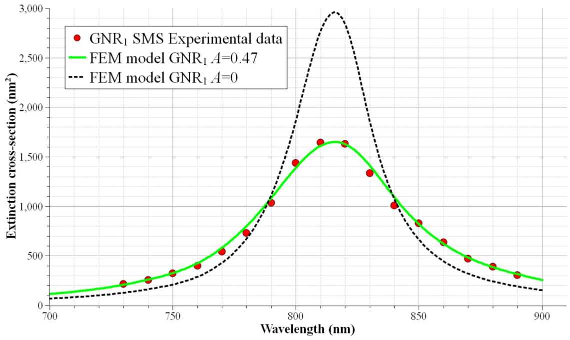
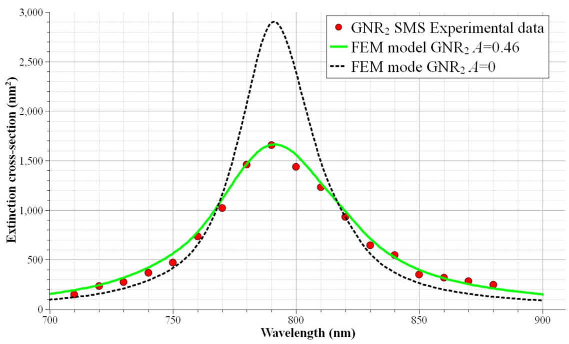


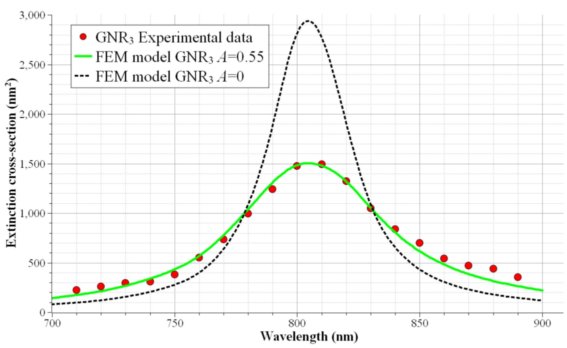
Figure 3: Single silica coated GNR model. (a) Mesh quality; (b) Electric field distribution (log|E|); (c) Scattered Electric field distribution (log|Escal).



(a)



(b)



(c)

Figure 4: Experimental and simulated extinction cross-section of three silica coated GNRs.