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The Importance of Surface Sensitivity in the Optimization of Plasmonic Sensors

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Summary:

In this work the importance of optimizing the surface sensitivity of nanoplasmonic sensors is discussed. In localized surface plasmon resonance (LSPR) based sensing, the size, shape and arrangement of the used nanoparticles all affect their sensitivity. We will demonstrate that by careful design and near-field engineering, the plasmon decay length of nanoparticle-arrangements can be fine-tuned, taking into account the size of receptor and target molecules used in the different sensing scenarios. By optimizing the surface sensitivity of the sensors, their performance in small molecule detection can be maximized.

Keywords: Localized Surface Plasmon Resonance; Refractive Index Sensitivity; Surface Sensitivity; Modelling adsorption interfaces.

Background

One of the main challenges of fabricating plasmonic nanostructures for chemical and biosensing applications is optimization of the structures in order to maximize their sensitivity. The most widely used parameter for benchmarking the fabricated sensors is the bulk refractive index sensitivity (*RIS*), which relates the spectral response ($\Delta\lambda_p$) to the RI changes in the medium surrounding the nanoparticles (Δn_b), as defined in Eq. 1 [1].

$$RIS = \frac{\Delta\lambda_p}{\Delta n_b} \tag{1}$$

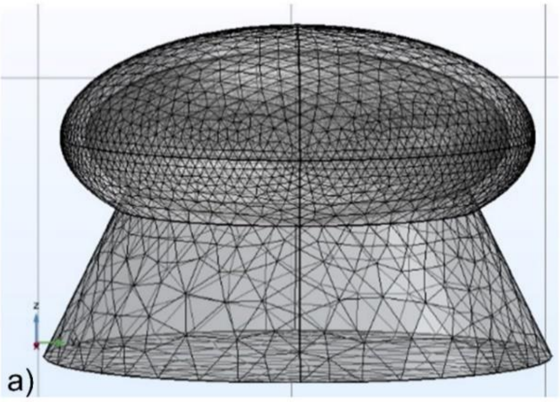
However, for small molecule sensing applications the surface sensitivity, that characterizes the spectral response in the function of the deposited layer thickness can be more meaningful. The surface sensitivity is usually evaluated by using Eq. 2, where nl and t are the refractive index and thickness of a deposited layer on the nanoparticle's surface, and ξ_D is the plasmon decay length [2].

$$\Delta\lambda_p = RIS(n_l - n_b)\left(1 - e^{\frac{-2t}{\xi_D}}\right) \tag{2}$$

Surface sensitivity is primarily depending on the plasmon decay length around the particles, affected by their size and shape and strongly influenced also by plasmonic coupling.

Description of the Used Methods

Surface sensitivity can be determined experimentally by using subsequential chemical deposition tools, such as the layer-by-layer (LbL) method, that uses positively and negatively charged molecules to create layers with increasing thickness and thus map the plasmon decay length of the particles. If the geometry of the used nanoparticles is known numerical simulations can also be used to map the response of the nanoparticles, as illustrated in Fig. 1.



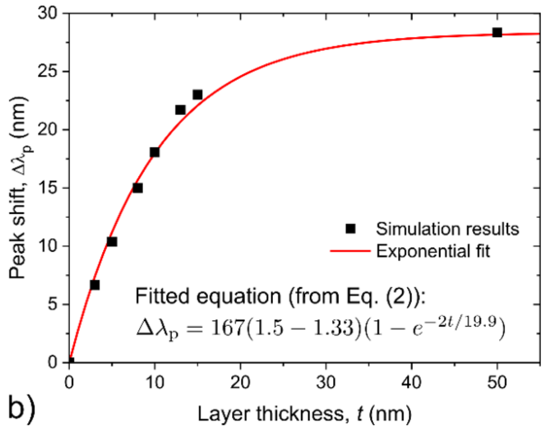


Fig. 1/a: The structure and mesh of the hexagonal (periodic) unit cell for modeling the response and surface sensitivity of densely-packed gold nanoparticle arrays. In the simulations a growing thickness of layers with known refractive index (n_l=1.5) is deposited onto the nanoparticles in water medium (n_b=1.33) [3]. b) The calculated peak shift values as a function of the deposited layer thickness. By fitting Eq. 2 on the dataset (with known RIS) the plasmon decay length of the structure can be obtained (19.9 nm in this case) [3].

Results

In this talk, the importance of optimizing the surface sensitivity of plasmonic nanostructures will be highlighted. It will be demonstrated through numerical simulations that if the size of the receptor-target molecules is known, the field decay length can be optimized to maximize the signal of target binding, through the interparticle gap engineering of densely-packed nanoparticle arrangements (see Fig. 2).

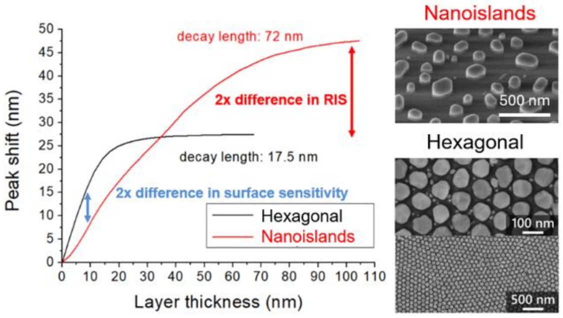


Fig. 2: Experimental comparison of the response of two realized nanoparticle arrangements. Top right, red: gold nanoislands on glass, created by sequential thin film deposition (sputtering) and solid-state dewetting. Bottom right, black: hexagonally arranged ellipsoidal gold nanoparticles, created by using a porous alumina template, thin film deposition and dewetting [4].

Fig. 2 demonstrates that while the gold nanoislands on glass (red curve) have a two-times higher bulk refractive index sensitivity compared to the hexagonally arranged gold nano-ellipsoids, the latter structures have significantly higher surface sensitivity. E.g. for binding a thin layer with only 10 nm thickness on the surface of the sensors, the hexagonal particle systems respond with a peak shift that is twice that of the nanoislands. This property is ultimately important for small molecule detection.

Two case studies will also be presented to illustrate the effect of layer formation on the interpreted signal during gas sensing and fluid refractive index sensing applications [3]. It will be demonstrated that during both gas sensing and fluid RI sensing, structures that have high surface sensitivity are increasingly affected by layer formation on their surface originating from the adsorption of gas molecules or ions (respectively for the different applications). Considering these layers during the sensing applications is essential for the correct interpretation of sensor signals.

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