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Sensitivity comparison of localized plasmon resonance structures and prism coupler

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ABSTRACT

Plasmon resonances are widely used in biomolecular sensing and continue to be an active research field due to the rich variety of surface and measurement configurations, some of which exhibit down to single molecule level sensitivity. The resonance wavelength shift of the plasmonic structure upon binding of molecules, strongly depends, among other parameters, on how well the field of the resonant mode is confined to the binding site. Here it is shown that, by using properly designed metal-insulator-metal type resonators, improved wavelength response can be achieved with localized surface plasmon resonators (LSPRs) compared to that of the commonly used Kretschmann geometry. Using computational tools we investigate theoretically the refractive index response of several LSPR structures to a 2 nm thin film of binding molecules. LSPR resonators are shown to feature improved sensitivity over conventional Kretschmann geometry in the wavelength interrogation scheme for such a thin film. Moreover, some of the LSPR modes are quasi-omnidirectional and such angular independence (up to 30° angle of incidence) allows higher numerical apertures to be used in colorimetric imaging. Results highlight the potential of LSPRs for biomolecular sensing with high sensitivity and high spatial resolution.

1. Introduction

Resonance excitation and detection of Surface Plasmon Polaritons (SPPs) provide a versatile platform for affinity based refractometric biomolecular sensing [1,2]. Typically, the refractive index profile near a metal surface is modified due to the refractive index contrast between a buffer solution and attached biomolecules and the resonance of the plasmonic modes are shifted frequency, which can be monitored using reflection or transmission measurements. Due to the ease of far field measurement and the simplicity of fabricating surfaces exhibiting plasmonic resonances, surface plasmon resonance (SPR) technique has been extensively used over the last few decades for biomolecular sensing [3–5]. Commonly, prism and grating coupling have been used to excite SPPs on planar or quasi-planar surfaces, and prism coupling is accepted to exhibit higher sensitivity [6]. The advent of nanofabrication tools allowed more and more top-down structures to be employed in plasmonic sensing. The rich physics of single or coupled plasmonic structures allows high performance biomolecular sensing using ordered periodic localized plasmon mode resonances (LSPR) [7–9]. Typically, SPR imaging allows simultaneous detection from multiple individually functionalized spots. As prism coupled SPR has a strong angular dependence of the resonant excitation, a low numerical aperture has to be used, and SPR imaging has been inherently a low spatial resolution technique [10,11]. In order to perform plasmon resonance imaging with high spatial resolution, it is desirable to have plasmonic surfaces that are not sensitive to angle of excitation (omnidirectionality), while maintaining a high refractive index sensitivity, thereby allowing sensing of monolayer biomolecular coatings. Periodic localized plasmon structures can be designed with optical band structures which exhibit quasi-omnidirectional response [12].

In this article, we explore theoretically the refractive index sensitivity of several plasmonic designs. Typically, a simple figure-of-merit (FOM), defined as the ratio of sensitivity (shift of the resonance wavelength in nm per change in the bulk refractive index of the sensing medium) to the full-width-at-half-maximum (FWHM) of the resonant line shape, is used to compare the performance [13,14]. We note that, in biomolecular sensing applications, the performance depends not on bulk refractive index change but on molecular binding. Therefore, instead of using the bulk refractive index change for the comparison of sensitivity of structures with that of a prism, a thin (2 nm thick) organic film (representing the adsorbed molecular layer) is used. Here we observe that, when a thin film is considered instead of the bulk refractive index, properly designed localized plasmon resonance sensors exhibit greater sensitivity compared to a prism coupler. Previously, various plasmonic resonators designed or fabricated on planar surfaces have been

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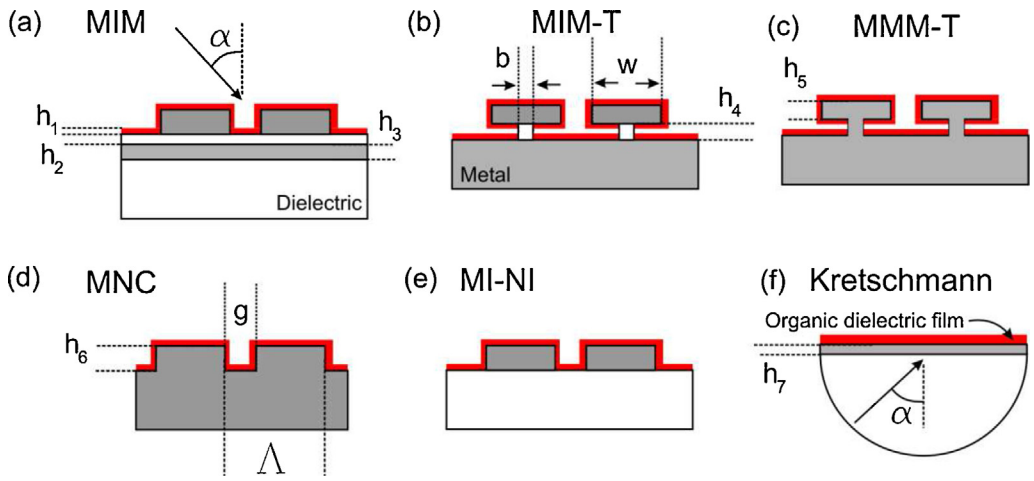

Fig. 1. Schematic cross section of plasmonic structures analyzed in this work. (a) A metal-insulator-metal (MIM) metasurface with a thin organic layer coated on top. (b) The MIM surface is further processed to partially eliminate the insulator and open up space that will allow binding of molecules in between metal layers (referred to as T-shaped MIM structure or, MIM-T). (c) All-metal version of the MIM-T structure (referred to as MMM-T). (d) Metallic nanocavity (MNC) structures. (e) Metal-insulator nano-islands (MI-NI). (f) Commonly used prism-coupled plasmon resonance geometry.

considered as plasmonic interfaces for LSPR sensing [15,16]. We also consider plasmonic structures on planar surfaces that can be fabricated by top-down methods. We choose geometric structures that are straightforward to implement using standard nanofabrication techniques such as electron beam lithography. Particularly, we investigate the structures shown in Fig. 1. The structures include a metal-insulator-metal (MIM) metasurface with a thin organic layer coated on top (Fig. 1a). The MIM surface is further processed to partially eliminate the insulator and open up space that will allow binding of molecules in between metal layers (referred to as T-shaped MIM structure or, MIM-T, Fig. 1b). An all metal version of the MIM-T structure, referred to as MMM-T is shown in Fig. 1c. In addition to MIM structures, a metallic nanocavity (MNC) array is studied (Fig. 1d). The MNC is similar to a lamellar grating, however the rectangular grooves are geometrically designed to feature broad localized plasmon modes confined primarily into the groove, therefore serving as plasmonic nanocavities. The MNC layer structure allows fabrication by a simple nanoimprinting process. Also a metal-insulator nano-island (MI-NI) array is considered (Fig. 1e). Such structures can be fabricated by top-down fabrication techniques such as electron beam lithography or the geometry can be used to approximate self-organized metal nanoislands, typically formed by dewetting of thin metal films on glass substrates [21]. We compare the surfaces with the commonly used prism-coupled plasmon resonance geometry (Fig. 1f).

2. Sensing by wavelength interrogation of plasmon resonance

The propagation constant k_{SPP} of the surface plasma wave propagating at the interface between a semi-infinite dielectric medium and a metal is given by [1]

$$k_{spp} = \frac{\omega}{c}\sqrt{\frac{\varepsilon_m \varepsilon_{do}}{\varepsilon_m + \varepsilon_{do}}} \geq k_0 = \frac{\omega}{c}\sqrt{\varepsilon_{do}} \tag{1}$$

where $\varepsilon_d = n_{do}^2$ and $\varepsilon_m = n_m^2$ are the dielectric functions of the dielectric and the metal, ω is the frequency and $k_o = 2\pi/\lambda_0$ is the free space wave vector. Since k_{SPP} is larger than k_0, free space resonant coupling to such a mode is not possible and a prism of refractive index $n_p > 1$ is generally used to couple free space light into the SPP mode. Resonant coupling takes place at an angle of incidence θ, when

$$k_{SPP} \cong n_p k_o \sin(\theta) \tag{2}$$

In the prism coupled scheme (Kretschmann geometry), a thin dielectric film, of refractive index n_d and thickness h, shifts the resonance according to [1]

$$\delta k_{SPP} \cong \frac{K_{SPP}^3}{K_0^2 n_{do}^3}[1 - \exp(-2\gamma_d h)]\delta n_d \tag{3}$$

where $\gamma_d = ik_o\varepsilon_{do}/(\sqrt{(\varepsilon_m + \varepsilon_{do})})$ and $h \ll 1/\mathrm{Re}\{\gamma_d\}$ and $\delta n_d = n_d - n_{do}$, n_{do} being the bulk refractive index of the dielectric medium. In LSPR sensing using an isolated plasmonic resonator where the mode field can be assumed to isotropically decay into the surrounding medium, the wavelength shift upon binding is given similarly by $\delta\lambda = S_\lambda \delta n_d(1 - \exp(-2h/1_d))$, where $s\lambda$ is the sensitivity factor (shift in resonance per RIU change in environment refractive index) and l_d is electromagnetic decay length of the LSPR mode [8–17]. However, for anisotropic resonators, such as prism or disk shaped, the mode field is non-uniformly distributed on the resonator and sensitivity of binding assumes a location dependent form. The same is true for resonators bound to a substrate, where only one side of the resonator is available for molecular binding. For a general plasmonic resonator including coupled resonators and cavities in buffer or on a surface, a simple equation including a single decay length cannot be used to express the wavelength shift. Instead, perturbation theory can be used to estimate the resonance wavelength shift upon a change in the refractive index profile through [18]

$$\frac{\delta\lambda_{res}}{\lambda_{res}} = -\frac{1}{2}\frac{\int |E(r)|^2 (n_d^2(r) - n_{do}^2) dv}{\int |E(r)|^2 dv} \tag{4}$$

where $E(r)$ is the electric field distribution (mode profile), n_{do} is the refractive index profile before perturbation, $n_d(r)$ is the refractive index profile after the perturbation and integration is performed over all space.

Sensitivity for wavelength interrogated detection is [6]

$$S_\lambda = \frac{\delta\lambda_{res}}{\delta n_d} \tag{5}$$