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Numerical evaluation of periodic nanowire-based phase sensitive surface plasmon resonance detection

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ABSTRACT

Localized surface plasmon (SP)-based amplification of sensitivity in a phase-sensitive SP resonance (SPR) biosensor is investigated numerically. SP is localized by surface-relief periodic nanowires. An optimum sample obtained by modeling DNA hybridization and water–glycerin mixture employed a gold grating with 300 nm period and 10 nm thickness on a gold film and an SF10 glass substrate. The localized SPR structure was shown to have the sensitivity enhanced by more than 10 times than conventional SPR for detecting DNA hybridization at an identical film thickness of 40 nm. The enhancement was found to be associated with field localization near the nanostructure. The results suggest a novel approach of improving the sensitivity of SPR detection.

1. Introduction

Surface plasmon (SP) is a longitudinal wave of electron concentration that is formed across a dielectric metal interface when TM polarized light is incident. SP resonance (SPR) condition varies in response to biomolecular interactions that occur on the metal surface. Because of the capability of label-free detection of refractive index changes in real-time, SPR has been used in biosensing extensively.

Recently, intensive efforts have been made to improve the sensitivity limit of SPR: for example, through signal amplification using functionalized nanoparticles [1], magneto-optic effects [2], nanostructure mediated localized SPR (LSPR) based on periodic nanowires and nanoposts [3–6], and phase-sensitive SPR detection schemes [7,8]. In particular, nanostructure-based LSPR, which is typically characterized by intensity measurement of periodic surface-relief corrugation patterns on the metallic film, enhances the sensitivity by means of creating local hot spots as well as increased reaction surface area. It was also found that the sensitivity enhancement depends on the biointeraction itself [9] and can be obtained additionally by localizing the target in the hot spots [10]. On the other hand, the detection of phase changes that accompany a biomolecular interaction was reported as a way to provide significant sensitivity improvement by using conventional thin film-based SPR biosensors, although dynamic range of phase-sensitive SPR detection is somewhat limited. An obvious next step to this overall direction is to improve the sensitivity by employing a plasmonic nanostructure in a phase detection scheme.

In this paper, we intend to achieve enhanced SPR detection sensitivity by measuring phase changes of a biomolecular interaction on plasmonic nanostructures that consist of periodic nanowires. In other words, an inherent motivation for this work is that a nanoplasmonic structure may amplify SPR intensity shifts as well as phase changes as a result of an interaction. The amplification may be associated with the presence of hot spots of localized electromagnetic waves created by the nanostructure.

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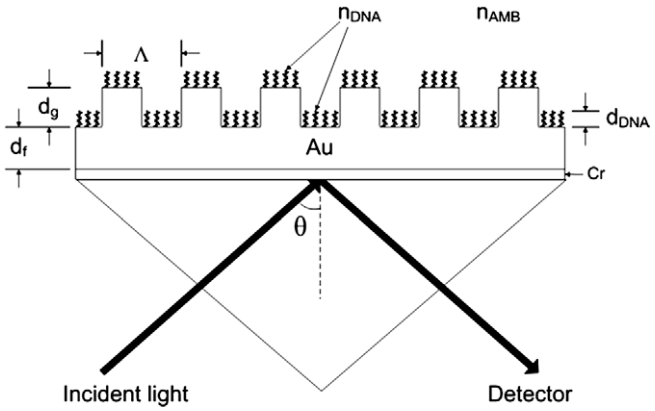

Fig. 1. Schematics of (a) DNA hybridization and (b) water–glycerin mixture on a periodic nanowire sample considered in this study.

2. Numerical methods

Sensitivity enhancement factor (SEF) has been used to evaluate the improvement of sensitivity. SEF may be defined as the ratio of $|\partial\varphi/\partial n|_{\mathrm{LSPR}}$ to $|\partial\varphi/\partial n|_{\mathrm{SPR}}$, where $\partial\varphi/\partial n$ denotes a differential phase change for a target index change in the case of phase-sensitive nanostructure mediated LSPR or a conventional thin film based SPR structure. The most widely available SPR system measures resonance angle changes in intensity. In this regard, sensitivity enhancement may be evaluated in terms of the ratio of $\partial\varphi/\partial n|_{\mathrm{LSPR}}$ to $\partial\theta_{\mathrm{SP}}/\partial n|_{\mathrm{SPR}}$. However, this ratio is relatively unclear to evaluate the degree of enhancement, because φ and θ_{SP} are not explicitly related quantities.

Phase dependence of a conventional SPR biosensor can be easily obtained from Fresnel coefficients. In the case of nanostructure-based LSPR, phase dependence is difficult to obtain analytically. One may take advantage of effective medium theory (EMT) for an effective permittivity of a nanostructure and then calculate Fresnel coefficient for a four-layer system. However, EMT tends not to provide converged data for metallic nanostructures and was, in fact, reported with significant disparity from exact results, particularly in the regime of strong plasmon resonance [11]. Therefore, we used rigorous coupled wave analysis (RCWA) for numerical calculation of phase variation in nanostructure-based LSPR biosensors, which has been successfully employed to simulate optical characteristics of periodic structures [12,13]. Twenty spatial harmonics was used to calculate the results. In addition, finite difference time domain (FDTD) method was used under periodic boundary condition to investigate the near-field characteristics at resonance.

We designed an optimum structure that provides the highest SEF for detecting two distinct targets, i.e., DNA hybridization process from single-stranded (ssDNA) to double-stranded DNA (dsDNA) and also a mixture of glycerin and pure water. The former represents the biomolecular interactions that form layers, while the latter corresponds to an overall ambient change. The calculation of DNA hybridization is based on the model shown in Fig. 1 with incident light wavelength at $\lambda = 632.8$ nm and parameters of ssDNA and dsDNA obtained from Ref. [14]. We assumed using 12-mer oligonucleotides so that the thickness of the film in the course of DNA hybridization is equal to $d_{\mathrm{DNA}} = 4$ nm. Optical parameters for metals (Au and Cr) were obtained from Ref. [15]. For modeling water–glycerin mixture, we assumed it as a change in ambience, which is identical to Fig. 1 except for the absence of the DNA layer and ambient refractive index changes. For all structures, it was assumed that metal film thickness $d_{\mathrm{f}} = 40$ nm on a substrate of SF10.

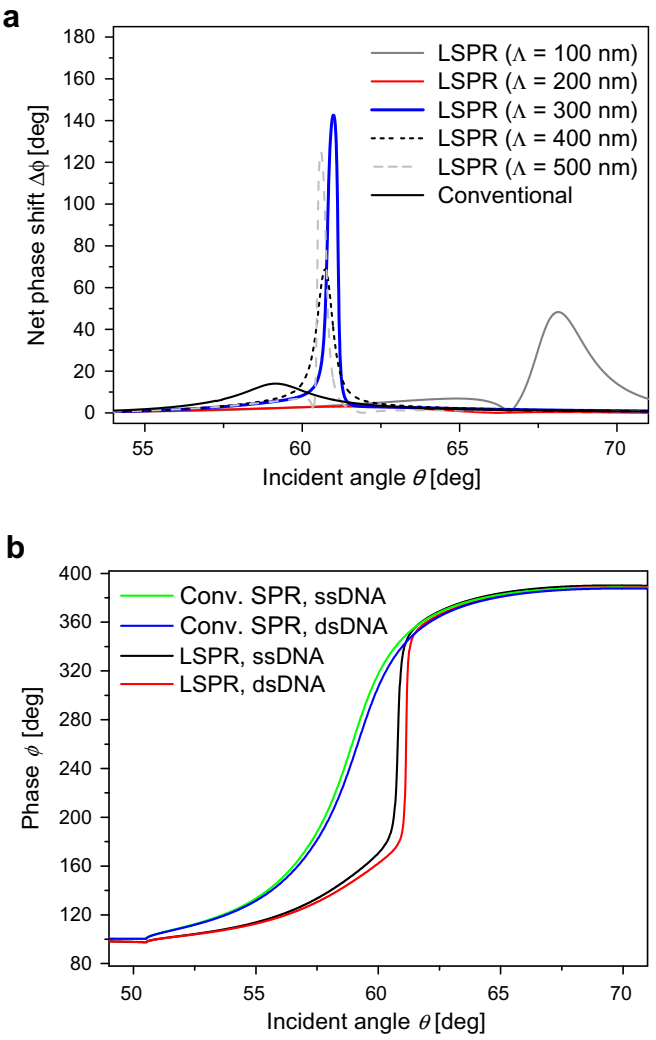

Fig. 2. (a) Net phase variation ($|\Delta\varphi|$) as a result of DNA hybridization for LSPR structures of various nanowire periods (Λ) in comparison with a thin film based conventional SPR structure. (b) Phase variation (φ) of resonance characteristics due to DNA hybridization for nanostructure-based LSPR at $\Lambda = 300$ nm and $d_g = 10$ nm and also conventional SPR.

3. Results and discussion

Fig. 2a shows net phase variation ($|\Delta\varphi|$) as a result of DNA hybridization as a function of incidence angle for LSPR structures of various structural parameters such as period (Λ) and thickness (d_g) in comparison with thin film based conventional SPR structures. The results indicate that the largest phase change is obtained with an LSPR structure at $\Lambda = 300$ nm and $d_g = 10$ nm. Compared to conventional SPR at $d_f = 40$ nm, SEF = 10.2. In other words, nanostructure-based LSPR may achieve sensitivity enhancement by an order of magnitude. Maximum sensitivity enhancement is obtained at $\theta = 60.99°$ for an LSPR biosensor with $\Lambda = 300$ nm and $d_g = 10$ nm and $\theta = 59.16°$ for conventional SPR at $d_f = 40$ nm. However, sensitivity enhancement is reduced to SEF = 4.6 if an attachment layer of chrome is considered. Fig. 2b represents the phase variation (φ) of resonance characteristics due to DNA hybridization for nanostructure-based LSPR at $\Lambda = 300$ nm and $d_g = 10$ nm and also conventional SPR. The difference of φ at any incidence angle corresponds to the net phase variation ($|\Delta\varphi|$) shown in Fig. 2a. The slope $\partial\varphi/\partial\theta$ near resonance angle is an indicator of sensitivity.

The net phase variation ($|\Delta\varphi|$) for water–glycerin mixture was calculated with design parameters that are similar to those of the