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Polarization-sensitive surface plasmon enhanced ellipsometry biosensor using the photoelastic modulation technique

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ABSTRACT

A surface plasmon enhanced ellipsometry (SPEE) biosensor scheme based on the use of a photoelastic modulator (PEM) is reported. We show that the polarization parameters of a laser beam, tan ψ, cos Δ and ellipse orientation angle ϕ, can be directly measured by detecting the modulation signals at the first and second harmonics of the modulated frequency under a certain birefringence geometry. This leads to accurate measurement of refractive index variations within the evanescent field region close to the gold sensor surface, thereby enabling biosensing applications. Our experimental results confirm that the new scheme offers a respectable detection limit of 6×10^{-7} refractive index unit (RIU) or 15 ng/ml of biomolecule solute concentration without any compromise in dynamic range. In addition, PEM offers the possibility of single-beam phase-sensitive SPR detection that drastically reduces the complexity of the optical system, thus readily making it possible for SPR to be adopted in label-free micro-array biochips.

1. Introduction

In recent years, surface plasmon resonance (SPR) has been going through continuous advancement in terms of sensitivity, sample throughput and areas of application [1–5]. Compared to other non-labeling biosensing techniques such as those using resonance mirrors [5] and cantilevers [6], SPR is gaining increasing acceptance in research areas related to biomolecular interaction analysis (BIA) because of the ever-growing need to understand and treat diseases [7].

Conventional SPR biosensors are generally implemented in a Kretschmann–Raether geometry through which the wave-vector of the incident photons may match with that of surface plasmon polaritons (SPP) so that energy may efficiently couple to a metal/dielectric interface. The resulting SPR effect is observed as a dip in angular or spectral interrogation of the reflected light intensity [7,8]. As biomolecular interaction events will lead to an increase in refractive index or thickness of the organic layer adsorbed by the gold sensor surface, which can in turn change the conditions for wave-vector matching, any biomolecular binding reaction taking place in the sensor surface will therefore result in a shift in the SPR dip both in the angular and spectral domains.

Until now, commercial SPR biosensor systems are predominantly based on the angular or intensity interrogation scheme, and their typical limit of detection is still not comparable with the level achievable by fluorescence tagging techniques, e.g. ELISA, Western blot [1,7], which can produce a signal associated with single molecular events. Despite of its less favorable sensitivity limit, SPR is still very attractive to the biomolecular research community because of its label-free and real-time quantification attributes. The continual pursue for improvement in detection limit is always on. Recently it has been reported that phase-sensitive SPR is a promising approach for improving the limit of detection by up to two orders of magnitude [9]. However, the relatively less attractive side of phase-sensitive SPR schemes is that they typically offer limited dynamic range because of the fact that steep phase change occurs only in a small refractive index range within the plasmon resonance dip [7,8].

In this paper, we report that indeed one can use ellipsometry to achieve high performance phase-sensitive SPR. Ellipsometry, being an established technique for simultaneously detecting the intensity and phase of light upon reflection is usually described by the complex ellipsometry equation: $\rho = r_p/r_s = \tan\psi \exp(i\Delta)$, where r_p and r_s are the electric field reflection coefficients for p- and s-polarization components. [10,11]. As it has been well established that surface plasmons (SP) can drastically enhance the electrical field intensity near the metal surface, using SP to enhance ellipsometric sensitivity is a natural choice. The concept of total internal reflection ellipsometry (TIRE) [12] or surface plasmon enhanced

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ellipsometry (SPEE) [13,14] has been proposed to combine the advantages of both SPR and ellipsometry to measure $\tan\psi$ and $\cos\Delta$, or more common intensity and phase variations. Until now, most of the reported cases employed a commercially available ellipsometer. Moreover, although TIRE or SPEE can offer as high as 10–100 times improvement in terms of detection sensitivity as compared to that of traditional ellipsometry, a common drawback is their limited dynamic range, especially when used in biosensors, while other non-ellipsometric phase-sensitive systems also suffer the same problem [12–14]. The common solution to this problem is to reduce the thickness of the Au or Ag sensor layer, typically from 50 nm to 25–30 nm, in order to achieve a reasonable compromise between sensitivity and dynamic range [12–14].

In this paper, we propose a new surface plasmon enhanced ellipsometry biosensor scheme that makes use of a photoelastic modulator (PEM) to temporally modulate the phase of the sensing beam in order to enable direct measurement of the polarization state through detecting the first and second harmonics of the modulated frequency under certain birefringence geometry. SPEE also collects information from the variations in both intensity and phase, i.e. $\tan\psi$ and $\cos\Delta$, of the reflected laser beam. Recently, phase-sensitive SPR detection based on polarimetry using a photoelastic modulator has been demonstrated to offer wide dynamic range and ultra-sensitive response [15,16]. Through detecting the second and third harmonic signals of the modulated frequency, this approach effectively measures the intensity as well as the SPR phase simultaneously. However, this is an indirect polarimetric technique which does not provide direct data on $\tan\psi$ and $\cos\Delta$ for further quantitative analysis. Here we show that under a certain birefringence geometry SPEE provides direct measurement on the polarization state of the p- component, which is sensitive to the SPR effect, and the s-component, which is unaffected by SPR. Experimental results confirm the expected improvement in detection limit while maintaining a reasonably wide dynamic range. Also, the new scheme has the merit of being less sensitive to external stray light and intensity fluctuations of the laser beam in comparison to conventional SPR schemes as well as the aforementioned phase-sensitive PEM systems.

2. Methodology and experimental setup

Fig. 1 shows a schematic diagram of our experimental SPEE set up. The light source is a 10 mW He–Ne laser operating at the wavelength of 632.8 nm. The beam passes through a quarter-wave plate and a polarizer to obtain a 45° linearly polarized laser beam. Then the laser beam is directed through the sensor head, which is made from a 60° equilateral prism coupler (BK7) coated with an Au layer (50 nm in thickness), interfacing with an aqueous dielectric media trapped in a flow chamber. The angle of incidence at the prism/Au interface is chosen such that maximum SPW coupling efficiency is achieved at Au/dielectric interface. Accompanying with the SPR effect is a drastic decrease of intensity and a sharp phase jump in the p-polarization component, while leaving the s-polarization component unchanged. All these will result in a change in the total polarization state of the optical beam. The reflected light beam then heads to a photoelastic modulator, whose fast axis is oriented at 45° from the *X*-axis in the *XY* plane. The PEM produces a sinusoidally modulated polarization state at a frequency, ω = 42 kHz. An analyzer with its orientation at 0° from the *X*-axis in the *XY* plane is placed between the PEM and the silicon detector. An amplifier is employed to separate the AC and DC components in the detector signal before they are fed to a lock-in amplifier with its external reference locked to the modulation frequency of the PEM. With all the necessary information already embedded in the frequency domain of the detected signal, the polarization state is extracted by analyzing the first and second harmonics of the modulated signal as well as the DC component simultaneously. In previous work we reported that the influence of residual birefringence to phase retardation of PEM [15,17]. In order to minimize this effect, the setup is located in an enclosed environment to ensure minimum temperature fluctuation caused by air circulation and proprietary frequency generator is used to provide stable modulation frequency.

The change of polarization state the system can be analyzed using Jones matrix transformation [10,11,16]. Since of the light is linearly polarized before impinging the SPR sensor head, one can describe the electric field components after SPR coupling as:

$$E_{\mathrm{spr}} = \begin{bmatrix} E_{\mathrm{P}} \\ E_{\mathrm{S}} \end{bmatrix} \quad \text{with} \quad E_{\mathrm{p}} = |a_{\mathrm{p}}|\exp(i\theta_{\mathrm{p}}), E_{\mathrm{s}} = |a_{\mathrm{s}}|\exp(i\theta_{\mathrm{s}}) \tag{1}$$

where a_s and a_p are the amplitudes of s- and p-polarization components, and θ_s, θ_p are their respective phase values. It should be noted that the amplitude and phase of the s-component do not change significantly by the SPR effect. Whereas the p-component experiences dramatic changes in both the amplitude and phase, which results in a shift in the overall polarization state as the light beam reflects off the sensor head. If we include all the parameters of the s- and p-components, the polarization state can be denoted by an ellipse orientation angle ϕ (or elongation), and ϕ can be calculated using the equation $\phi = 1/2\,\mathrm{arc}\left\{\tan[(2a_s a_p)/((a_s)^2 - (a_p)^2)\cos(\theta_s - \theta_p)]\right\}$ [10,11,18]. In the proposed scheme, we fix the system at given birefringence geometry, and the orientation angle (ϕ) is found directly using an ellipsometer constructed from a PEM and a set of polarization analyzers.

With its aperture plane oriented at 45° from the *X*-axis in the *XZ* plane, the PEM generates modulation in both the s- and p-polarization components of laser beam at a frequency ω. The periodic modulation of the polarization state can be represented by a Jones matrix as shown here [10]:

$$J_{\mathrm{PEM}} = \begin{pmatrix} \cos\left(\frac{\delta}{2}\right) & i\sin\left(\frac{\delta}{2}\right) \\ i\sin\left(\frac{\delta}{2}\right) & \cos\left(\frac{\delta}{2}\right) \end{pmatrix} \tag{2}$$

Here $\delta = A\cos(\omega t)$, A is the modulation amplitude of PEM, δ is the relative phase retardation of between the two orthogonal polarization components.

The phase modulated sensing beam passes through an analyzer with its axis set to 0°. The Jones matrix of the analyzer is:

$$J_{\mathrm{p}} = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \tag{3}$$

The finial intensity of the laser beam after having passed through the analyzer becomes:

$$I = E^{*}E \quad \text{where} \quad E = J_{\mathrm{p}} J_{\mathrm{PEM}} E_{\mathrm{spr}} \tag{4}$$

Consequently, for the resultant intensity we have:

$$I = \frac{1}{2}\left[a_s^2 + a_p^2 + (a_s^2 - a_p^2)J_0(A) - 2J_1(A)(2a_s a_p\cos(\varphi))\cos(\omega t) - 2J_2(A)(a_s^2 - a_p^2)\cos(2\omega t) + \cdots\right] \tag{5}$$

where $\varphi = \theta_s - \theta_p$, and J_0, J_1, J_2 are spherical Bessel functions of first kind.

Using a lock-in amplifier, the harmonics of the modulation signal can be detected, particularly with the help from Fourier transformation. Respectively the DC and first two harmonics signal are:

$$H_{\mathrm{DC}} = a_s^2 + a_p^2 + (a_s^2 - a_p^2)J_0(A) \tag{6}$$

$$H_1 = 2a_s a_p\cos(\varphi)J_1(A) \tag{7}$$

$$H_2 = (a_s^2 - a_p^2)J_2(A) \tag{8}$$