Phase-sensitive silicon-based total internal reflection sensor

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Abstract: A concept of phase-sensitive Si-based Total Internal Reflection bio- and chemical sensor is presented. The sensor uses the reflection of light from an internal edge of a Si prism, which is in contact with analyte material changing its index of refraction (thickness). Changes of the refractive index are monitored by measuring the differential phase shift between p- and s-polarized components of light reflected from the system. We show that due to a high refractive index of Si, such methodology leads to a high sensitivity and dynamic range of measurements. Furthermore, the Si-based platform offers an easy bioimmobilization step and excellent opportunities for the development of multi-channel microsensors taking advantage of the advanced state of development of Si-based microfabrication technologies.

OCIS codes: (120.5050) Instrumentations, measurement, and metrology; (240.2130) Optics at surfaces.

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1. Introduction

Refractive index (RI) of a physical or chemical substance is a fundamental parameter, which characterizes its optical properties and can give important information on its condition [1]. When examined in organic thin films on the surface, refractive index monitoring can be used to control the thickness of the films and thus follow the course of biological (chemical) binging/recognition events [2]. While refractive index of a bulk medium can be sensitively characterized by a variety of interferometric or other methods [1], the monitoring of RI in thin films is a much more challenging task with a limited number of opportunities in optics. Normally, such monitoring requires evanescent wave excitation, which can be achieved in total internal reflection (TIR) or Surface Plasmon Resonance (SPR) geometry. These methods use features in intensity distribution of light reflected from a dielectric/dielectric or dielectric/metal interface, respectively, to control the refractive index of the adjacent dielectric medium. Since SPR provided a higher reflectivity-based sensitivity due to essentially resonant nature of sensing response [3,4], it has been much more frequently used in biological and chemical sensing compared to TIR-based implementations [5,6]. Nevertheless, a recent introduction of phase-sensitive methods in SPR [7-9] changed our vision of parameters and conditions of evanescent wave sensing. The main idea here is the use of phase of light reflected from a system as a sensing parameter instead of intensity. Having an extremely sharp jump under SPR, phase can be extracted by interferometry or polarimetry methods to provide an up to two order of magnitude upgrade in sensing sensitivity. In fact, the SPR-based phase-sensitivity show the record to date of sensitivity to refractive index change in thin films [8].

Generally speaking, the phase jump under a refection of light from an optical system is a rather general phenomenon, which does not necessarily require the presence of a SPR-based dip in the reflected intensity. In particular, similar phase jump takes place in a simplest case of light reflection from an external edge of a prism in Total Internal Reflection (TIR) geometry [1]. Since methodologies of phase-sensitive measurements are also rather general, it seems logical to examine phase under TIR as a potential parameter of sensing. However, the phase jump under TIR is known to rather smooth in simplest case of a glass prism configuration, preventing an achievement of high sensitive sensing response [10-14].

In this paper, we show that the phase sensitivity in TIR geometry can be drastically improved by using Si-based TIR geometry. In this case, sensing characteristics become extremely interesting, opening new opportunities for the development of microsensors taking advantage of state-of-art development of Si-based micro fabrication technology. Earlier we showed that similar concept of Si-based platform can be used to miniaturize and integrate SPR-based sensor devices [15-17].

2. Results and discussion

A schematic of the proposed sensor is depicted in Fig. 1(a). Near-infrared light was directed through a hemi-cylindrical prism and reflected from its edge, which was in contact with an analyte solution in a flow cell. The reflected intensity was examined as a function of the incident angle. To simulate sensing effect in the system, we developed a theoretical framework on the basis of total internal reflection model. In our calculations, the data for dispersion characteristics of silicon were obtained from the approximation of experimental dependencies reported by Herzinger et al. [18]. Under conditions of total internal reflection (TIR), at angles of incidence φ above the critical angle $\varphi_c = arcsin\ 1/n$ the phase shifts δ_p, δ_s and phase difference $\Delta = \delta_p - \delta_s$ are given by [1]:

$$\Delta = 2\arctan\frac{n(n^2\sin^2\varphi-1)^{1/2}}{\cos\varphi} - 2\arctan\frac{(n^2\sin^2\varphi-1)^{1/2}}{n\cos\varphi} = 2\arctan\frac{(n^2\sin^2\varphi-1)^{1/2}}{n\sin\varphi\tan\varphi} \quad (1)$$

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At TIR $n_0 > n_m$ and $n = n_0/n_m$ where n_0 and n_m are refractive indices of the incident and ambient (evanescent) media, respectively.

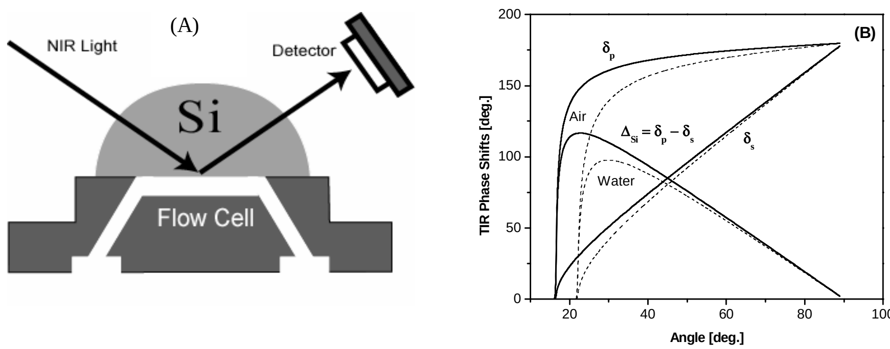

Fig. 1. (a). Schematic of Si-based TIR sensor; (b) Phase shifts for the s- and p – polarized light and differential phase shift for Si prism under TIR (wavelength 1200nm). The results are presented for air and water as ambient media

Figure 1(b) presents calculation results for the phase shifts of s- and p – polarized light components and a differential phase shift under TIR at the Si-air or Si-water interface. Here, we can mention a sharp jump of phase δ_p for p – polarized light from zero up to 150 deg. under a slight change of the angle of light incidence. As shown in the Fig. 1(b), difference of phases Δ of p- and s-components also experiences a strong change reaching 120-130 deg. Although the absolute variation for Δ is lower compared to δ_p , we will later use Δ as the main sensing parameter in order to take advantage of common-path phase-shift interferometry, which enables to easily exclude phase noises caused by different factors such as e.g., temperature drifts, mechanical vibrations, light-source fluctuations etc.

Figure 2(a) presents differential phase shifts for the three prism materials: Si (n_{Si} = 3.525, λ = 1200nm); glass SF11 (n_{SF11} = 1.779, λ = 633nm); glass BK7 (n_{BK7} = 1.515, λ = 633nm). Calculations were performed for water and typical biological medium with refractive indices 1.33 and 1.4, respectively. It is clear that due to a higher RI, Si provides much larger and sharp variation of Δ compared to glasses (100 deg. compared to 35 deg. and 15 deg. for SF11 and BK7, respectively), suggesting a higher sensitivity and wider dynamic range of Si-based scheme to refractive index changes.

To estimate the efficiency of the system in sensing, it is important to examine several characteristics such as the sensitivity, dynamic range, maximal measurable value of RI and finally the simplicity of experimental set-up. To calculate these parameters we have performed the theoretical modeling of phase shifts for s- and p- components under Fresnel interface reflection [Eq. (1)]. Here, incident angles and refractive indexes of tested media were main variable parameters.

Angular dependences of differential phase for different RI of the ambient media are shown in Fig. 2(b). One can clearly see a gradual decrease of maximal differential phase and a shift of a critical angle φ_c as the refractive index of the medium increases from 1.33 (water) to larger values close to n_{Si}. Here, for relatively low RI close to 1.33 the differential phase shows a sharp jump with a total change of the magnitude reaching 110° deg. Such phase characteristics give a promise for using phase as a parameter of sensing similar to how it was done in conditions of Surface Plasmon Resonance [8-10]. In this case, two phase-sensitive methodologies can be applied to monitor the refractive index of the tested medium n_m: (i) determination of an angular position of the maximum of differential phase (line A); (ii) control of the differential phase at a fixed angle of incidence (line B) close to the φ_c. It is

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evident that the first methodology must provide a wide dynamic range of refractive index measurements, while the second one promises a better sensitivity.

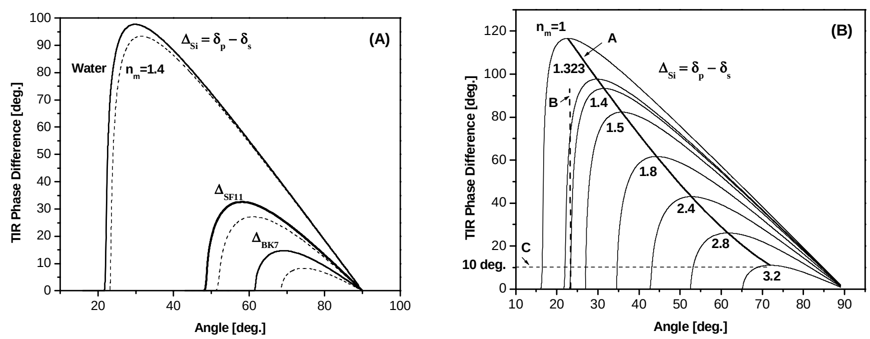

Fig. 2. (a). Differential phase shifts for three prism materials: Si, SF11 and BK7 glasses. Solid and dashed lines show results for water and medium with $n_m = 1.4$, respectively; (b) Differential phase in Si-based TIR configuration for different refractive indices of the external medium (n_m)

Considering the first method based on the analysis of line A, the maximum differential phase shift occurs at the angle ϕ_m where $\partial\Delta/\partial\varphi = 0$. Using Eq. (1), one can find [11]:

$$F = \frac{\partial\Delta}{\partial\varphi} = \frac{\cos^2(\Delta/2)(2(1-n^2)\tan^2\varphi + 4)}{n\sin\varphi\tan^2\varphi(n^2\sin^2\varphi - 1)^{1/2}} \tag{2}$$

The latter equation yields to the maximum of Δ_m at $\varphi_m = \arcsin\left(\sqrt{\frac{2}{n^2+1}}\right)$. In this case, the sensitivity of the method can be determined by differentiating the expression for Δ_m: $S_m = \frac{\partial\Delta_m}{\partial n_m} = -\frac{4n_0}{n_0^2 + n_m^2}$.

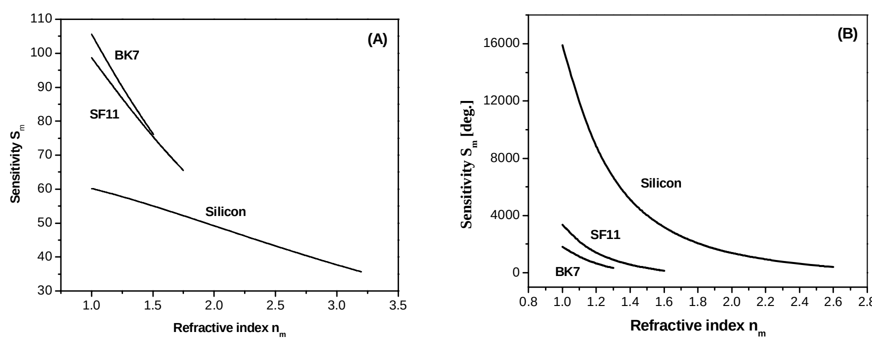

Fig. 3. Calculated sensitivities in cases of maximum differential phase control (a) and a fixed angle of incidence (b).

Figure 3(a) shows the sensitivity S_m as a function of the RI of the external medium for three different prism materials. One can see that despite a slightly lower sensitivity, Si-based configuration provides a much wider dynamic range. In addition, this configuration gives a unique opportunity to characterize high RI media.

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To obtain the sensitivity $S_a = \frac{\partial \Delta}{\partial n_m}$ in the case of the second methodology based on the analysis of phase at a fixed angle [Line B in Fig. 2(b)], we performed a calculation of differential phase as a function of n_m. S_a was determined in points where the phase difference was easily measurable and equal to 10 degree [line C in Fig. 2(b)]. As one can see from Fig. 3(b), high refractive index silicon provided much better sensitivities compared to glasses.

Since the second methodology uses a fixed angle of incidence, we need to assess whether the dynamic range of measurements is wide enough. This parameter was estimated by calculating the change of refractive index Δn_m when the angle of incidence passes from φ_m to φ_c, as shown in Fig. 4(a): $\Delta n_m = n_{\varphi_m} - n_{\varphi_c} = n_m\left(\sqrt{\frac{2}{1+\frac{1}{n^2}}} - 1\right)$. Figure 4(b) presents the separation $\varphi_m - \varphi_c = \arcsin\left(\frac{(\sqrt{2}-1)\sqrt{n^2-1}}{n\sqrt{n^2+1}}\right)$ and dynamic range of Si-based TIR sensor as a function of the ambient RI. One can see that the dynamic range can reach 0.4-0.45 in terms of RI units. In fact, such value is quite sufficient to cover the range of typical changes of RI in biosensing experiments. Another interesting feature is related to the fact that the widest dynamic range is achieved for values of RI close to biosensing conditions (n_m = 1.33-1.42). Such a match is a pleasant surprise granted by particular dispersion properties of Si.

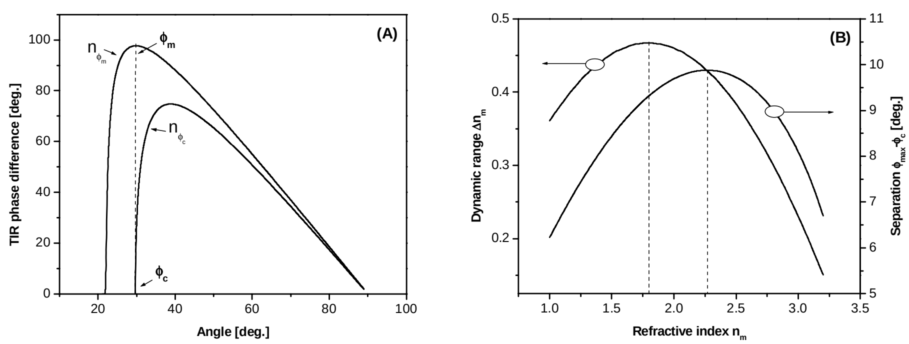

Fig. 4. (a). Methodology for dynamic range estimation; (b) Separation φ_m - φ_c and dynamic range as a function of ambient RI

Thus, theoretical studies show that phase of light reflected in the Si-based TIR geometry can provide a sensitive response to a refractive index change associated with biological binding events on the Si surface. To verify the validity of the proposed sensor concept, we carried out a series of experiments in the TIR geometry, shown schematically in Fig. 1. The TIR coupling system was placed onto a rotary block of a variable angle spectroscopic ellipsometer (Woollam VASE® ellipsometer, J.A. Woollam, Lincoln, NE) to allow for a very fine variation of the angular prism position with respect to the optical path of the ellipsometer. The system was illuminated by a monochromatic 1200 nm light and the reflected beam from the coupling system was analyzed by a detector. The experiments were performed at a fixed wavelength with the resolution of 0.01° in angular measurements.

To study the influence of the ambient RI on the phase difference Δ_m, we brought the sensing surface into contact with solutions of glycerin of different concentration. The solutions were pumped through the channel of the flow cell. Figure 5(a) shows angular

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dependences of Δ_m when water was replaced by two glycerin solutions changing the refractive index by $\Delta n_1 = 3.1 \cdot 10^{-3}$ and $\Delta n_2 = 9.3 \cdot 10^{-3}$. One can see that such small variations of refractive index led to a drastic change of phase Δ_m by 8° and 10° deg., respectively.

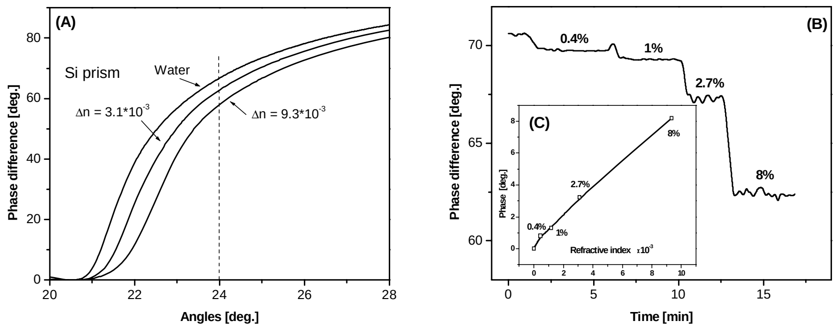

Fig. 5. (a). Phase difference curves for water and two glycerin-water solutions having refractive index difference of $\Delta n = 3.1*10^{-3}$ and $\Delta n = 9.3*10^{-3}$ with respect to water; (b) Real time differential phase measurements of glycerin-water mixtures with various weight ratios; (c) Calibration curve of Si-based TIR sensor.

Figure 5(b) summarizes results of phase shifts for different concentrations of glycerin, while Fig. 5(c) shows the calibration curve for the proposed Si-based TIR sensor. One can see almost linear dependence of the phase on the concentration of glycerin. Another feature is related to a good signal/noise ratio. Taking account the level of noise in the system, we can estimate the detection limit. In our experimental setup, the minimal detectable refractive index variation was about $5*10^{-4}$ in terms of refractive index units. Since this value is mainly limited by temperature drifts in the system, we anticipate further improvements of the sensitivity by at least one order of magnitude through the optimization of the incident angle value and application of active thermostating to better control temperature of solutions. In this case, the sensitivity of Si-based TIR will become comparable with sensitivities of amplitude-sensitive Surface Plasmon Resonance devices. It should be noted that the proposed sensor have some advantages over SPR technology. First, methods for bioimmobilization are much easier and cheaper for the Si/SiO_2 surface compared to gold. In particular, a variety of silanization methods are well elaborated to functionalize Si. Another advantage consists in a possibility of testing high refractive index materials, including novel liquid- or polymer-based sensing elements. Finally, the use of Si-based platform opens opportunities for an easy miniaturization and integration of the proposed sensor, taking advantage of the advanced state of development of Si-based microfabrication technologies. Work on the implementation of silicon based Integrated TIR sensor is now in progress and will be published elsewhere.

3. Conclusions

A novel method to sensitively characterize thin films is proposed. The method is based on the analysis of differential phase in Si-based TIR geometry. We show that the proposed methodology is capable of providing the detection limit of at least $5*10^{-4}$ in terms of the refractive index change, which is comparable with sensitivities of currently available biosensing devices. The Si-based platforms also offers other advantages such an easy bioimmobilization step, as well as a good potential for sensor miniaturization using Si-based microfabrication technologies.

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