

Silicon based total internal reflection bio and chemical sensing with spectral phase detection

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Abstract: Si-based total internal reflection (TIR) bio/chemical sensor presents an attractive alternative to Surface Plasmon Resonance (SPR) technology due to a relatively simple optical arrangement and technological implementation, as well as a relatively easy bio/chemical immobilization on Si/SiO₂ surface with a number of novel attractive applications. This sensor is based on the control of phase difference between p- and s-polarized components of light reflected from Si/air or Si/water interface in TIR geometry and a high sensitivity of the sensor is granted by a high refractive index of Si (3.56 at 1200 nm). We study properties of TIR sensors in a configuration of spectral phase detection and identify conditions of maximal phase sensitive response. We also experimentally show that the detection limit of Si-based TIR sensor can be lowered down to a level of detection of commercially available SPR devices (10⁻⁶ Refractive Index Units, RIU) under the use of a proper low-noisy method of the phase control. The concept of Si-based TIR opens attractive prospects for the miniaturization of sensor devices, taking advantage of the advanced state of development of Si-based microfabrication technologies, while the proposed spectral phase detection scheme offers much easier packaging and calibration steps

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1. Introduction

Optical transduction-based bio- and chemical sensors become increasingly popular for last years for real time label-free characterization of bio/chemical interactions [1,2]. The optical transduction mechanism implies the control of a biological or chemical binding/recognition event through refractive index monitoring in thin films. Biosensors take advantage of the fact that most biological species have higher refractive index than water environment (1.44-1.5 compared to 1.33). A reactant is normally immobilized on a solid/liquid interface connected to the transducer. If a selective partner of the reactant (analyte) is present in the liquid environment, it binds to the reactant changing the refractive index of a thin near-surface layer (normally from several nm to several tens of nm). In contrast, chemical sensors use polymers, deposited on the working solid/gas interface, which change their index of refraction under the presence of a particular gas or volatile substance [3]. Methods for efficient refractive index monitoring in thin films normally employ evanescent waves, as in the cases of integrated planar [4] or fiber waveguides [5] or Surface Plasmon Resonance (SPR) biosensors [2,6–8]. Employing resonant conditions of plasmon excitation over a thin gold film to record refractive index changes near gold, SPR sensors provide the lowest detection limit reaching $(1-10) \cdot 10^{-6}$ Refractive Index Units (RIU) in commercially available devices [6–8]. The detection limit can be further improved down to 10^{-8} RIU using phase-sensitive schemes SPR interferometry [9–11] or SPR polarimetry [12,13].

We recently showed that the use of phase as a sensing parameter can lead to a good sensitivity to refractive index changes even in the absence of SPR-supporting gold layer, when light is reflected from an edge of a Si prism in TIR geometry [14]. The use of Si prism is a key element in such sensor, since this is a relatively high refractive index (RI) of Si (more than 3.5 in near-infrared) that conditions a drastic sharpening of phase characteristics and an amplification of phase response to RI changes compared to glasses (BK7, SF11 etc.) [14]. Using a relatively noisy ellipsometer-based arrangement, we were also able to achieve the detection limit of 10^{-4} RIU, which is not too far from the sensitivities of commercially available SPR devices. Advantages of the proposed Si-based TIR sensor include a relatively simple sensor block arrangement, the ease of immobilization of biospecies on Si/SiO₂ surface with a number of new applications not possible with gold, and excellent miniaturization prospects through the application of Si-based microfabrication methods. However, with the chosen angular phase detection scheme it is not easy to run the calibration procedure in miniaturized designs due to an inherent difficulty of adjusting the angle of incidence.

In this paper, we report the implementation of Si-based TIR sensor in configuration of spectral phase detection and examine characteristics of such sensor. We also demonstrate the possibility of a considerable (2 orders of magnitude) improvement of the detection limit of Si-based TIR sensor by applying low-noise polarimetry-based phase detection schemes. The spectral phase detection scheme is expected to simplify strategies for miniaturization due to the possibility of fine spectral tuning of phase characteristics around the TIR feature.

2. Calculation results

A schematic of the proposed sensor is depicted in Fig. 1(a). Near-infrared light is directed through a triangular silicon prism and reflected from its polished surface, which is in contact with a gaseous or aqueous medium in a flow cell. The reflected intensity is examined as a function of the pumping wavelength. Total internal reflection (TIR) is the reflection of the total amount of incident light at the boundary between two media when the incident angle θ_i is larger than the critical one $\theta_c = \arcsin(n_m/n_0)$, where n_0 and n_m are refractive indices of the incident and ambient media, respectively. In this case, phases of p- and s-polarization, δ_p and δ_s , at $n_m/n_0 > 1$ can be expressed as the following [15–17]:

$$\delta_p = 2 \arctan \frac{(n_m/n_0) \left[(n_m/n_0)^2 \sin^2 \theta_i - 1 \right]^{1/2}}{\cos \theta_i}, \quad \delta_s = 2 \arctan \frac{\left[(n_m/n_0)^2 \sin^2 \theta_i - 1 \right]^{1/2}}{(n_m/n_0) \cos \theta_i},$$

Therefore, phase difference Δ is given by: $\Delta = \delta_p - \delta_s = 2 \arctan \frac{\left[(n_m/n_0)^2 \sin^2 \theta_i - 1 \right]^{1/2}}{(n_m/n_0) \sin \theta_i \cdot \tan \theta_i}$.

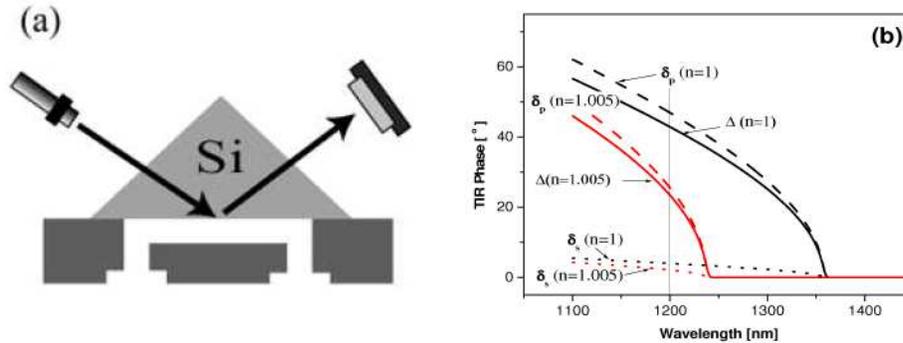


Fig. 1. (a) Schematic of Si-based TIR sensor; (b) Calculated spectral dependences for phases δ_p , δ_s , and phase difference Δ in the sensor with air as a sample medium. The dependences are given for the incident angle of 16.6°

Figure 1(b) shows typical spectral phase dependences of s- (δ_s) and p-polarized (δ_p) components, as well as the phase difference Δ , when Si-based sensor interface is in contact with a gaseous medium. To assess the response of all characteristics in gas sensing, we give the dependences for two different refractive indices: $n = 1$ (black curves) and $n = 1.005$ (red curves). One can see that phase of the p-polarized component δ_p experiences a substantial decrease as the pumping wavelength increases. For conditions given in Fig. 1(b), the total range of phase variation (60° Deg.) is limited by the cut-off silicon transparency wavelength (1100 nm), but its value can reach 120-150° Deg. through a proper selection of the light incident angle. As we showed in [14], such a significant phase dynamic range is granted by a relatively high refractive index of Si compared to glasses. In contrast, phase of s-polarized component δ_s only slightly depends on the wavelength. It is clear that in practice the phase difference Δ looks more promising as the sensing parameter, since it enables to take advantage of common-path phase-shift interferometry and polarimetry in order to easily exclude noises caused by temperature drifts, mechanical vibrations, light-source fluctuations etc. As shown in Fig. 1(b), the phase difference Δ follows the general downward trend of δ_p as the wavelength increases, whereas the dynamic range of phase variations is comparable. One can note that the change of the refractive index by 0.005 RIU leads to a drastic change of both δ_p and Δ , whereas δ_s looks almost unaffected. In particular, at 1200 nm the phases δ_p and Δ change by substantial 25.5° and 19.23° Deg., which the change of δ_s did not exceed 5° Deg.

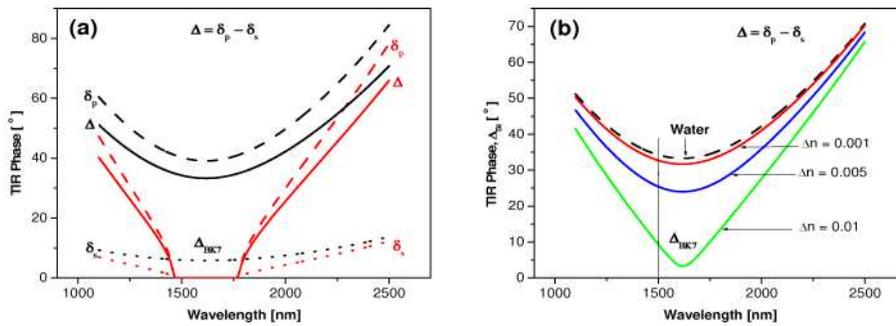


Fig. 2. (a) Calculated spectral dependences for phases δ_p , δ_s and phase difference Δ in TIR Si-based sensor with water as a sample medium. The dependences are given for the incident angles of 22.4° (black curves) and 22.2° (red curves); (b) Calculated response of the phase difference Δ to variations of the refractive index

Quite different spectral phase characteristics are observed when the Si-based sensor interface is in contact with an aqueous medium. Figure 2(a) presents phases δ_s , δ_p and Δ as a function of the pumping wavelength. One can see that for such medium spectral phase dependences critically depend on light incidence. For the angle of incidence of 22.4° Deg., all phase characteristics (δ_s , δ_p and Δ) demonstrate a minimum at 1600 nm. However, at 22.2° Deg. the minimum is transformed in a zero phase plane between 1450 and 1800 nm, which is explained by spectral dispersions of silicon and water refractive indexes in the near-infrared region. As we mentioned in [14], angular phase characteristics also strongly depend on the pumping wavelength. These data show the symmetry of the system when different parameters are employed for interrogation. Figure 2(b) demonstrates how a change of refractive index affects spectral phase dependences. All data are given for the incident angle of 22.4° Deg. As shown in this figure, the maximal phase response takes place near a relative minimum of the spectral phase dependence (1600 nm). For example, a change of the refractive index by 0.01 RIU leads to the phase shifts of 13° and 25° Deg. for 1200 nm and 1500 nm, respectively. It should be noted that in practice the angle of incidence should be selected to maximise the dynamic range of phase measurements and minimize zero plane effects. Thus, the incident angle of 22.4° Deg. (Fig. 2(b)) looks more adequate for sensing tasks.

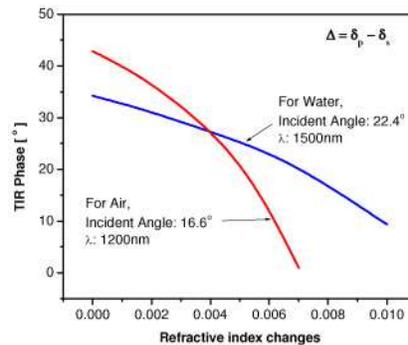


Fig. 3. Spectral TIR phase difference as a function of the medium refractive index changes in air and in water

Figure 3 presents phase of light reflected under TIR as a function of the RI the adjacent medium (relatively to refractive indices of air and water) for optimum wavelengths. Here, for air sample medium we selected 1200 nm, which is short enough to provide a highly sensitive response according to dependences of Fig. 1(b) and still larger than the cut-off wavelength (1050-1100 nm) corresponding to silicon absorption. For aqueous tested medium, we selected 1500 nm, which is in the range of the most sensitive response (Fig. 2(b)) and matches operation wavelengths of inexpensive laser diodes. One can see that a refractive index change

of 0.007 RIU causes a phase change of 42° Deg. and 22° Deg. for air and water environments, respectively. Although the response of the sensor in gas sensing is almost 2 times stronger, the sensitivity in both situations looks very promising taking into account that interferometry and polarimetry methods are capable of measuring phase variations of lower than 10⁻³ Deg [17].

3. Measurements results

In our last paper [14], we used an ellipsometer (Woollam VASE, J. A. Woollam, Lincoln, NE) to examine angular phase characteristics of Si-based TIR sensor and assess its sensitivity. Being one of best instruments for characterizing spectral and angular characteristics of various structures and devices, the ellipsometer is however not well suited for sensitivity tests because of a relatively high level of phase and amplitude noises. As a result, the measured detection limit level was not low enough (10⁻⁴ RIU), being 2 orders of magnitude higher when compared to amplitude-sensitive SPR devices. In this paper, we employ another polarimetry-based phase-sensitive scheme, which provides much lower level of phase noises and, as a consequence, is capable of measuring much smaller variations of refractive index. The scheme is a modification of a recently introduced photoelastic modulator (PEM) - based methodology with temporal phase modulation [18]. A schematic diagram of the proposed method is shown in Fig. 4(a). A laser diode operating at a wavelength of 1310 nm is used as the light source. Light is passed through a polarizer to obtain a 45° Deg. linearly polarized beam. After passing a combination of half and a quarter wave plates, which serve to optimize the initial phase retardation, light is directed to the PEM, which is used to sinusoidally modulate p-component at a fixed frequency of $\phi = 50$ kHz. The beam is then directed through a custom-made prism to be reflected under TIR from one of its facets. The angle of light incidence (16.7° Deg.) is selected to provide the sharpest spectral phase feature for the pumping wavelength. A polarizer (analyzer) is placed just after the sensing block and oriented 45° Deg. in front of the detector. Information on the phase of light reflected under SPR is obtained by measuring F1 and F2, the 1st and 2nd harmonics of the modulated signal, with the help of a lock in amplifier. Thus, for first two harmonics, we have:

$$F1 = A \cdot J_1(M) \cdot \cos(\phi) \quad F2 = A \cdot J_2(M) \cdot \sin(\phi)$$

where J_n are Bessel functions and M is the modulation amplitude. Phase information can now be extracted if we divide Equations for the two harmonics. In this case, amplitude noises can be subtracted by setting the modulation amplitude of PEM to zero point of the zero order Bessel function [19]. Thus, for the phase signal we have:

$$\tan(\phi) = F2 \cdot J_1(M) / F1 \cdot J_2(M)$$

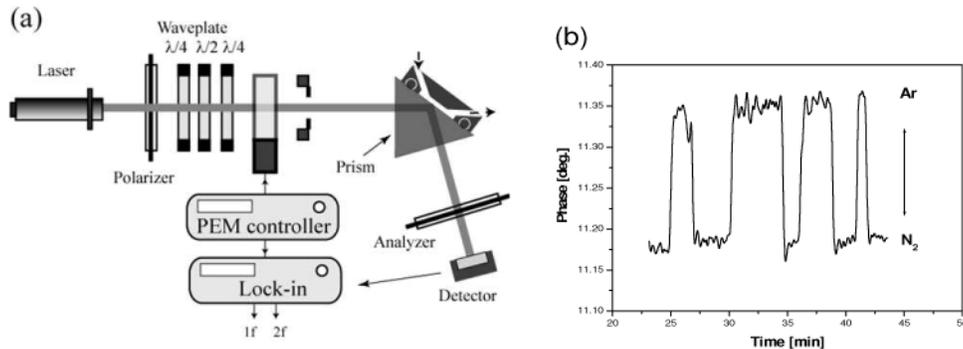


Fig. 4. (a) Schematics of phase measurements in Si-based TIR geometry using PEM; (b) Response of the system under the replacement of pure Ar by pure N2

To assess the detection limit of Si-based TIR sensor, we used a well-established gas methodology to simulate tiny refractive index (RI) variations Δn [9,20]. This method consists in measuring the system response while different inert gases with known refractive indices are brought into a contact with the gold film. In our experiments Ar and N₂ were used, for which the refractive indices differ by $\Delta n \cong 1.5 \cdot 10^{-5}$ RIU under the normal conditions [21]. The gases were flown through a long spiral copper tube to equalize their temperatures with the room one and then brought into contact with the sensor interface of the Si prism. Figure 4(b) demonstrates the response of the system when 100% of Ar was replaced by a 100% of N₂. One can see that such a tiny RI change leads to a significant phase shift of almost 0.18° Deg. Taking into account that the noise level did not exceed 0.1° Deg. (Fig. 4(b)), we can conclude that the experimental detection limit of the system is better than 10⁻⁶ RIU. In fact, this means that the sensitivity of TIR-based sensor with spectral phase interrogation is comparable with the one of commercial SPR sensors based on amplitude interrogation. Further improvements of the detection limit can be achieved through the employment of IR stabilized laser sources and active thermostabilization of the sensing block. Notice that for simplicity we placed Si prism in direct contact with tested medium. In real bio- and chemical sensing it is preferable to use cheap slides based on double side polished silicon wafer. When these slides are placed in close contact with bulk silicon prism, the system works as if the slide and the prism represent the same block without any parasitic reflections [22,23]. It should be also noted that a thin layer of native oxide (~1 nm) can appear on silicon surface after its exposition to air or water environment. However, since the thickness of this layer is extremely small compared to the penetration depth of evanescent field in Si-based TIR (1 μm), oxidation phenomena can be neglected in our conditions.

It is important that the proposed TIR sensor is implemented with a Si prism. This makes possible the application of powerful Si-based technologies for sensor miniaturization similar to the case of Si-based SPR [22,23]. In particular, one of miniaturized implementations implies the formation, through plasma etching or Focused Ion Beam, of a microprism connected to a microfluidic channel. It is clear that in miniaturized designs the wavelength is a preferable interrogation parameter compared to the angle of incidence, since it makes possible an easier device packaging and much simpler calibration step.

4. Conclusion

We considered the implementation of Si-based TIR sensor in configuration of spectral phase detection and identify conditions of maximal response in bio and chemical sensing. Employing a sensitive polarimetry-based phase detection setup, we showed the possibility of obtaining the measurement detection limit of the order of 10⁻⁶ RIU, which is comparable with commercial SPR devices. The proposed sensor looks very promising for miniaturization by using Si-based microfabrication methodologies.

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