PHASE INTERROGATION OF AN INTEGRATED OPTICAL SPR SENSOR

Anna K. Sheridan¹, Richard D. Harris¹, Philip N. Bartlett² & James S. Wilkinson¹

1) Optoelectronics Research Centre, University of Southampton, Highfield, Southampton, Hampshire, SO17 1BJ, UK.
2) Department of Chemistry, University of Southampton, Highfield, Southampton, Hampshire, SO17 1BJ, UK.

Abstract

A theoretical analysis of the phase behaviour of an integrated optical surface plasmon resonance (SPR) sensor is presented, in which the complex transmittance of a dielectric waveguide coated with a thin film of gold is determined for varying superstrate index. Exploitation of the phase sensitivity is studied theoretically by incorporating gold-coated waveguides on both branches of a Mach-Zehnder interferometer, one or both of which may be exposed to superstrate index variations or thin adlayers. It is proposed that this device can be used to suppress unwanted refractive index changes common to both branches, for example due to temperature variations, and may show enhanced sensitivity.

Keywords

Surface plasmon resonance, Mach Zehnder Interferometer, Integrated Optics, Sensor
1. Introduction

The use of surface plasmon resonance (SPR) for biological and chemical sensing is well established [1]. The high sensitivity of this technique to surface phenomena makes it ideal for use in bio- and chemical sensors where a very small change in the refractive index must be detected. Thin gold films are most often employed as they are chemically inert and provide an excellent surface for attachment of sensing films. The fabrication of the first integrated optical sensor based on the principle of surface plasmon resonance in the late 1980's [2] has led to extensive work in this area with the demonstration of several integrated optical SPR sensors [3-6]. In comparison with other integrated optical sensors based upon detecting changes in the refractive index of surface films, integrated SPR sensors offer the advantage of providing a metallic surface for combined electrochemical and optical measurements [7,8].

In the conventional (Kretschmann) prism configuration, the refractive index of the analyte is determined by measuring the angle of maximum attenuation of the incident beam or, at a fixed angle, by the reflectivity. Such a resonant absorption must be accompanied by a strong phase excursion, and it has been confirmed by ellipsometric measurements [9,10] that the resonant absorption is accompanied by a sharp jump in the phase of the reflected beam as the incident angle passes through the resonance. Nikitin et al. have since realised an SPR sensor based on the Kretschmann configuration, which uses interferometry to detect this phase shift [11,12]. It was shown both theoretically and experimentally that an increase in the sensitivity of the device may be achieved, compared with conventional intensity-based measurements.

In this paper, the possibility of using this phase behaviour to increase the sensitivity of an integrated optical surface plasmon resonance sensor is explored. The Mach Zehnder interferometer (MZI) is used as a model device, as it is one of the simplest integrated optical devices available to study relative phase variations in waveguides. An MZI incorporating identical gold-coated waveguides on both branches, and a phase bias in one branch, is modelled to yield the transmittance as a function of superstrate index. The effect of a small variation of the superstrate index on one branch is determined, to yield the sensitivity to specific changes. The capacity of this device to suppress unwanted refractive index changes which occur on both branches of the device - the common mode rejection ratio (CMRR) - is also determined. The results are compared with those of a simple integrated optical SPR sensor employing absorption in a single channel waveguide coated with gold. The sensitivity of such a device to the addition of a thin dielectric film representative of a protein layer is evaluated. The modelling is performed for a simple device which is not optimised for operating in a medium of a particular refractive index. However, the principles are general and the design could be optimised for analyte refractive index in a particular application by adjusting the waveguide index or incorporating a low-index film between the waveguide and the gold film [4].

2 Complex Transmittance of a Gold-Coated Waveguide

Figure 1 shows the structure of the gold-coated waveguide section used in all devices modelled in this paper. This is first studied on its own, to determine the phase and
amplitude behaviour and to provide results for the conventional waveguide SPR sensor as a benchmark, and the results are then incorporated in the model of a Mach-Zehnder interferometer in Section 3. The waveguide structure corresponds approximately to a single mode waveguide produced by potassium ion-exchange in Pyrex glass, with a gold film coating a short length. A wavelength of 633nm and a substrate with a refractive index of 1.471 were chosen so that the device could be optimised for use in aqueous media [13]. For optimisation, the thickness of the gold film was varied between 50nm and 70nm, while the length of the gold-coated section was varied between 0.5mm and 2.0mm. A lower limit of 50 nm for the gold film thickness was chosen as films thinner than this would not be robust enough for repeated experimental use.

To perform the calculations, a waveguide model described in detail in earlier publications [4,13] was extended to determine the phase, in addition to the intensity, of the light emerging from the gold-coated section, relative to the phase and intensity of the light entering the gold-coated region. If the field in the input waveguide is given as $E = E_1 e^{-i\theta_1}$, where $A_1$ is the magnitude of the amplitude transmission coefficient and $\theta_1$ is the phase, with the power transmittance being $A_1^2$. The model approximates the structure as a planar slab waveguide with a homogeneous refractive index, while the waveguides used in practice are often channel waveguides with gradient index profiles. However the approach used is considerably simpler than other models and provides useful scientific insights into the operation of the sensor. The model divides the waveguide into three parts - the input waveguide, the multi-layer gold-coated region, and the output waveguide. In the multi-layer region a transfer matrix approach is used to generate the system eigenvalue equation and Müllers method [14] is used to determine the complex modal effective indices. The number of modes is counted in the complex plane, using the argument principle method [15], to ensure that all relevant guided modes are taken into account in subsequent calculations. Overlap integrals are then used to calculate the excitation of these normal modes in the metal-clad region from the input waveguide section. These modes are then propagated through the metal coated region and finally recombined (with their amplitudes and phases) in the monomode output waveguide.

The transmittance of the gold-coated waveguide and the phase of the light emerging from the gold-coated region are plotted against superstrate index in Figure 2. Figures 2a, b & c show how the transmittance and phase varies for waveguides coated with gold film thicknesses of 50, 60 & 70nm, respectively. In each case the results are shown for lengths of gold coating of 0.5, 1.2 and 2.0mm. The phase relative to that at the input of the gold-coated region is not particularly significant here, so that the phase has been fixed at 0 at the centre of the resonance. The gradient of phase with respect to superstrate index (between the maximum and minimum points), $d\theta/dn$, is shown in Figure 3 for all the cases modelled. The phase shows the behaviour expected of a resonant system, with the magnitude of the phase excursion depending upon the depth and sharpness of the resonance. Increasing the gold film length leads to an increased phase excursion as the phase change experienced on propagation through a particular waveguide is proportional to its length. Increasing the gold film thickness results in a complex change in phase behaviour due to the combined effects of the increase in volume of absorbing material, the redistribution of the modal fields and hence the modal absorptions, and the variation in the coupling to each mode from the
input waveguide. It can be seen that the thinnest gold film leads to the deepest resonance, the largest phase excursion, and the highest rate of change of phase with change in index. Further, as expected, the phase change becomes greater as the gold film becomes longer. This is because the phase of each of the modes in the gold-coated region will be proportional to the length of the gold film. However, there is a limit to the useful length of the gold-coated region as the attenuation also increases with length and, to allow accurate measurement of the phase, measurable power must emerge from the gold-coated region.

3 MZI with Gold-Coated Branches

3.1 Device configuration

The phase of the light emerging from a gold-coated section of a waveguide has been shown above to vary strongly in the region of the surface plasmon resonance. In a practical system this phase variation may be measured by placing the gold-coated section in an interferometer, resulting in an intensity variation. In such a configuration the interferometric component of the intensity change on recombination in an MZI, for example, may augment or dominate the absorptive component of the intensity change in the gold film, and provide higher sensitivity than the conventional single-waveguide configuration.

To study the potential for sensitivity enhancement and common-mode rejection, two devices are modelled and compared. In each case the response was quantified by determining the transmittance with and without a thin dielectric adlayer of refractive index 1.4 and a thickness of 1 pm. These values were chosen to represent the adsorption of, for example, a thin protein film. In practice a protein monolayer has a thickness much greater than 1 pm, however as the detection limit of such a system may be defined in terms of the minimum detectable change in thickness, it is convenient to study the signal due to the addition of a 1 pm thick film.

The devices modelled were as follows:

i) An optical waveguide coated with a thin film of gold over a short section, as in Section 2, to establish the baseline performance of a conventional integrated optical surface plasmon resonance sensor.

ii) A Mach-Zehnder Interferometer incorporating in each branch an identical gold-coated waveguide section to that above. However the thin adlayer is only added on one branch and the other branch incorporates an additional fixed phase shift.

In the latter, the additional phase shift was added to one of the branches of the MZI to allow biasing of the device to a sensitive operating point. Incorporating a gold layer in only one branch of the interferometer would lead to poor contrast and low visibility of the interference, as the gold-coated region is strongly absorbing. Placing an identical gold layer in both branches but rendering only one branch sensitive to the measurement ensures that the field in each branch is of similar magnitude and that good interference contrast is achieved.

The schematic diagram in Figure 4 shows both the single waveguide device (4a) and the Mach Zehnder device (4b) studied in this work. The blocks enclosed with broken lines represent waveguide sections as modelled in Section 2 and shown in Figure 1.
The block labelled "Φ" represents the phase bias of the MZI, an additional design parameter in this configuration. The ratio of output power to input power, or transmittance, of both devices is determined as a function of superstrate index, with and without the adlayer, using the model described above. Conventional theory [16] gives the transmittance of the Mach-Zehnder interferometer as:

\[ T = \frac{1}{4} | (A_1 e^{-j\theta_1} + A_2 e^{-j\theta_2}) |^2 \]

where \( A \) and \( \theta \) are the amplitudes and phases of the light combining in the output coupler. Here \( A \) and \( \theta \) are taken to be the amplitude and the phase of the light emerging from the gold-coated sections as determined by the model in Section 2, and the subscript refers to branch 1 or 2 of the MZI.

### 3.2 Sensitivity and Detection Limit

The sensitivity, \( S \), to the addition of a thin adlayer may be defined as the difference in transmittance with and without the adlayer, \( \Delta T \), divided by the adlayer thickness, so that \( S = \Delta T / \Delta x \). This may be conveniently expressed, for comparison with other work, in terms of change in transmittance per picometre change in adlayer thickness. The use of this measure of sensitivity to optimise a particular device depends upon the limiting noise source in the complete sensing system. For practical transmitted power levels of order 1\( \mu \)W, the fundamental limiting noise source is normally thermal noise in the receiver, assuming that input light fluctuations can be referenced out by suitable device design. For receiver bandwidths of order 1Hz, typically employed for biosensing systems, the noise equivalent power (NEP) due to thermal noise is usually below 1pW. However, it is found in practice that the minimum detectable change in signal is limited by other instabilities to about 0.1%, some orders of magnitude worse than the thermal noise limit [13]. Under these circumstances, the limit of detection may be optimised by maximising the signal change for a given stimulus as a proportion of the transmitted power, using a normalised sensitivity, \( S' = S / T \), where \( T \) is the average of the transmittance with and without the adlayer. In the following, the normalised sensitivity and the change in transmittance for the single waveguide device and for the Mach-Zehnder device will be referred to as \( S'_{SP} \) and \( \Delta T_{SP} \) and \( S'_{MZ} \) and \( \Delta T_{MZ} \), respectively. Figure 5 shows the power transmission of the single waveguide device and the Mach-Zehnder device with and without the 1pm adlayer and the corresponding sensitivities \( S'_{SP} \) and \( S'_{MZ} \) for devices with gold length of 1.2 mm and thickness of 50 nm, as an example, with the phase bias \( \Phi \) set to zero. It can be seen that the behaviour of the conventional device and the Mach-Zehnder is rather similar, except that the sensitivity of the latter is about half that of the former. This is because, with \( \Phi = 0 \), the MZI is operating at a point of minimum phase sensitivity, and the absorptive component of the change in transmittance is reduced as only that power travelling through one arm of the MZI is affected by the adlayer. Figure 6 shows the sensitivity of the Mach-Zehnder device in comparison with that of the single waveguide device for the same gold film length and thickness but with a phase bias \( \Phi \) of 90°, 150° and 175°.

Figure 6 shows that the peak of the sensitivity of the MZI curve corresponds to the minimum of the SPR transmission curve. This is because the change in the phase with respect to refractive index is greatest close to the surface plasmon resonance as shown in Figure 2. It can be seen that the power transmission of the MZI decreases as the
phase bias is increased as expected in an interferometer, where the transmission is maximum when both arms are in phase and minimum when the arms are 180° out of phase. The sensitivity, shown in the upper panel, also increases as the phase bias increases, such that at a bias of 90°, the maximum (at a superstrate index of 1.363) sensitivity is comparable with that of the conventional device, whereas at a bias of 175° the sensitivity of the Mach Zehnder device is about 15 times higher. The sensitivity continues to increase as the phase bias approaches 180° as this is the point of maximum phase sensitivity for the MZI. This is shown clearly in Figure 7, where the ratio of the sensitivity of the MZI to the conventional device is plotted for phase shifts between 0 and 360 degrees. Figure 7 shows that with a phase bias greater than 90° the sensitivity ratio is greater than 1, so that the Mach Zehnder configuration is more sensitive than the single waveguide device. As the phase shift increases towards 180° the sensitivity increases rapidly. This shows that it should be possible to make an extremely sensitive device by fabricating a MZI to take advantage of the phase dependence near the surface plasmon resonance. However, as the phase bias approaches 180°, the transmitted power falls towards zero, so that thermal noise will ultimately create a lower bound for the minimum detection limit which can be achieved. In principle, for typical values such as a detection bandwidth of 1Hz, for which a change in received optical power of 1pW is readily detectable, and a device input power of 1mW, a device transmittance of -90dB would yield a measurable signal. However, this would be difficult to achieve in practice, principally because of spurious light scattered in the substrate.

The conventional waveguide SPR sensor modelled in Figure 5 shows maximum sensitivity at a superstrate index of 1.359 which falls to half of its peak value at Δn = ± 0.012. The combined MZI/SPR sensor with, for example, a 175° phase bias modelled in Figure 6c shows a peak sensitivity at a superstrate index of 1.363, and its sensitivity is higher than the peak sensitivity of the conventional waveguide SPR sensor for Δn = ± 0.014. Thus the MZI/SPR sensor exhibits a higher sensitivity than the conventional waveguide SPR sensor over a broader range of indices, shifted to a slightly higher superstrate index (at which the phase is changing most rapidly). These designs have not been optimised for operation in a specific analyte (superstrate) medium, but the peak sensitivity may be shifted to a desired operating point (for instance water with an index of 1.33) by minor adjustments to the waveguide parameters [4]. It should be noted that the regions of enhanced sensitivity correspond to regions where the device transmittance is below -80dB, and this would represent a significant challenge for practical devices.

3.3 Common Mode Rejection Ratio

Non-specific effects, such as the attachment of substances other than the target material layer on to the gold window or changes in ambient temperature, degrade the achievable detection limit in single waveguide SPR sensors. Both affect the apparent refractive index of the superstrate medium and may be misinterpreted as a change in concentration of the target material. In a conventional Mach Zehnder interferometer, refractive index changes which occur on both branches of the device can be suppressed as both branches of the device undergo an equal phase change, leaving the MZI output unchanged.
In order to determine whether the combined MZI/SPR device proposed here shows suppression of these spurious changes, the common mode rejection ratio (CMRR) has been calculated. This is defined as the ratio of the change in output power due to a change on one branch of the MZI to the change in output power measured when the same change is present on both branches of the Mach-Zehnder, and can be written as:

\[ \text{CMRR} = \frac{T_{m0} - T_{m1}}{T_{m0} - T_{m2}} \]

Where \( T_{m0} \) is the transmittance of a Mach-Zehnder device shown in Figure 4 with no additional layer on either of the gold windows. For this work \( T_{m1} \) has been chosen to be the transmittance of the Mach-Zehnder device with an additional layer of refractive index 1.4 and thickness 1pm which models a biological adlayer, and \( T_{m2} \) has been chosen to be the transmittance of the Mach-Zehnder device with the same adlayer on both the gold windows. The former represents the change due to a specific film binding to one arm of the device, while the latter represents the same film binding to both arms, representing a non-specific effect. If the magnitude of the CMRR is greater than 1, then the unwanted signals from the non-specific refractive index changes have been partially suppressed when compared with the conventional waveguide SPR sensor.

Figure 8 shows the magnitude of the CMRR as a function of refractive index for phase biases of 90, 150 and 175° over the superstrate index range 1.32 - 1.38 where the region of improved sensitivity lies. These curves all show a peak close to a superstrate index of 1.363, which corresponds to the surface plasmon resonance. The height of this peak increases rapidly as the phase bias is increased towards 180°. The horizontal line in Figure 8 indicates that the magnitude of the CMRR = 1, and values above this line indicate that the MZI device shows suppression of effects common to both branches. Figure 8 shows that at 90°, only devices operating in a very narrow band of refractive indices (from 1.361 to 1.364) will partially suppress unwanted signals. For a phase bias of 150 degrees all refractive indices below 1.339 and between 1.357 and 1.366 also demonstrate a CMRR greater than 1. The CMRR increases with bias so that with a phase bias of 175°, devices operating at a wide range of superstrate indices of interest have a CMRR greater than 1.

4 Conclusions

We have presented a theoretical analysis of the phase behaviour of an integrated optical surface plasmon resonance (SPR) sensor, in which the complex transmittance of a dielectric waveguide coated with a thin film of gold is determined for varying superstrate index. Exploitation of the phase sensitivity is studied theoretically by incorporating gold-coated waveguides on both branches of a Mach-Zehnder interferometer, one or both of which may be exposed to superstrate index variations and thin adlayers. By applying a phase bias in one branch of the interferometer an increase in the sensitivity of the device is achieved compared to a conventional waveguide SPR sensor, although further work is required to generate practical designs for specific applications. It is further shown that this configuration can be used to partially suppress unwanted refractive index changes common to both branches, for example due to temperature variations in the analyte medium or to non-specific binding. This paper demonstrates the principles of a novel device, and specific
designs must be generated depending on the analyte medium to match sensitivity and common-mode rejection ratio to the specific application. Work is now in progress to demonstrate such a device experimentally.

Acknowledgments
The Optoelectronics Research Centre is an Interdisciplinary Research Centre partially supported by the UK Engineering and Physical Sciences Research Council.
Figure Captions

1) Structure of the gold-coated waveguide section.
2) Transmittance and phase for propagation through the gold-coated waveguide section against superstrate index for gold thicknesses of a) 50 nm, b) 60 nm & c) 70 nm. In each case curves corresponding to gold films of lengths 0.5, 1.2 and 2.0mm are shown.
3) Maximum phase gradient, d\(\theta\)/dn, against film thickness for gold films of lengths 0.5, 1.2 and 2.0mm.
4) Schematic diagrams of the structures modelled a) a single waveguide with a gold film on one branch, b) a Mach-Zehnder interferometer with gold film on both branches and an additional phase shift \(\Phi\) in one branch.
5) Upper panel: The sensitivity as a function of refractive index for a single waveguide (solid line) and a Mach-Zehnder Interferometer (dotted line). Lower panel: Theoretical transmission for each of the devices shown in Figure 2. Devices with adlayer (dotted line) and devices without adlayer (solid line).
6) Upper panel: The sensitivity as a function of refractive index for a single waveguide (dotted line) and a Mach-Zehnder Interferometer (solid line). Lower panel: Theoretical transmission for each of the devices shown in Figure 2, devices with adlayer (dotted line) and devices without adlayer (solid line) for an additional phase shift of 90, 150 and 175 degrees.
7) Ratio of the maximum sensitivity of the MZI device to that of the single waveguide against additional phase shift.
8) Common mode rejection ratio (CMRR) for the Mach-Zehnder device for an additional phase shift of 90, 150 and 175 degrees.
References:


Figure 1

```
1.2 mm

n = 1.3 - 1.42
Superstrate

n = 1.4
Protein layer

n = 1.97 + j3.446
Gold film

2 μm
n = 1.4784
Waveguide

n = 1.471
Substrate
```

11
Figure 2
Figure 3
Figure 5
Figure 6

a) $\Phi = 90^\circ$

b) $\Phi = 150^\circ$

c) $\Phi = 175^\circ$
Figure 7
figure 8

a) $\Phi = 90^\circ$

b) $\Phi = 150^\circ$

c) $\Phi = 175^\circ$