Bulk and surface sensitivities of surface plasmon waveguides

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New Journal of Physics 10 (2008) 105010 (37pp)
Received 1 May 2008
Published 28 October 2008
Online at http://www.njp.org/
doi:10.1088/1367-2630/10/10/105010

Abstract. The potential of surface plasmon waveguides for bulk and surface (bio)chemical sensing was assessed theoretically, anticipating their use in an integrated optics sensor such as a Mach–Zehnder interferometer (MZI). The performance of a generic MZI implemented with attenuating waveguides was assessed initially, revealing that attenuating waveguides constrain the sensing length to an optimal length equal to the propagation length of the mode used. The MZI sensitivities for bulk and surface sensing were found to be proportional to the ratio of the waveguide sensitivity to its normalized attenuation: $H = (\partial n_{\text{eff}}/\partial n_c)/k_{\text{eff}}$ for bulk sensing and $G = (\partial n_{\text{eff}}/\partial a)/k_{\text{eff}}$ for surface sensing. Maximizing $H$ or $G$ maximizes the corresponding MZI sensitivity and minimizes its detection limit, leading to preferred waveguide designs and operating wavelengths. The propagation constant, the sensitivities, and the $H$ and $G$ parameters were then determined for the surface plasmon in the single interface, the $s_b$ mode in the metal–insulator–metal (MIM) waveguide and the $s_b$ mode in three variants of the insulator–metal–insulator (IMI) waveguide, as a function of dimensions, for wavelengths spanning $600 \leq \lambda_0 \leq 1600$ nm, assuming Au and H$_2$O as the materials and adlayers representative of biochemical matter. The principal findings are: (i) the surface sensitivity in the thin MIM can be $100 \times$ larger than in the single interface, whereas that in the thin IMI is up to $5 \times$ smaller; (ii) the bulk sensitivity in the thin MIM can be $3 \times$ larger than in the single interface, whereas that in the IMI is slightly smaller; (iii) $G$ in the thin MIM can be $3 \times$ larger than in the single interface, whereas $G$ in the IMI is about $10 \times$ larger; and (iv) $H$ in the thin MIM can be
10× smaller than in the single interface, whereas $H$ in the thin IMI is about 10× larger. The IMI and the MIM both offer an improvement in sensitivity and detection limit for surface sensing over the single interface in an integrated MZI (or Kretschmann–Raether) configuration, despite the fact that they are at opposite ends of the confinement–attenuation trade-off. Preferred wavelengths for surface sensing were found to be near the short wavelength edge of the Drude region, where detection limits of about 0.1 pg mm$^{-2}$ are predicted. With regard to bulk sensing, only the IMI offers an improvement over the single interface. The results are collected in a form that should be useful for investigating other sensor architectures implemented with these waveguides or variants thereof.

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1. **Introduction**

Surface plasmon polaritons (SPPs) in their simplest form are transverse-magnetic (TM) polarized optical surface waves propagating typically along the planar interface between optically semi-infinite metal and dielectric regions [1, 2]. Such ‘single-interface’ SPPs are known to have large bulk and surface sensitivities, but also large attenuations [1]–[3].

New Journal of Physics 10 (2008) 105010 (http://www.njp.org/)
The large surface sensitivity of SPPs has been heavily exploited in gas and (bio)chemical sensors ever since the initial demonstrations about two decades ago [3]. The conventional and mainstream approach to SPP sensing [3, 4] rests on the ever popular Kretschmann–Raether attenuated total reflection (ATR) prism arrangement [1, 5], where the metal film is in intimate contact with the prism, conveniently deposited directly on the latter or on an intervening glass slide of the same material as the prism. This arrangement is interrogated by launching a TM-polarized optical beam incident beyond the critical angle and monitoring the intensity of the reflected beam. As the angle of incidence or the wavelength of the incident beam is scanned, the intensity of the reflected beam goes through a minimum, indicating conditions under which the input beam couples evanescently into the SPP at the metal/fluid (gaseous or liquid) interface. The conditions for efficient evanescent coupling to the SPP depend strongly on the characteristics (thickness and index) of an adlayer located at the metal/fluid interface and on the index of the fluid itself, motivating the use of this arrangement as a chemical to optical transducer in (bio)chemical sensors. The coupling of an incident beam with the SPP in this manner is also referred to as surface plasmon resonance (SPR). Reviews describing the performance of various SPR sensors and interrogation schemes have recently been published [6]–[8].

Meanwhile, ongoing research in plasmonics has uncovered interesting and potentially useful attributes for many other metallic structures as reviewed in e.g. [9]–[12], such as apertures (e.g. [13]–[16]), particles (e.g. [17]–[20]), corrugated gratings (e.g. [21]–[23]), waveguides (e.g. [24]–[46]—discussed further in section 2.1), and integrated optic structures (e.g. [47]–[60]), such as bends [38], [48]–[50], [53]–[58], splitters/combiners [47]–[50], [53, 55, 58, 60], couplers [48]–[50], [55, 56], resonators [58, 59], Bragg gratings [51, 52, 54, 58, 59], and interferometers [47, 48, 55, 58, 60]. Given that the SPPs are tightly bound to the surface of the metal in the aforementioned structures, (bio)chemical sensors are often suggested as a good direction for potential applications. Indeed, this direction has led to success with conventional [3, 4] and other prism- or grating-based SPR sensors [6]–[8].

Work to date on (bio)chemical sensing with apertures [61]–[63] and metal particles [64]–[67] indicates that these approaches have potential for applications along this direction. But, perhaps surprisingly, the potential of approaches based on plasmonic waveguides [24]–[46] in integrated optic structures [47]–[60] has yet to be explored. Thus, the purpose of this paper is to assess theoretically the potential of such approaches for (bio)chemical sensing.

More particularly, we determine the bulk and surface sensitivities of modes in three popular SPP waveguides tailored to (bio)chemical sensing, and additionally, in two variants suitable for supporting long-range SPPs (LRSPPs). We adopt the single-output Mach–Zehnder interferometer (MZI) as the prototypical transducer structure, and estimate its sensitivity and detection limit when implemented with SPP waveguides. Using an SPP waveguide in a MZI follows similar implementations based on dielectric waveguides (e.g. [68]–[76]), or on metal-clad dielectric waveguides (e.g. [77]–[79]), and this is commented on in general terms at the end of the paper.

The paper is organized as follows: section 2 describes the SPP waveguides investigated and defines various quantities including the sensitivities. Section 3 discusses the sensitivity and detection limit of a generic single output MZI implemented with attenuating waveguides. Sections 4 to 7 collect results for the SPP waveguides of interest as a function of geometry and operating wavelength. Section 8 discusses the results and section 9 gives concluding remarks.
Figure 1. Sketches of 1D surface plasmon waveguides. The relative permittivity of the metal (yellow) and dielectric (light blue) regions is $\epsilon_{r,m}$ and $\epsilon_{r,c}$, respectively. The dielectric adlayers (red) have a thickness $a$ and a relative permittivity $\epsilon_{r,a}$. The real part of the main transverse electric field component (Re$\{E_y\}$) of the mode of interest is sketched in green on all structures. (a) Single-interface between a metal and a dielectric. (b) Metal-bounded dielectric film of thickness $t$. (c) and (d) dielectric bounded metal film of thickness $t$. (e) Metal film of thickness $t$, on a dielectric membrane of thickness $d$ and relative permittivity $\epsilon_{r,d}$, bounded by dielectric.

2. Waveguides investigated and definitions

2.1. Waveguide structures

The waveguides and operating modes of interest are sketched in figure 1. These are one-dimensional (1D) waveguides providing field confinement along the $y$-direction only with propagation occurring along the $z$-direction (out of the page). Figure 1(a) shows the conventional single-interface metal–dielectric waveguide (e.g. [1, 2]), figure 1(b) shows a metal-clad dielectric film of thickness $t$ henceforth referred to as the metal–insulator–metal (MIM) waveguide (e.g. [39, 40, 80, 81]), and figure 1(c) shows a dielectric-clad metal film of thickness $t$ henceforth referred to as the insulator–metal–insulator (IMI) waveguide (e.g. [40], [80]–[82]). The profile of the main transverse electric field component (Re$\{E_y\}$) of the mode of interest in each structure is also sketched: in the single interface, it is the usual purely bound (non-radiative) SPP, and in the MIM and IMI it is the purely bound symmetric coupled mode, denoted herein.
as $s_b$. Thin dielectric adlayers of thickness $a$ are also shown. The structures of figures 1(d) and (e) are variants of the IMI and will be discussed further below.

There are many reasons for considering these 1D waveguides operating in the mode highlighted: (i) the geometrical design space of 1D waveguides is small and so can easily be spanned (no thicknesses are needed to define the single-interface, and only one thickness is needed to define the MIM and IMI). (ii) The 1D structures require little numerical effort for an accurate analysis. (iii) The modes of interest can all be excited in an end-fire arrangement. (iv) The structures complement each other in that they capture different confinement–attenuation trade-offs [40, 81]. The single interface is, nominally, in the ‘middle’ of the trade-off, whereas the MIM and IMI are at opposite ends for small $t$. For small $t$, the $s_b$ mode of the MIM is strongly confined but also strongly attenuated (e.g. [39]) and thus often termed a short-range SPP (SRSPP), whereas the $s_b$ mode in the IMI is weakly confined but also weakly attenuated (e.g. [82]) and thus is often termed an LRSPP. Consequently, it is expected that the bulk and surface sensitivities of the modes in these waveguides will be at opposite ends, thus leading to very different sensor designs. (v) The 1D waveguides are limits of their 2D variants having a large width and operating in their main mode. For example, the metal stripe in an asymmetric dielectric environment (e.g. [25], [28]–[30], [41]) supports single-lobed modes having a single extremum along the width of the stripe (among others) localized along either of the metal–dielectric interfaces, and these modes have characteristics (effective index, attenuation, perpendicular confinement, ...) comparable to the corresponding single-interface SPP (figure 1(a)). The $ss_0^0$ mode in the metal stripe bounded by symmetric dielectrics (e.g. [24, 26, 32, 42]) is similar to the $s_b$ mode of the IMI (figure 1(c)). The main symmetric mode of gap and wedge waveguides (e.g. [31, 33], [35]–[38], [45]) bear a strong resemblance to the $s_b$ mode of the MIM (figure 2(b)). Therefore, it is expected that the trends uncovered for the 1D waveguides would carry over to their 2D variants. (vi) Although the bulk and surface sensitivities of the single-interface SPP are reasonably well understood (e.g. [8, 83]), the structure is included here in order to facilitate comparisons, explain trends, and assess sensor designs based on its 2D variants.

2.2. Modelling approach and material parameters

An $e^{j\omega t}$ time dependence is assumed throughout and modes propagate in the $+z$-direction according to $e^{-j\gamma z}$. The complex propagation constant $\gamma$ in m$^{-1}$ expands as $\gamma = \alpha + j\beta$, where $\alpha$ and $\beta$ are the attenuation and phase constants, respectively. The normalized propagation constant $\gamma_{\text{eff}}$ is given by $\gamma_{\text{eff}} = \gamma/\beta_0 = \alpha/\beta_0 + j\beta/\beta_0 = k_{\text{eff}} + jn_{\text{eff}}$ where $\beta_0 = 2\pi/\lambda_0 = \omega/c_0$ is the phase constant of plane waves in free space, $\lambda_0$ the wavelength in free space, and $c_0$ the speed of light in free space. The complex effective index of a mode $N_{\text{eff}}$ is then given by $N_{\text{eff}} = -j\gamma/\beta_0 = \beta/\beta_0 - j\alpha/\beta_0 = n_{\text{eff}} - jk_{\text{eff}}$. The mode power attenuation (MPA) in dB m$^{-1}$ is given by MPA = $a20\log_{10}e$, and the $1/e$ propagation length by $L_c = 1/(2\alpha)$. The relative permittivity is obtained from the optical parameters $n$, $k$ in the usual way $\varepsilon_r = (n - jk)^2$. The relative permittivity of the metal breaks down into real and imaginary parts as $\varepsilon_{r,m} = -\varepsilon_R - j\varepsilon_I$. All numerical modelling is conducted using the method of lines (MoL) [26, 84, 85].

The materials assumed for the structures of figure 1 are Au for the metal ($\varepsilon_{r,m}$) and H$_2$O for the background dielectric ($\varepsilon_{r,c}$). This choice is based on relevance to biosensors where an aqueous solution is typically used as a carrier fluid for the analyte. Au is a good plasmonic metal and not very reactive. Also, good thiol-based chemistries are readily available to functionalize Au surfaces.
The optical parameters adopted for the materials are plotted in figure 2 for $\lambda_0$ ranging from 600 to 1600 nm. The data points from the original sources are plotted as well as the spline interpolations used for the numerical modelling. The parameters for Au originate from [86] (and a single source therein [87]), those for H$_2$O from [88], and those for Si$_3$N$_4$ from [89] (Si$_3$N$_4$ is used for the structure of figure 1(e), as described further below). Figure 3 plots the relative permittivity of Au and H$_2$O (from the splines) over a slightly broader wavelength range from $\lambda_0 = 400$ to 2000 nm. At wavelengths shorter than about 650 nm the optical absorption in Au increases due to the onset of interband transitions so the range considered in the modelling is taken from $\lambda_0 = 600$ to 1600 nm, with the availability of good, inexpensive and compact optical sources justifying our upper limit of 1600 nm.

Each adlayer depicted in figure 1 is modelled as a homogeneous plane-parallel longitudinally invariant dielectric region. A relative permittivity representative of biological material is adopted, $\varepsilon_{\text{rel}} = 1.5^2$ (e.g. [90]), and assumed constant over the wavelength range considered.
2.3. Definition of waveguide sensitivities

The bulk sensitivity is defined as $\partial N_{\text{eff}}/\partial n_c$ and is determined without adlayers on the waveguides ($a = 0$). This sensitivity breaks down into real and imaginary parts yielding the effective index bulk sensitivity $\partial n_{\text{eff}}/\partial n_c$ and the normalized attenuation bulk sensitivity $\partial k_{\text{eff}}/\partial n_c$.

The surface sensitivity can be defined in a number of ways \[8, 83\], \[91\]–\[93\]. The definition adopted here is $\partial N_{\text{eff}}/\partial a$, where $a$ is the thickness of the adlayer. This sensitivity breaks down into real and imaginary parts yielding the effective index surface sensitivity $\partial n_{\text{eff}}/\partial a$ and the normalized attenuation surface sensitivity $\partial k_{\text{eff}}/\partial a$.

These sensitivities are computed directly by approximating the differentials via $O(h^2)$ central finite-difference formulae (e.g. \[64\]):

$$\frac{\partial N_{\text{eff}}}{\partial n_c} = \frac{N_{\text{eff}} (n_c + h_c - j k_c) - N_{\text{eff}} (n_c - h_c - j k_c)}{2 h_c}$$

and

$$\frac{\partial N_{\text{eff}}}{\partial a} = \frac{N_{\text{eff}} (a + h_a) - N_{\text{eff}} (a - h_a)}{2 h_a}.$$  

These approximations improve in accuracy as $h_c, h_a \rightarrow 0$ (to within numerical accuracy). In the computations, we used $h_c = 10^{-5} \ll n_c$ and $h_a = 0.1 \text{ nm} \ll a$ with $a = 3 \text{ nm}$ as a prototypical adlayer thickness, representing, say, a thin monolayer of receptor molecules. The surface sensitivity depends on $a$, but only weakly so for $a$ small compared with the mode width. (We neglect in equation (1) corresponding perturbations to $k_c$ necessarily arising through the Kramers–Kronig relations.)

Alternatively, following perturbation methods (e.g. \[95\]), it is possible to determine the change in the phase constant of a mode due to a longitudinally invariant perturbation in the permittivity distribution of the structure. The effective index sensitivity can then be determined from this change in phase constant. Neglecting losses, the unperturbed ($u$) and perturbed ($p$) fields are written as:

$$E_u(x, y, z) = E_u(x, y) e^{-j \beta_u z}, \quad H_u(x, y, z) = H_u(x, y) e^{-j \beta_u z}, \quad E_p(x, y, z) = E_p(x, y) e^{-j \beta_p z}, \quad H_p(x, y, z) = H_p(x, y) e^{-j \beta_p z}.$$  

The perturbed permittivity distribution is related to the unperturbed one by

$$\epsilon_p(x, y) = \epsilon_u(x, y) + \Delta \epsilon(x, y).$$

From \[95\], the change in the phase constant due to the perturbation is given by

$$\beta_p - \beta_u = \frac{\int \int_{A_{\infty}} \Delta \epsilon E_p \cdot E_u^* dA}{\int \int_{A_{\infty}} (E_u^* \times H_p + E_p \times H_u^*) \hat{z} dA},$$

where $\hat{z}$ is the unit vector in the $z$-direction and $A_{\infty}$ is the infinite transverse cross-sectional area. In the case of small perturbations $\Delta \epsilon$, the perturbed fields can be taken as the unperturbed ones. Under this assumption, and writing $\Delta \epsilon$ in terms of refractive index distributions:

$$\Delta \epsilon(x, y) = \epsilon_0 \left( n_p^2(x, y) - n_u^2(x, y) \right),$$

the following expression for the change in effective index is obtained:

$$n_{\text{eff},p} - n_{\text{eff},u} = \sqrt{\frac{\epsilon_0}{\mu_0}} \frac{1}{4 P_{\text{AVE}}} \int \int_{A_{\infty}} \left( n_p^2 - n_u^2 \right) |E_u|^2 dA,$$
where $P_{\text{AVE}}$ is the time-averaged real power carried by the mode:

$$P_{\text{AVE}} = \frac{1}{2} \int \int_{A_{\infty}} \text{Re} \left\{ E_u \times H_u^* \right\} \hat{z} \, dA. \quad (8)$$

For the case where the perturbation corresponds to the appearance of an adlayer of cross-sectional area $A_a$ and index $n_a$:

$$n_p^2(x, y) - n_c^2(x, y) = \begin{cases} n_a^2 - n_c^2 & \text{for } x, y \text{ in } A_a \\ 0 & \text{elsewhere} \end{cases}, \quad (9)$$

then equation (7) simplifies to:

$$\Delta n_{\text{eff}} = n_{\text{eff}, p} - n_{\text{eff}, u} = \sqrt{\frac{\varepsilon_0}{\mu_0}} \frac{n_a^2 - n_c^2}{4 P_{\text{AVE}}} \int \int_{A_a} |E_u|^2 \, dA. \quad (10)$$

From the above, it is observed that $\Delta n_{\text{eff}} > 0$ if $n_a^2 > n_c^2$, which is generally the case for biosensors. Clearly, maximizing $\Delta n_{\text{eff}}$ means maximizing the overlap of the mode fields with the adlayer area $A_a$. Fields outside of $A_a$ do not contribute to the integral, so, if possible, (i) the mode area should be similar to $A_a$, and (ii) the mode fields should peak within $A_a$. Point (ii) is met for all of the modes sketched in figure 1. If the adlayer is very thin, then based on point (i), the $s_b$ mode in a thin MIM (small $t$) should have the greatest surface sensitivity, followed by the SPP in the single interface, and then finally by the $s_b$ mode in the thin IMI (small $t$), since their mode widths range from smallest to largest, respectively [82]. (It is possible to add a thick molecular binding layer such as a hydrogel matrix [96] to the surface of the metal in order to increase its binding capacity per unit area and the overlap of the mode fields with the sensing region; this is particularly relevant for the single-interface SPP and the thin IMI.)

For the case where a perturbation $h_c$ with $h_c \ll n_c$ is applied to the index of the carrier fluid occupying the area $A_c$:

$$n_p^2(x, y) - n_c^2(x, y) = \begin{cases} (n_c + h_c)^2 - n_c^2 & \text{for } x, y \text{ in } A_c \\ 0 & \text{elsewhere} \end{cases}, \quad (11)$$

then equation (7) simplifies to

$$n_{\text{eff}, p} - n_{\text{eff}, u} = \sqrt{\frac{\varepsilon_0}{\mu_0}} \frac{n_c h_c}{2 P_{\text{AVE}}} \int \int_{A_c} |E_u|^2 \, dA. \quad (12)$$

Considerations similar to those discussed with regard to increasing the perturbation due to an adlayer apply to this case as well.

Equations (10) and (12) could be used to determine the surface and bulk sensitivities, respectively, instead of the finite-difference approximations given by equations (1) and (2); the latter were used here since they are easier to compute.

3. MZI

The prototypical transducer considered is the single-output equal-arm equal-split/combine MZI sketched in figure 4. It is assumed that the MZI is implemented end-to-end using a 2D variant of a 1D waveguide sketched in figure 1, and as mentioned earlier, that the waveguide essentially retains the performance characteristics of its 1D counterpart (propagation constant and sensitivities—this was verified for the LRSPP of the symmetric metal stripe [26]). The MZI sketch then represents the optical path of the mode as it propagates along the structure rather
than a specific geometrical feature. Fluidic channels are also sketched overlapping with each arm of the MZI, where one is identified as the sensing arm and the other as the reference arm. The length of the sensing region is \( L \), and \( L_0 \) (not shown) is the optical path length needed for the input and output access lines and the splitter and combiner. *We emphasize that the relationships derived in this section hold for a generic MZI implemented with attenuating waveguides, including dielectric waveguides that have losses.*

Figure 4(a) shows the MZI configured as a biosensor, where the sensing arm is coated with receptor molecules specific to the target analyte, and the reference arm is coated with a blocking chemical. The carrier fluid with analyte flows over both arms through the fluidic channels, and as analyte binds to the receptors (but not to the blocking chemical) the effective index of the sensing arm changes, which in turn changes the phase relationship between the arms causing the MZI output power to vary. Figure 4(b) shows the MZI used as a bulk sensor, where the fluid to be monitored flows over the sensing arm and a reference fluid flows over the reference arm. Differences in their bulk index cause changes in the MZI output power.

The output power of the MZI, implemented with a waveguide having an attenuation of \( \alpha \) (the attenuation cannot be neglected in SPP waveguides) is given by

\[
P_{\text{out}} = P_{\text{in}} e^{-2\alpha(L_0+L)} \frac{1}{2} (1 + \cos \phi_0),
\]

(13)
where $\phi_D$ is the difference between the insertion phase of the sensing (s) and reference (r) arms:

$$\phi_D = \frac{2\pi L}{\lambda_0} (n_{\text{eff},s} - n_{\text{eff},r}).$$

(14)

$P_{\text{out}}$ is the only measurable in this simple structure, so we define the MZI phase sensitivity as $\partial P_{\text{out}}/\partial \phi_D$, which works out to

$$\frac{\partial P_{\text{out}}}{\partial \phi_D} = -P_{\text{in}} \frac{1}{2} e^{-2\alpha(L_0+L)} \sin \phi_D.$$

(15)

As is evident from the above (and as is well known), the maximum sensitivity occurs for $\phi_D = p\pi/2$ with $p = \pm 1, \pm 3, \pm 5, \ldots$ and vanishes for $\phi_D = q\pi$ with $q = 0, \pm 1, \pm 2, \pm 3, \ldots$. This ‘sensitivity fading’ can be mitigated by adding phase modulation to one of the MZI arms (say thermo-optically by heating one of the arms) in order to maintain operation at $\phi_D \sim p\pi/2$.

From equation (15), it is immediately apparent that the waveguide attenuation introduces a ‘power penalty’ relative to a lossless waveguide, in that $P_{\text{in}}$ needs to be increased in order to recover the lossless ($\alpha = 0$) MZI phase sensitivity.

3.1. MZI surface sensitivity and detection limit

The MZI surface sensitivity is defined as $\partial P_{\text{out}}/\partial a = (\partial P_{\text{out}}/\partial \phi_D)(\partial \phi_D/\partial a)$. From equation (14), the phase surface sensitivity $\partial \phi_D/\partial a$ is

$$\frac{\partial \phi_D}{\partial a} = \frac{2\pi L}{\lambda_0} \frac{\partial n_{\text{eff},s}}{\partial a}.$$

(16)

Combining the above with equation (15) yields

$$\frac{\partial P_{\text{out}}}{\partial a} = -P_{\text{in}} \frac{1}{2} e^{-2\alpha(L_0+L)} \sin \phi_D \frac{2\pi L}{\lambda_0} \frac{\partial n_{\text{eff},s}}{\partial a}.$$

(17)

In addition to the points made above with regard to equation (15), the magnitude of the MZI surface sensitivity $|\partial P_{\text{out}}/\partial a|$ increases with increasing $P_{\text{in}}$ and $\partial n_{\text{eff},s}/\partial a$, and with decreasing $\lambda_0$ and $\alpha$. For a surface plasmon waveguide, the parameters $\partial n_{\text{eff},s}/\partial a$ and $\alpha$ are not independent and both depend on $\lambda_0$. Also, it is not clear from the above how $L$ should be selected: increasing $L$ causes $|\partial P_{\text{out}}/\partial a|$ to increase linearly but also to decrease exponentially. We rewrite the above as

$$\frac{\partial P_{\text{out}}}{\partial a} = -P_{\text{in}} \frac{1}{2} e^{-2\alpha L_0} \sin \phi_D \frac{U(L)}{\lambda_0} \frac{2\pi L}{\lambda_0} \frac{\partial n_{\text{eff},s}}{\partial a},$$

(18)

from which we observe that $L$ could be selected to maximize $U$. This is achieved by setting $\partial U/\partial L$ to zero and finding the roots:

$$\frac{\partial U}{\partial L} = e^{-2\alpha L}(1 - 2\alpha L) = 0,$$

(19)

which are $L \rightarrow \infty$ and $L = 1/(2\alpha)$. Using L’Hôpital’s rule, it is easy to verify that the limit $U(L \rightarrow \infty)$ is 0, so the MZI surface sensitivity vanishes for this length. Inspection of the sign of $\partial^2 U/\partial L^2$ at $L = 1/(2\alpha)$ reveals that this solution corresponds to a maximum, hence we refer
to this length as the optimal sensing length, and we note, interestingly, that it corresponds to the propagation length of the mode $L_e$. The maximum value of $U$ is thus

$$ U(L_e) = \frac{1}{2a\epsilon}. \tag{20} $$

Substituting the above into equation (18) yields

$$ \frac{\partial P_{\text{out}}}{\partial a}(L = L_e) = -P_{\text{in}} \frac{1}{\lambda_0} e^{-(1+2aL_0)} \sin(\phi_D) \frac{2\pi}{\lambda_0} \frac{1}{\alpha} \frac{\partial n_{\text{eff},s}}{\partial a}, \tag{21} $$

which we rewrite as

$$ \frac{\partial P_{\text{out}}}{\partial a}(L = L_e) = -P_{\text{in}} \frac{1}{\lambda_0} e^{-(1+2aL_0)} \sin(\phi_D) G \tag{22} $$

defining

$$ G \equiv \frac{2\pi}{\lambda_0} \frac{1}{\alpha} \frac{\partial n_{\text{eff},s}}{\partial a} = \frac{\beta_0}{\alpha} \frac{\partial n_{\text{eff},s}}{\partial a} = \frac{\partial n_{\text{eff},s}}{\partial a}, \tag{23} $$

$G$ lumps into a single parameter, $\lambda_0$ and the wavelength-dependent waveguide parameters $\partial n_{\text{eff},s}/\partial a$ and $\alpha$. From (22) it is clear that maximizing $|\partial P_{\text{out}}/\partial a|$ means maximizing $G$; therefore, the best waveguide design and operating wavelength for surface sensing are those that maximize $G$.

Noting that the transmittance of the sensing section is $T_L = e^{-2aL}$, it is observed from the above equations that maximizing $U$ then maximizing $G$ is equivalent to maximizing the product $T_L \cdot \partial \phi_D/\partial a$, which is the transmittance times the phase surface sensitivity (equation (16)) of the sensing region. This is intuitively satisfying since the MZI sensitivity was defined in terms of its (absolute) output power.

It is also interesting to note that de Bruijn et al [97] arrived at a ratio similar to equation (23) when considering the reflectance response of the fixed-angle fixed-wavelength Kretschmann–Raether SPR setup.

As the attenuation of the waveguide vanishes $\alpha \to 0$, the propagation length and optimal sensing length diverge $L_e \to \infty$, the maximum value of $U$ diverges $U(L_e) \to \infty$ (equation (20)), $G$ diverges (equation (23)), and consequently the MZI surface sensitivity diverges $|\partial P_{\text{out}}/\partial a| \to \infty$ (equation (22)). Indeed this final point is also directly observed from equation (17) by setting $\alpha = 0$ and letting $L \to \infty$; i.e. we recover the usual result for lossless waveguides that the MZI surface sensitivity $|\partial P_{\text{out}}/\partial a|$ increases linearly with the sensing length. We emphasize however that as soon as attenuating waveguides are used, the solution $L \to \infty$ leads to vanishing sensitivity ($|\partial P_{\text{out}}/\partial a| \to 0$, equation (18)) and the optimal sensing length becomes $L_e$.

If we assume, irrespective of the waveguide, that the splitter/combiner and any input/output access lines can be designed such that their total path length $L_0$ is also equal to $1/(2\alpha)$ then equation (22) simplifies to

$$ \frac{\partial P_{\text{out}}}{\partial a}(L = L_0 = L_e) = -P_{\text{in}} \frac{1}{\lambda_0} e^{-2} \sin(\phi_D) G. \tag{24} $$

The surface mass coverage $\Gamma (\text{g} \text{m}^{-2})$ is related to the adlayer parameters by [98]:

$$ \Gamma = \frac{a(n_a - n_c)}{\partial n/\partial c}, \tag{25} $$

where $\partial n/\partial c$ (m$^3$ g$^{-1}$) is the index variation with analyte concentration (typically, $\partial n/\partial c \sim 187$ mm$^3$ g$^{-1}$). Changes in surface mass coverage $\Delta \Gamma$ are related to changes in adlayer thickness
\[ \Delta a \text{ via the above, and changes in } P_{\text{out}} \text{ are related to } \Delta a \text{ via:} \]

\[ \Delta P_{\text{out}} = \frac{\partial P_{\text{out}}}{\partial a} \Delta a. \]  

(26)

Denoting the smallest measurable change in output power as \( \Delta P_{\text{out,min}} \), we obtain for the detection limit \( \Delta \Gamma_{\text{min}} \) of the MZI the following expression by combining equations (25) and (26):

\[ \Delta \Gamma_{\text{min}} = \frac{(n_a - n_c)}{\partial n/\partial c} \frac{1}{\partial P_{\text{out}}/\partial a} \Delta P_{\text{out,min}}. \]  

(27)

It is noted that the detection limit \( \Delta \Gamma_{\text{min}} \) decreases as the MZI surface sensitivity \( |\partial P_{\text{out}}/\partial a| \) increases.

### 3.2. MZI bulk sensitivity and detection limit

In the case of bulk sensing, the phase bulk sensitivity \( \partial \phi_D/\partial n_c \) is obtained from equation (14) as

\[ \frac{\partial \phi_D}{\partial n_c} = \frac{2\pi L}{\lambda_0} \frac{\partial n_{\text{eff,s}}}{\partial n_c}. \]  

(28)

Then, following a similar derivation, the MZI bulk sensitivity \( \partial P_{\text{out}}/\partial n_c \) is found to be given by equation (17) except that \( \partial n_{\text{eff,s}}/\partial a \) is replaced with \( \partial n_{\text{eff,s}}/\partial n_c \). For \( L = L_c \), the expression simplifies to

\[ \frac{\partial P_{\text{out}}}{\partial n_c} (L = L_c) = -P_m \frac{1}{4} e^{-(1+2\alpha L_0)} \sin(\phi_D) \frac{2\pi}{\lambda_0} \frac{1}{\alpha} \frac{\partial n_{\text{eff,s}}}{\partial n_c}. \]  

(29)

which we rewrite as

\[ \frac{\partial P_{\text{out}}}{\partial n_c} (L = L_c) = -P_m \frac{1}{4} e^{-(1+2\alpha L_0)} \sin(\phi_D) H \]  

defining

\[ H \equiv \frac{2\pi}{\lambda_0} \frac{1}{\alpha} \frac{\partial n_{\text{eff,s}}}{\partial n_c} = \frac{\beta_0}{\alpha} \frac{\partial n_{\text{eff,s}}}{\partial n_c} = \frac{\partial n_{\text{eff,s}}}{\partial n_c} \frac{1}{k_{\text{eff}}}. \]  

(31)

From (30) it is clear that maximizing \( |\partial P_{\text{out}}/\partial n_c| \) means maximizing \( H \); therefore, the best waveguide design and operating wavelength for bulk sensing are those that maximize \( H \). If \( L_0 \) is set to \( L_c \) then equation (30) simplifies to equation (24) except that \( G \) is replaced with \( H \).

The detection limit for bulk sensing is

\[ \Delta n_{c,\text{min}} = \frac{1}{\partial P_{\text{out}}/\partial n_c} \Delta P_{\text{out,min}}, \]  

(32)

from which we note that it decreases as the MZI bulk sensitivity \( |\partial P_{\text{out}}/\partial n_c| \) increases.

### 3.3. Relationship between MZI sensitivities and detection limits

For identical MZIs operating at the same \( P_m \) and \( \lambda_0 \), and near \( \sin(\phi_D) \sim 1 \) we find

\[ \frac{\partial P_{\text{out}}/\partial a}{\partial n_{\text{eff,s}}/\partial n_c} \]  

(33)

Assuming furthermore identical \( \Delta P_{\text{out,min}} \), the ratio of the detection limits simplifies to

\[ \frac{\Delta \Gamma_{\text{min}}}{\Delta n_{c,\text{min}}} = \frac{(n_a - n_c)}{\partial n/\partial c} \frac{\partial n_{\text{eff,s}}/\partial n_c}{\partial n_{\text{eff,s}}/\partial a}, \]  

(34)

which is useful for relating one detection limit to the other.

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3.4. Effect of the attenuation sensitivity

In the foregoing development, we have assumed that the normalized attenuation sensitivities, $\partial k_{\text{eff}}/\partial a$ and $\partial k_{\text{eff}}/\partial n_c$ (or the attenuation sensitivities $\partial \alpha/\partial a$ and $\partial \alpha/\partial n_c$), of the sensing waveguide are small enough not to cause an appreciable change in the transmittance of the sensing arm or in the performance of the MZI. We derive, in this subsection, bounds under which this assumption remains valid.

The output power of the single-output MZI, in the more general case where the reference and sensing waveguides have different attenuations $\alpha_r$ and $\alpha_s$ respectively, is given by

$$P_{\text{out}} = P_{\text{in}} e^{-2\alpha_r L_0} \left( e^{-2\alpha_r L} + e^{-2\alpha_s L} \right)^{\frac{1}{4}} (1 + V \cos \phi_D).$$

(35)

The MZI visibility $V$ is

$$V = \frac{2 e^{-\alpha_r L} e^{-\alpha_s L}}{e^{-2\alpha_r L} + e^{-2\alpha_s L}}$$

(36)

If $\alpha_r = \alpha_s = \alpha$ then $V = 1$ and equation (13) is recovered.

We assume, as in the previous subsections, that the MZI is implemented with identical waveguides, and furthermore, that the attenuation of the sensing waveguide only is perturbed by a small amount $\Delta \alpha_s$ due to surface or bulk sensing. Thus, we substitute $\alpha_r = \alpha$ and $\alpha_s = \alpha + \Delta \alpha_s$ into the above and simplify, yielding:

$$P_{\text{out}} = P_{\text{in}} e^{-2\alpha(L_0+L)} \frac{W}{\cosh(\Delta \alpha_s L)} \left( e^{-\Delta \alpha_s L} \cosh(\Delta \alpha_s L) \right)^{\frac{1}{2}} (1 + V \cos \phi_D)$$

(37)

and

$$V = \frac{1}{\cosh(\Delta \alpha_s L)}.$$ 

(38)

From these equations, we note that the perturbation $\Delta \alpha_s$ causes a decrease in the visibility $V$ of the MZI, and depending on its sign, an increase or a decrease in the power penalty relative to lossless waveguides through the factor $W$.

If $\Delta \alpha_s L$ is bounded by $-0.1 \leq \Delta \alpha_s L \leq 0.1$ (for example), then from equations (37) and (38) $W$ and $V$ are bounded by $0.91 < W < 1.11$ and $V > 0.995$, so $W \sim V \sim 1$, equation (13) holds, and the MZI sensor performs essentially as described in the previous subsections. For surface sensing, the perturbation is written $\Delta \alpha_s = \Delta a \partial \alpha_s/\partial a$, and for bulk sensing, it is $\Delta \alpha_s = \Delta n_c \partial \alpha_s/\partial n_c$. Assuming that the sensing length is selected as the nominal propagation length $L = L_c = 1/(2 \alpha)$, the bounds $-0.1 \leq \Delta \alpha_s L \leq 0.1$ imply

$$\left| \frac{\Delta a \partial \alpha/\partial a}{2 \alpha} \right| = \frac{\Delta a \partial k_{\text{eff}}/\partial a}{2 k_{\text{eff}}} \leq 0.1$$

(39)

and

$$\left| \frac{\Delta n_c \partial \alpha/\partial n_c}{2 \alpha} \right| = \frac{\Delta n_c \partial k_{\text{eff}}/\partial n_c}{2 k_{\text{eff}}} \leq 0.1.$$ 

(40)

As demonstrated in the following sections, these bounds are generally amply satisfied for all of the waveguide cases investigated, even for large changes $\Delta a$ or $\Delta n_c$, except near the SPP energy asymptote (the fractional attenuation sensitivities increase as $\lambda_0$ decreases—as discussed below).
4. Single interface

4.1. Approximate analytic expressions

Approximate analytic expressions for the sensing parameters can be derived in the case of the single-interface SPP. These are useful for uncovering trends and helping to explain results for more complex waveguides.

The normalized propagation constant of the single-interface SPP in the case of no adlayer (figure 1(a), \( a = 0 \)) is [1]:

\[
\gamma_{\text{eff},0} = \left( \frac{\varepsilon_{r,m}\varepsilon_{r,c}}{\varepsilon_{r,m} + \varepsilon_{r,c}} \right)^{1/2}, \tag{41}
\]

In the case of no absorption in the carrier fluid (\( k_c = 0 \)), its effective index and normalized attenuation are given by the following approximate expressions, respectively [1]:

\[
n_{\text{eff},0} \approx \left( \frac{\varepsilon_R\varepsilon_{r,c}}{\varepsilon_R - \varepsilon_{r,c}} \right)^{1/2}, \tag{42}
\]

\[
k_{\text{eff},0} \approx \frac{\varepsilon_1}{2\varepsilon_R} \left( \frac{\varepsilon_{r,c}\varepsilon_R}{\varepsilon_R - \varepsilon_{r,c}} \right)^{3/2} = \frac{\varepsilon_1}{2\varepsilon_R^2} n_{\text{eff},0}^3. \tag{43}
\]

If Au was lossless \((\varepsilon_1 = 0)\), then from equation (41) or (42), and from figure 3, \( n_{\text{eff},0} \) would diverge as \( \lambda_0 \) decreases from the infrared, and thus as \( \varepsilon_R \to \varepsilon_{r,c} \), becoming infinite when \( \varepsilon_R = \varepsilon_{r,c} \) at \( \lambda_0 \approx 480 \text{ nm} \) (i.e. at the SPP energy asymptote on an \( \text{E}--\text{k} \) curve [82]). In practice though, \( \varepsilon_1 \neq 0 \), so \( n_{\text{eff},0} \) never becomes infinite. From equation (43) and figure 3, it is apparent that \( k_{\text{eff},0} \) also increases as \( \lambda_0 \) decreases, and evidently, at a much greater rate.

Differentiating equations (42) and (43) with respect to \( n_c \) leads to the effective index and normalized attenuation bulk sensitivities:

\[
\frac{\partial n_{\text{eff},0}}{\partial n_c} = \frac{n_{\text{eff},0}^3}{3 n_c^2} = \frac{n_{\text{eff},0}^3}{n_c^3}, \tag{44}
\]

\[
\frac{\partial k_{\text{eff},0}}{\partial n_c} = \frac{3 \varepsilon_1 n_{\text{eff},0}^5}{2\varepsilon_R^2 n_c^3} \frac{\partial n_{\text{eff},0}}{n_c} = \frac{3 \varepsilon_1}{2\varepsilon_R^2} n_{\text{eff},0}^3 \frac{\partial n_{\text{eff},0}}{n_c} = \frac{3 \varepsilon_1}{2\varepsilon_R^2} n_{\text{eff},0}^3 \frac{\partial n_{\text{eff},0}}{n_c}. \tag{45}
\]

Equation (44) agrees with that given in [83]. The \( H \) parameter is then

\[
H = \frac{\partial n_{\text{eff},0}}{k_{\text{eff},0} n_c} = \frac{2}{\varepsilon_R^2} \frac{\varepsilon_t^2}{n_c^3} = \frac{2}{\varepsilon_R^2} \frac{\varepsilon_t^2}{n_c^3}. \tag{46}
\]

From equation (44), it is noted that \( \partial n_{\text{eff},0}/\partial n_c \) is always greater than 1 since \( n_{\text{eff},0} \) is always greater than \( n_c \) for a purely bound (non-radiative) SPP. From equation (45), it is noted that \( \partial k_{\text{eff},0}/\partial n_c \ll \partial n_{\text{eff},0}/\partial n_c \) as long as \( \varepsilon_1 \ll \varepsilon_R \), which holds for \( \lambda_0 \) greater than about 600 nm (figure 3). In light of the points made above with regards to equations (41)–(43), both bulk sensitivities increase as \( \lambda_0 \) decreases, but comparing equations (44) and (45) reveals that \( \partial k_{\text{eff},0}/\partial n_c \) increases at a greater rate (fifth power in \( n_{\text{eff},0} \)), becoming comparable to \( \partial n_{\text{eff},0}/\partial n_c \) near the SPP energy asymptote. From equation (46), the \( H \) parameter remains large as long as \( \varepsilon_1 \ll \varepsilon_R \).
The introduction of the thin adlayer of thickness $a$ perturbs $n_{\text{eff},0}$ (to first order) \([5, 99]\):

$$n_{\text{eff}} \approx n_{\text{eff},0} + \frac{2\pi a}{\lambda_0} \left( \frac{\varepsilon_{r,a} - \varepsilon_{r,c}}{\varepsilon_{r,a}} \right) \left( \frac{\varepsilon_{r,a} + \varepsilon_R}{\varepsilon_{r,c} + \varepsilon_R} \right) \left( \varepsilon_{r,c} \varepsilon_R \right)^{-1/2} n_{\text{eff},0}^4 \quad (47)$$

and the normalized attenuation $k_{\text{eff},0}$ (to second order) \([99]\):

$$k_{\text{eff}} \approx \frac{k_{\text{eff},0}}{2\varepsilon_R^2 n_{\text{eff},0}^4} + \frac{1}{2\varepsilon_R} \left( \frac{\varepsilon_{r,a} - \varepsilon_{r,c}}{\varepsilon_{r,a}} \right) \left( \frac{\varepsilon_{r,a} + \varepsilon_R}{\varepsilon_{r,c} + \varepsilon_R} \right) \left( \varepsilon_{r,c} \varepsilon_R \right)^{-1/2} n_{\text{eff},0}^4 \cdot (48)$$

Equation (47) is similar to that derived by de Bruijn et al \([100]\). From these equations, it is noted that the adlayer increases both $n_{\text{eff},0}$ and $k_{\text{eff},0}$ (the nominal values for $a = 0$), and that the strength of the perturbation increases with $a$. Differentiating equations (47) and (48) with respect to $a$ leads trivially to the effective index and normalized attenuation surface sensitivities:

$$\frac{\partial n_{\text{eff}}}{\partial a} \approx \frac{2\pi}{\lambda_0} \left( \frac{\varepsilon_{r,a} - \varepsilon_{r,c}}{\varepsilon_{r,a}} \right) \left( \frac{\varepsilon_{r,a} + \varepsilon_R}{\varepsilon_{r,c} + \varepsilon_R} \right) \left( \varepsilon_{r,c} \varepsilon_R \right)^{-1/2} n_{\text{eff},0}^4 \quad (49)$$

and

$$\frac{\partial k_{\text{eff}}}{\partial a} \approx \frac{1}{2\varepsilon_R} \frac{2\pi}{\lambda_0} \left( \frac{\varepsilon_{r,a} - \varepsilon_{r,c}}{\varepsilon_{r,a}} \right) \left( \frac{\varepsilon_{r,a} + \varepsilon_R}{\varepsilon_{r,c} + \varepsilon_R} \right) \left( \varepsilon_{r,c} \varepsilon_R \right)^{-1/2} n_{\text{eff},0}^4 = \frac{1}{2\varepsilon_R} \frac{\partial n_{\text{eff}}}{\partial a}. \quad (50)$$

Equation (49) agrees with that given in \([83]\) when $(\varepsilon_{r,a} + \varepsilon_R)/(\varepsilon_{r,c} + \varepsilon_R) \sim 1$. The $G$ parameter is then

$$G = \frac{\partial n_{\text{eff}} / \partial a}{k_{\text{eff},0}} \approx \frac{2\pi^2}{\varepsilon_R^2} \frac{4\pi}{\varepsilon_1} \lambda_0 \left( \frac{\varepsilon_{r,a} - \varepsilon_{r,c}}{\varepsilon_{r,a}} \right) \left( \frac{\varepsilon_{r,a} + \varepsilon_R}{\varepsilon_{r,c} + \varepsilon_R} \right) \left( \varepsilon_{r,c} \varepsilon_R \right)^{-1/2}. \quad (51)$$

It is observed that both surface sensitivities (equations (49) and (50)) increase with the contrast between the adlayer and the carrier fluid $(\varepsilon_{r,a} - \varepsilon_{r,c})$. From equation (50), it is noted that $\partial k_{\text{eff}} / \partial a \ll \partial n_{\text{eff}} / \partial a$ as long as $\varepsilon_1 \ll \varepsilon_R$, which holds for $\lambda_0$ greater than about 600 nm (figure 3).

In light of the points made with regard to equations (41)–(43), both surface sensitivities increase as $\lambda_0$ decreases, and comparing equations (49) and (50) reveals that they increase roughly at a similar rate (fourth power in $n_{\text{eff},0}$), but the $\varepsilon_1/(2\varepsilon_R)$ term in equation (50) makes $\partial k_{\text{eff}} / \partial a$ comparable near the SPP energy asymptote. From equation (51), the $G$ parameter remains large as long as $\varepsilon_1 \ll \varepsilon_R$.

The ratio of the surface to the bulk effective index sensitivities is given by

$$\frac{\partial n_{\text{eff}} / \partial a}{\partial n_{\text{eff},0} / \partial n_c} = \frac{2\pi}{\lambda_0} \left( \frac{\varepsilon_{r,a} - \varepsilon_{r,c}}{\varepsilon_{r,a}} \right) \left( \frac{\varepsilon_{r,a} + \varepsilon_R}{\varepsilon_{r,c} + \varepsilon_R} \right) \frac{\varepsilon_{r,c} \varepsilon_R}{\pi^2 \varepsilon_R^2} n_{\text{eff},0}^4. \quad (52)$$

The fractional normalized attenuation sensitivity for bulk sensing is

$$\frac{\partial k_{\text{eff},0} / \partial n_c}{n_{\text{eff},0}^4} = \frac{3}{n_c^2} n_{\text{eff},0}^4. \quad (53)$$

Assuming $n_{\text{eff},0} \sim n_c$, with $n_c$ given by the data plotted in figure 2, yields $\sim 2.3$ for the above. Substituting this into equation (40) reveals that the bound is amply satisfied even for large bulk index changes (e.g. $\Delta n_c \sim 10^{-2}$). The fractional normalized attenuation sensitivity for surface sensing is:

$$\frac{\partial k_{\text{eff}} / \partial a}{k_{\text{eff},0}} = \frac{2\pi}{\lambda_0} \left( \frac{\varepsilon_R}{\varepsilon_{r,c}} \right)^{1/2} \left( \frac{\varepsilon_{r,a} - \varepsilon_{r,c}}{\varepsilon_{r,a}} \right) \left( \frac{\varepsilon_{r,a} + \varepsilon_R}{\varepsilon_{r,c} + \varepsilon_R} \right) n_{\text{eff},0}. \quad (54)$$
Assuming $n_{\text{eff,0}} \sim n_c$, and the data plotted in figures 2 and 3, yields $\sim 10^{-2}\ \text{nm}^{-1}$ for the above. Substituting this into equation (39) reveals that the bound is amply satisfied even for large adlayer thickness changes (e.g. $\Delta a \sim 10\ \text{nm}$). These fractional sensitivities are proportional to $n_{\text{eff,0}}$ (or its square), so they increase as $\lambda_0$ decreases.

4.2. Relationship to the waveguide figures of merit

In the case of the single-interface SPP, the parameters $G$ and $H$, which might be regarded as surface and bulk sensing figures of merit, respectively, are connected to the waveguide figures of merit [82, 101]. The figures of merit are defined as benefit-to-cost ratios, where the ‘benefit’ is taken as a confinement measure and the ‘cost’ as the attenuation. They are defined as follows [82]:

\[ M_1 \equiv \frac{1}{\delta w \alpha} \sim \frac{2}{\varepsilon_{t,c}} \frac{\varepsilon_R^{3/2}}{\varepsilon_1}, \quad \text{for} \quad \varepsilon_R \gg \varepsilon_{t,c}, \tag{55} \]

\[ M_2 \equiv \frac{\beta - \beta_c}{\alpha} = \frac{n_{\text{eff}} - n_c}{k_{\text{eff}}} \sim \frac{\varepsilon_R}{\varepsilon_1} \quad \text{for} \quad \varepsilon_R \gg \varepsilon_{t,c}, \tag{56} \]

and

\[ M_3 \equiv \frac{1}{\lambda_{g}} \alpha \sim \frac{1}{\varepsilon_{t,c}^2} \frac{\varepsilon_R^{3/2}}{\varepsilon_1} \quad \text{for} \quad \varepsilon_R \gg \varepsilon_{t,c}, \tag{57} \]

where the inverse $1/\varepsilon$ mode field width ($1/\delta w$), the mode’s distance from the light line ($n_{\text{eff}} - n_c$) and the inverse guided wavelength ($1/\lambda_{g}$) are used as the confinement measures in these definitions, respectively. For waveguides with low dispersion, $M_3$ becomes proportional to $Q$, the unloaded quality factor.

The approximate forms on the right-hand side of equations (55)–(57) hold specifically for the single-interface SPP under the condition that $\varepsilon_R \gg \varepsilon_{t,c}$ [82]. Comparing them with equations (46) and (51) yields the following relationships

\[ H \sim \frac{2\pi}{\varepsilon_{t,c}^{1/2}} M_3 \quad \tag{58} \]

and

\[ G \sim \left( \frac{\varepsilon_{t,a} - \varepsilon_{t,c}}{\varepsilon_{t,a}} \right) \frac{2\omega_p}{c_0} M_2, \quad \tag{59} \]

where $\varepsilon_R \gg \varepsilon_{t,a}$ was also assumed. The Drude model [82]

\[ \varepsilon_{t,m} = -\varepsilon_R - j\varepsilon_1 = 1 - \frac{\omega_p^2}{\omega^2 + 1/\tau_D^2} - j \frac{\omega_p^2/\tau_D}{\omega(\omega^2 + 1/\tau_D^2)} \tag{60} \]

was used to represent the metal permittivity in deriving equation (59) ($\omega_p$ is the plasma frequency and $\tau_D$ the relaxation time).

These relationships to the figures of merit (equations (58) and (59)) are useful in that they relate trends and behaviours deduced for the latter (e.g. [82]) to $G$ and $H$: neglecting the wavelength dependence of $\varepsilon_{t,c}$ and $\varepsilon_{t,a}$, we expect from equation (58), that $H$ will increase with $\lambda_0$ into the Drude region following $M_3$, and from equation (59), we expect $G$ to peak on the short wavelength side of the Drude region following $M_2$ [82].

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Figure 5. Propagation constant and bulk sensing parameters of the single-interface SPP (figure 1(a) with $a = 0$). The solid blue curves are computed using the MoL and the dotted magenta curves are computed using approximate formulae as follows: (a) $n_{\text{eff}}$ with equation (42), (b) $k_{\text{eff}}$ with equation (43) (and then MPA), (c) $\partial n_{\text{eff}}/\partial n_c$ and $\partial k_{\text{eff}}/\partial n_c$ with equations (44) and (45), respectively, and (d) $H$ with equation (46).

We do not expect equations (58) and (59) to hold generally, since they depend implicitly through $G$ and $H$ on the definitions adopted for the waveguide sensitivities (i.e. subsection 2.3), on the geometry and disposition of the bulk and surface sensing region(s) (i.e. $A_a$ and $A_c$ in equations (10) and (12)), and on the actual waveguide design (i.e. figure 1).

4.3. Numerical results

Figure 5 gives the propagation constant and bulk sensing parameters computed for the single-interface SPP (figure 1(a), $a = 0$), figure 6 gives the associated figures of merit $M_2$ and $M_3$, and figure 7 gives the associated surface sensing parameters for $a = 3$ nm. In these figures,
Figure 6. $M_2$ and $M_3$ figures of merit of the single-interface SPP (figure 1(a) with $a = 0$). The solid blue curves are computed using the MoL and the dotted magenta curves are computed using approximate formulae as follows: (a) $M_2$ with equation (56) (approximate form), (b) $M_3$ with equation (57) (approximate form).

Figure 7. Surface sensing parameters of the single-interface SPP (figure 1(a) with $a = 3$ nm). The solid blue curves are computed using the MoL and the dotted magenta curves are computed using approximate formulae as follows: (a) $\partial n_{\text{eff}}/\partial a$ and $\partial k_{\text{eff}}/\partial a$ with equations (49) and (50), respectively, and (b) $G$ with equation (51).
equations (49) and (50) in figure 7(a), and $G$ with equation (51) in figure 7(b). In all cases, the approximate forms agree reasonably well with the modelled results.

The trends with $\lambda_0$ discussed in the previous subsections are observed and confirmed from these results: from figure 5, it is noted that: $n_{\text{eff}}$, MPA, $\partial n_{\text{eff}}/\partial n_c$ and $\partial k_{\text{eff}}/\partial n_c$ increase as $\lambda_0$ decreases, $\partial n_{\text{eff}}/\partial n_c > 1 \gg \partial k_{\text{eff}}/\partial n_c$ over the wavelength range considered, and $H$ decreases as $\lambda_0$ decreases following $M_3$ in figure 6(b) (comparing figures 5(d) and 6(b) verifies equation (58)). The left-hand side of equation (40) was computed assuming $\Delta n_c = 10^{-2}$, yielding a maximum value of 0.014 over the wavelength range considered, which amply satisfies the bound of equation (40) (as pointed out with regard to equation (53)). From figure 7(a), $\partial n_{\text{eff}}/\partial a$ and $\partial k_{\text{eff}}/\partial a$ increase as $\lambda_0$ decreases and $\partial n_{\text{eff}}/\partial a \gg \partial k_{\text{eff}}/\partial a$, while figure 7(b) shows that $G$ peaks near the short wavelength side of the Drude region for Au, at $\lambda_0 \sim 840$ nm, following $M_2$ in figure 6(a) (comparing figures 6(a) and 7(b) validates equation (59)). The left-hand side of equation (39) was computed assuming $\Delta a = 10$ nm, yielding a maximum value of 0.075 over the wavelength range considered, which amply satisfies the bound of equation (39) (as pointed out with regard to equation (54)).

From figure 5(b), a bump in the MPA is observed at $\lambda_0 \sim 1450$ nm, corresponding to a peak in the absorption of H$_2$O at this wavelength, as is apparent from figures 2(b) and 3(b). The absorption of H$_2$O therefore begins to have a significant effect on the performance of the waveguide at this wavelength (and beyond). Valleys are also noted in $H$, $M_2$, $M_3$ and $G$ near this wavelength.

From figure 7(b), preferred wavelengths for operating a surface sensor would be in the near-infrared, with $\lambda_0 = 850$ nm being a good choice since this is where $G$ is largest. From figure 5(d), it is apparent that preferred wavelengths for operating a bulk sensor are found deeper into the infrared where $H$ is largest, with $\lambda_0 = 1310$ nm (just on the left of the H$_2$O absorption peak at $\lambda_0 \sim 1450$ nm) being a good choice.

5. MIM

Figure 8 gives the propagation constant and bulk sensing parameters computed for the $s_b$ mode in the MIM with no adlayers (figure 1(b), $a = 0$) as a function of $t$. Figure 9 gives the associated surface sensing parameters for $a = 3$ nm, assuming adlayers on both metal surfaces as sketched in figure 1(b).

The smallest value of $t$ considered is 20 nm. In selecting a minimum value for $t$, consideration must be given to the flow conditions of the carrier fluid (increasing resistance to flow as $t$ decreases [102]), to the thickness needed for the receptor layers (e.g. $a$), and to the size of the target (bio)chemical analyte (many nanometres for large biomolecules). It seems then that $t \sim 20$ nm would be close to a lower limit.

As expected, the propagation constant and the sensing parameters all converge to those of the single-interface SPP as $t$ increases. At small $t$, the $s_b$ mode is more tightly confined, so the propagation constant and the sensitivities are larger: at $t = 20$ nm for example, $n_{\text{eff}}$ and $\partial n_{\text{eff}}/\partial n_c$ are larger by factors of about 1.5–3, whereas the MPA and $\partial n_{\text{eff}}/\partial a$ are larger by factors of about 10–100 over the wavelength range investigated. Consequently, $H$ decreases with decreasing $t$, as is evident from figure 8(e), because the bulk sensitivity increases at a smaller rate than the attenuation. Indeed, $H$ is smaller at $t = 20$ nm by a factor of about 3–20 compared with the single-interface SPP. On the other hand, $G$ increases with decreasing $t$, as is evident from figure 9(c), because the surface sensitivity increases at a greater rate than the
Figure 8. Propagation constant and bulk sensing parameters of the $s_b$ mode in the MIM (figure 1(b) with $a = 0$). In (a), (b), (c), and (e), the red curve corresponds to the case $t = 20$ nm and the blue curves are plotted for increasing $t$ in steps of 10 nm. (c) and (d) $\partial n_{\text{eff}} / \partial n_c$ plotted as semi-log curves and contours, respectively. (e) $H$ plotted as semi-log curves. (f) $\log_{10}(H)$ plotted as contours.

attenuation. Indeed, $G$ is greater at $t = 20$ nm by a factor of about 2–4 compared with the single-interface SPP.

Except for $H$, the wavelength responses of the propagation constant and sensing parameters are similar to those of the single-interface SPP, in that the main features and trends are retained as $t$ decreases. For instance, $G$ still peaks near the short wavelength side of the
Figure 9. Surface-sensing parameters of the $s_0$ mode in the MIM (figure 1(b) with $a = 3$ nm). In (a) and (c), the red curve corresponds to the case $t = 20$ nm and the blue curves are plotted for increasing $t$ in steps of 10 nm. (a) and (b) $\partial n_{\text{eff}}/\partial a$ in nm$^{-1}$ plotted as semi-log curves and contours, respectively. (c) $G$ in nm$^{-1}$ plotted as semi-log curves. (d) $\log_{10}(G)$ with $G$ in nm$^{-1}$ plotted as contours.

Drude region, at a similar value of $\lambda_0$. The response of $H$, however, changes as $t$ decreases: at $t = 20$ nm for example, $H$ is largest near the short wavelength side of the Drude region (like $G$), whereas for large $t$, and for the single-interface SPP, $H$ increases with $\lambda_0$ into the infrared.

The normalized attenuation sensitivities, $\partial k_{\text{eff}}/\partial n_c$ and $\partial k_{\text{eff}}/\partial a$ (not shown), remain one to two orders of magnitude smaller than the effective index sensitivities, $\partial n_{\text{eff}}/\partial n_c$ and $\partial n_{\text{eff}}/\partial a$, over the parameter ranges investigated, and equation (40) remains amply satisfied (for $\Delta n_c = 10^{-2}$). Equation (39) is satisfied over most of both ranges (for $\Delta a = 10$ nm), but the left-hand side increases to about 0.15 at $t = 20$ nm and $\lambda_0 = 600$ nm.

The surface sensitivity (and propagation constant) depend increasingly on $a$ as $t$ decreases because the mode width essentially follows $t$ [82], and $t$ can become comparable to $a$. Indeed, for $t = 20$ nm, the two adlayers ($2a = 6$ nm) cover a significant portion of the mode width.
This is of course the reason for the very large surface sensitivities, but it also means that the computations depend on the value selected for \(a\) (3 nm herein).

6. IMI

6.1. Two adlayers

Figure 10 gives the propagation constant and bulk sensing parameters computed for the \(s_b\) mode in the IMI with no adlayers (figure 1(c), \(a = 0\)) as a function of \(t\), down to \(t = 5\) nm. Figure 11 gives the associated surface sensing parameters for \(a = 3\) nm, assuming adlayers on both sides of the metal film as shown. (Note that the MPA is plotted on a different scale.)

In selecting a minimum value for \(t\), consideration must be given to the mechanical stability of a free-standing metal film in a flow channel, the situation sketched in figure 1(c). Corrugated free-standing metal films, having a thickness down to \(t = 26\) nm, and bounded by air, have been demonstrated experimentally, with both coupled modes \((s_b\) and \(a_b)\) excited thereon [21]. It seems then that \(t \sim 20\) nm would be an aggressive lower limit. Furthermore, it is expected that the optical parameters of an Au film (say, as-evaporated) would begin to change from their bulk values as the thickness becomes thinner than \(t \sim 20\) nm (e.g. [34]), so the computations for decreasing \(t\) beyond about 20 nm could deviate increasingly from reality.

As expected, the propagation constant and sensing parameters all converge to those of the single-interface SPP as \(t\) increases. At small \(t\), the \(s_b\) mode is less tightly confined, so the propagation constant and the sensitivities are smaller: at \(t = 20\) nm for example, \(n_{\text{eff}}\) and \(\partial n_{\text{eff}}/\partial n_c\) are slightly smaller, while the MPA and \(\partial n_{\text{eff}}/\partial a\) are smaller by factors of about 100 and 10, respectively, over the wavelength range investigated. Consequently, both \(H\) and \(G\) increase with decreasing \(t\), as is evident from figures 10(e) and 11(c) because both the bulk and surface sensitivities decrease at a smaller rate than the attenuation. Indeed, \(H\) and \(G\) are greater at \(t = 20\) nm by more than one order of magnitude compared with the single-interface SPP.

The wavelength responses of the propagation constant and sensing parameters evolve somewhat from those of the single-interface SPP as \(t\) decreases, in part because the attenuation of the \(s_b\) mode decreases so the absorption spectrum of \(H_2O\) becomes more dominant, as is readily noted from figure 10(b) at small \(t\). In particular, a sharp increase in the MPA is noted for small \(t\) at long wavelengths due to the increase in the absorption of \(H_2O\) in the infrared. Still, the propagation constant and the sensitivities increase as \(\lambda_0\) decreases, which is consistent with the single-interface SPP. From figures 10(f) and 11(d), it is noted that both \(H\) and \(G\) are largest near the short wavelength side of the Drude region.

The normalized attenuation sensitivities, \(\partial k_{\text{eff}}/\partial n_c\) and \(\partial k_{\text{eff}}/\partial a\) (not shown), remain one to a few orders of magnitude smaller than the effective index sensitivities, \(\partial n_{\text{eff}}/\partial n_c\) and \(\partial n_{\text{eff}}/\partial a\), over the parameter ranges investigated, and equation (40) remains amply satisfied (for \(\Delta n_c = 10^{-2}\)). Equation (39) is satisfied over most of both ranges (for \(\Delta a = 10\) nm), but the left-hand side increases beyond 0.1 for \(t < 20\) nm at short wavelengths.

6.2. One adlayer

Figure 12 gives the surface sensitivity and the \(G\) parameter computed for the \(s_b\) mode in the IMI with one adlayer (figure 1(d), \(a = 3\) nm) as a function of \(t\). The propagation constant remains similar to the case plotted in figures 10(a) and (b).

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Figure 10. Propagation constant and bulk sensing parameters of the $s_b$ mode in the IMI (figure 1(c) with $a = 0$). In (a), (b), (c) and (e), the red curve corresponds to the case $t = 5$ nm and the blue curves are plotted for increasing $t$ in steps of 5 nm. (c) and (d) $\partial n_{\text{eff}}/\partial n_c$ plotted as semi-log curves and contours, respectively. (e) $H$ plotted as semi-log curves. (f) $\log_{10}(H)$ plotted as contours.

This structure represents a situation where the bottom fluidic region is replaced with a solid substrate index-matched to the fluid. In this situation, only one adlayer may form on the metal film, at the top metal/fluid interface. This structure seems more practical than the previous one (figure 1(c)), where the metal film was assumed to be free-standing within the fluid. The smallest
value of $t$ considered is still 5 nm. The complex index of H$_2$O was adopted as the index of the solid substrate, for simplicity and to ensure perfect index matching to the fluidic region above the metal.

It is noted from figure 12 that the sensing parameters vanish at large $t$ instead of converging to those of the single-interface SPP plotted in figure 7. This is in contrast to the two adlayer situation explored in the previous subsection and plotted in figure 11. As observed from figure 12(a), the surface sensitivity first increases to reach a maximum value near $t \sim 60$–$80$ nm (depending on $\lambda_0$), and then tends to zero as $t$ increases further. The MPA also increases with $t$ (figure 10(b)) but at a greater rate so $G$ remains largest at small $t$. $\partial n_{\text{eff}}/\partial a$ and $G$ are slightly smaller than in the two adlayer case at small $t$ over the wavelength range investigated (by a factor of about 2 for $t = 20$ nm).

**Figure 11.** Surface sensing parameters of the $s_b$ mode in the IMI with two adlayers (figure 1(c) with $a = 3$ nm). In (a) and (c), the red curve corresponds to the case $t = 5$ nm and the blue curves are plotted for increasing $t$ in steps of 5 nm. (a) and (b) $\partial n_{\text{eff}}/\partial a$ in nm$^{-1}$ plotted as semi-log curves and contours, respectively. (c) $G$ in nm$^{-1}$ plotted as semi-log curves. (d) $\log_{10}(G)$ with $G$ in nm$^{-1}$ plotted as contours.
Figure 12. Surface sensing parameters of the $s_b$ mode in the IMI with one adlayer (figure 1(d) with $a = 3$ nm). In (a) and (c), the red curve corresponds to the case $t = 5$ nm and the blue curves are plotted for increasing $t$ in steps of 5 nm. (a) and (b) $\partial n_{eff}/\partial a$ in nm$^{-1}$ plotted as semi-log curves and contours, respectively. (c) $G$ in nm$^{-1}$ plotted as semi-log curves. (d) $\log_{10}(G)$ with $G$ in nm$^{-1}$ plotted as contours.

The behaviour observed for $\partial n_{eff}/\partial a$ is explained by inspection of the mode fields with increasing $t$: the $s_b$ mode becomes more tightly confined as $t$ increases from small values (e.g. $t \sim 20$ nm) causing $\partial n_{eff}/\partial a$ to increase. But the $s_b$ mode also evolves with increasing $t$ into the single-interface SPP localized at the bottom metal/dielectric interface opposite the adlayer (on the low index side), causing $\partial n_{eff}/\partial a$ to become zero at large $t$. These processes compete such that a maximum in $\partial n_{eff}/\partial a$ arises.

The wavelength response of the propagation constant and sensing parameters are similar to the two adlayer situation explored in the previous subsection at small $t$. In particular, it is noted that $G$ is also largest near the short wavelength side of the Drude region.

The normalized attenuation sensitivity $\partial k_{eff}/\partial a$ (not shown) remains one to a few orders of magnitude smaller than the effective index sensitivity $\partial n_{eff}/\partial a$ over the parameter ranges.
investigated, and equation (39) is satisfied (for Δa = 10 nm) over most of the ranges, but the left-hand side increases beyond 0.1 at short wavelengths.

The bulk sensitivity and H parameter have not been computed, but would need to be assessed with regard to changes in the index of the fluidic region above the metal film only, and thus both are expected to be smaller by a factor of about 2 compared with the structure explored in the previous subsection (a free-standing metal film in fluid—figure 1(c)).

7. Membrane waveguide

The structure depicted in figure 1(e) represents an alternative approach to sensing using the IMI. It consists of a metal film on a thin free-standing dielectric membrane of thickness $d$ and relative permittivity $\varepsilon_{r,d}$ bounded on both sides by the sensing fluid [46, 102, 103]. This structure seems more practical than the free-standing metal film of figure 1(c) since the dielectric membrane could provide stronger mechanical support.

This ‘membrane waveguide’ allows a perturbed version of the $s_b$ mode to propagate in any transparent sensing fluid, in the gaseous or liquid states, always ensuring that the structure remains essentially symmetric. If the membrane is not too thick (optically) then the perturbed $s_b$ mode retains the main characteristics of the unperturbed one ($d = 0$), including a low attenuation at small $t$. The design and performance of this structure was discussed in detail in [103]; here, we focus on its surface sensing parameters.

The top metal/fluid interface is used as the sensing surface with the adlayer located thereupon. $\text{Si}_3\text{N}_4$ is assumed as the membrane material with the measured optical parameters plotted in figure 2 [89] used for the computations.

7.1. Results at $\lambda_0 = 1310$ nm

Figure 13 gives the propagation constant and surface sensing parameters of the $s_b$ mode in this waveguide ($a = 3$ nm) at $\lambda_0 = 1310$ nm over a portion of the $t$, $d$ design space, including $t = d = 1$ nm as the inferior limit (for the purposes of recovering limiting waveguides). From figure 13 it is noted that $n_{\text{eff}}$, MPA and $\partial n_{\text{eff}}/\partial a$ all increase with $t$ and $d$ over the ranges considered. This behaviour is explained as follows. For a fixed $t$, the mode fields become more confined and localized to the metal/fluid interface opposite the membrane as $d$ increases, causing $n_{\text{eff}}$, MPA and $\partial n_{\text{eff}}/\partial a$ to increase. The same trend is observed for a fixed $d$ as $t$ is increased. Increasing both $t$ and $d$ eventually transforms the $s_b$ mode into the single-interface SPP localized at the metal/fluid interface opposite the membrane (i.e. the metal interface with the low-index medium). The $s_b$ mode remains long-range as long as $t$ and $d$ both remain small, below about 30 nm in this example implementation.

Thus, four extreme cases are recovered for extreme values of $t$ and $d$: for $d = 0$, the IMI with a single adlayer (figure 1(d)) is recovered; for $t$ and $d$ both large the single-interface SPP localized at the metal/fluid interface (figure 1(a)) is recovered; for $t$ and $d$ both small the long-range $s_b$ mode is recovered; and for $t = 0$ the dielectric slab is recovered.

From figure 13(d), it is noted that $G$ is largest for large $d$ and $t \to 0$, since this structure tends to a high-confinement $\text{Si}_3\text{N}_4$ core in $\text{H}_2\text{O}$ with no Au, and therefore without its loss contribution. The MPA for $t \to 0$ is nonzero due to absorption in $\text{H}_2\text{O}$, and since a small $d$ provides less surface sensitivity (figure 13(c)), then $G$ decreases as $t$, $d \to 0$, as observed.
Figure 13. Propagation constant and surface sensing parameters of the $s_b$ mode in the membrane waveguide (figure 1(d) with $a = 3$ nm) at $\lambda_0 = 1310$ nm. (a) $n_{\text{eff}}$. (b) MPA (dB mm$^{-1}$). (c) $\partial n_{\text{eff}}/\partial a$ (nm$^{-1}$); the three back dotted contours correspond to $\partial k_{\text{eff}}/\partial a = 0$ and $\partial k_{\text{eff}}/\partial a = \pm 5 \times 10^{-7}$ nm$^{-1}$ (the negative one is to the right of $\partial k_{\text{eff}}/\partial a = 0$). (d) $\log_{10}(G)$ with $G$ in nm$^{-1}$; the grey solid curve highlights a ridge in $G$.

Although the largest $G$ is achieved for $t = 0$, the Au film remains desirable for its chemical and electrical properties.

Choosing an extremely thin Au film ($t \sim 2$ nm) on a thick membrane might seem like a good idea since a large $G$ is achieved. However, a high quality extremely thin metal film is impractical to fabricate and would likely exhibit optical parameters that are significantly different from the bulk (as mentioned earlier). Interestingly, a $G$ ‘ridge’ persists into the region $t$, $d < 30$ nm along the grey solid curve sketched onto the plot. Good designs are located along and near this ridge. For $t$ selected in this region, say $t = 15$ nm, increasing $d$ improves $G$ up to a local maximum from which it then decreases. Likewise, for $d$ selected in this region, say $d = 15$ nm,
increasing $t$ improves $G$ to a local maximum from which it then decreases. Therefore, adding a dielectric membrane to a metal film in H$_2$O increases $\partial n_{\text{eff}}/\partial a$ of the $s_b$ mode more rapidly than its attenuation. Alternatively, covering a thin dielectric slab in H$_2$O with a thin metal film achieves the same result. Insofar as $G$ is concerned, a good design region in this example is $t$ and $d$ in the range of 15–30 nm. The $M_2$ figure of merit computed for this structure [103] is distributed similarly to $G$ over the $t$, $d$ design space, so good choices for $t$ and $d$ yield high values for both.

As noted from figure 13(c), the normalized attenuation sensitivity $\partial n_{\text{eff}}/\partial a$ has a zero contour over a portion of the design space, and remains one to a few orders of magnitude smaller than $\partial n_{\text{eff}}/\partial a$. Equation (39) is amply satisfied near this region. The sign of $\partial k_{\text{eff}}/\partial a$ can be positive or negative.

7.2. Results at $\lambda_0 = 850$ nm

Figure 14 gives the propagation constant and surface sensing parameters of the $s_b$ mode at $\lambda_0 = 850$ nm over the same region of the $t$, $d$ design space. The $s_b$ mode behaves in a qualitatively similar manner at this wavelength, but greater surface sensitivities and $G$ values are accessible compared with $\lambda_0 = 1310$ nm.

7.3. Wavelength response

The four movies, available from stacks.iop.org/NJP/10/105010/mmedia, show how $n_{\text{eff}}$, $\log_{10}$MPA, $\log_{10} \partial n_{\text{eff}}/\partial a$ and $\log_{10} G$ of the $s_b$ mode vary over the wavelength range $600 \leq \lambda_0 \leq 1600$ nm (in wavelength steps of 10 nm), for the full $t$, $d$ design space considered in figures 13 and 14.

Figure 15 summarizes the wavelength response of the propagation constant and surface sensing parameters of the $s_b$ mode for two membrane thicknesses ($d = 20$ and 30 nm) and three metal thicknesses ($t = 15$, 20 and 25 nm) representative of good designs. The propagation constant and the sensitivity are noted to increase as $\lambda_0$ decreases. The sharp increase in the MPA at long wavelengths (figure 15(b)) is due to the increase in the absorption of H$_2$O in this region (figure 3(b)). Except for the case $t = 15$ nm and $d = 20$ nm, the short wavelength side of the Drude region yields the largest $G$ (figure 15(d)), with $\lambda_0 = 850$ nm being a good choice for the operating wavelength. $\lambda_0 = 1310$ nm is located near the edge of the drop in $G$ on the long wavelength side, and provides lower values of $G$, but remains nonetheless a reasonably good choice.

8. Discussion

From the results given and described in the previous sections, trends are summarized and the structures are compared as follows.

For the single interface, MIM and IMI (two adlayers—figure 1(c)), the $n_{\text{eff}}$, MPA and all of the waveguide sensitivities ($\partial n_{\text{eff}}/\partial n_c$, $\partial k_{\text{eff}}/\partial n_c$, $\partial n_{\text{eff}}/\partial a$ and $\partial k_{\text{eff}}/\partial a$) increase with confinement, so they all increase with decreasing $\lambda_0$, with decreasing $t$ in the MIM, and with increasing $t$ in the IMI. The trends with $\lambda_0$ also hold for the variants of the IMI (figures 1(d) and (e)), but the trends with thickness are different: in the case of the one adlayer IMI (figure 1(d)), the maximum surface sensitivity is observed at a specific $t$, near 60 nm, and in the case of the
Figure 14. Propagation constant and surface sensing parameters of the $s_b$ mode in the membrane waveguide (figure 1(d) with $a = 3$ nm) at $\lambda_0 = 850$ nm. (a) $n_{\text{eff}}$. (b) MPA (dB mm$^{-1}$). (c) $\partial n_{\text{eff}}/\partial a$ (nm$^{-1}$); the three back dotted contours correspond to $\partial k_{\text{eff}}/\partial a = 0$ and $\partial k_{\text{eff}}/\partial a = \pm 1 \times 10^{-6}$ nm$^{-1}$ (the negative one is to the right of $\partial k_{\text{eff}}/\partial a = 0$). (d) $\log_{10}(G)$ with $G$ in nm$^{-1}$.

In order of largest to smallest surface sensitivity, the structures are ranked as follows: (i) thin MIM ($t \sim 20$ nm), (ii) single-interface, (iii) thin IMI with two adlayers ($t \sim 20$ nm), (iv) thin membrane waveguide ($t \sim 15$ nm and $d \sim 20$ nm) and (v) thin IMI with one adlayer ($t \sim 20$ nm). The surface sensitivity of the thin MIM can be up to two orders of magnitude...
larger than that of the single interface, whereas the surface sensitivity of the three thin IMIs are smaller by a factor of about 2–5.

But as emphasized in section 3, it is maximizing $G$ (or $H$) that matters for maximizing the sensitivity (and minimizing the detection limit) of a single-output MZI sensor. $G$ increases as $t$ decreases in the IMI and MIM, so both of these structures offer improvements for surface sensing over the single interface in this geometry.

In order of largest to smallest $G$, the structures are ranked as follows: (i) thin IMI with two adlayers ($t \sim 20$ nm), (ii) thin membrane waveguide ($t \sim 15$ nm and $d \sim 20$ nm), (iii) thin IMI with one adlayer ($t \sim 20$ nm), (iv) thin MIM ($t \sim 20$ nm) and (v) single-interface. $G$ of the thin IMI with two adlayers is about one order of magnitude larger than $G$ of the single interface, whereas $G$ of the membrane waveguide and of the MIM are larger by factors of about 6 and 3, respectively.

Figure 15. Wavelength response of the propagation constant and surface sensing parameters of the $s_b$ mode in the membrane waveguide (figure 1(d) with $a = 3$ nm) for six combinations of $t$ and $d$: solid blue line $t = 15$ nm, $d = 20$ nm; dashed blue line $t = 15$ nm, $d = 30$ nm; solid green line $t = 20$ nm, $d = 20$ nm; dashed green line $t = 20$ nm, $d = 30$ nm; solid red line $t = 25$ nm, $d = 20$ nm; dashed red line $t = 25$ nm, $d = 30$ nm. (a) $n_{\text{eff}}$. (b) MPA (dB mm$^{-1}$). (c) $\partial n_{\text{eff}} / \partial a$ (nm$^{-1}$). (d) $G$ nm$^{-1}$.
In order of largest to smallest bulk sensitivity, the structures are ranked as follows: (i) thin MIM \((t \sim 20\text{ nm})\), (ii) single-interface and (iii) thin IMI \((t \sim 20\text{ nm})\). The bulk sensitivity of the thin MIM can be larger than that of the single interface by a factor of about 3, whereas the bulk sensitivity of the thin IMI is slightly smaller than that of the single interface.

On the other hand, \(H\) increases with decreasing \(t\) in the IMI, but \(H\) decreases with decreasing \(t\) in the MIM, so only the IMI offers an improvement over the single interface for bulk sensing. In order of largest to smallest \(H\), the structures are ranked as follows: (i) thin IMIs \((t \sim 20\text{ nm})\), (ii) single-interface and (iii) thin MIM \((t \sim 20\text{ nm})\). \(H\) of the thin IMI is about one order of magnitude larger than \(H\) of the single interface, whereas \(H\) of the thin MIM is about one order of magnitude smaller.

\(G\) is largest near the short wavelength side of the Drude region \((\sim 850\text{ nm})\) in all structures. \(H\) is also largest near the short wavelength side of the Drude region for the MIM and IMI, but is largest deeper into the infrared for the single interface.

Tables 1 and 2 summarize one good design for each structure at \(\lambda_0 = 850\) and 1310 nm, respectively. The MZI sensitivities, \(\partial P_{\text{out}}/\partial a\) and \(\partial P_{\text{out}}/\partial n_c\), are computed using equations (22) and (30), respectively, assuming \(P_{\text{in}} = 1\text{ mW}\), \(L_0 = L_e\) and \(\sin(\phi_D) = -1\). The detection limits \(\Delta\Gamma_{\text{min}}\) and \(\Delta n_c,\text{min}\) are computed using equations (27) and (32) assuming \(\Delta P_{\text{out},\text{min}} = 10\text{ nW}\). This power level \((\Delta P_{\text{out},\text{min}} = 10\text{ nW} = -50\text{ dBm})\) is well above the noise floor of good detection optoelectronics, but could still prove challenging to achieve given the potential sources of noise and fluctuation along the sensor from the laser to the output. Other MZI architectures offering differential detection and power referencing (such as the dual output MZI) might prove advantageous in this regard, as would phase modulation applied to one of the arms.

From tables 1 and 2, it is observed that an MZI implemented with the single interface has detection limits for bulk and surface sensing that are comparable to conventional SPR [6]–[8], whereas an IMI-based MZI could improve on these by an order of magnitude or more, depending on how low \(\Delta P_{\text{out},\text{min}}\) and how high \(P_{\text{in}}\) can be in practice.

It is interesting to note that the IMIs and the MIM offer an improvement for surface sensing over the single interface, despite the fact that they are at opposite ends of the confinement–attenuation trade-off and have very different dimensional scales: an MZI implemented with an IMI would be about 1–4 mm long, whereas an MZI implemented with a MIM would be about 3–5 \(\mu\text{m}\) long. An MZI implemented with the single interface would have a length in between these extremes, of about 50–200 \(\mu\text{m}\).

These lengths refer to the end-to-end length of the MZI, which is taken as \(L + L_0 = 2L_e\), and therefore assumes that the splitter/combiner and any input/output access lines can be designed such that their total path length \(L_0\) is also equal to \(L_e\) (as mentioned in simplifying to equation (24)). We have verified that this assumption holds approximately for the LRSPP in the symmetric metal stripe [26, 57] \((L_0 \approx 1.3L_e)\), and note that it is consistent (perhaps conservative) for an implementation based on MIM-like structures [38, 48, 53, 58] or on the single interface [47, 54, 60]. The exponential in equation (13) then reduces to \(e^{-2}\), which is the MZI attenuation factor (corresponding insertion loss of 8.69 dB) when the interference term is set to 1.

The surface sensitivity of a typical dielectric waveguide designed for sensing (e.g. [83]) is much lower than that of the single interface or thin MIM, but comparable to that of the thin IMIs. However, the attenuation of dielectric waveguides is also generally lower than all of the SPP waveguides so larger \(G\) and \(H\) parameters (and thus more sensitive MZIs) should be achievable. However, and equally importantly, the availability of good surface chemistries for
Table 1. Summary of waveguide designs and operating parameters at $\lambda_0 = 850$ nm. The MZI sensitivities, $\partial P_{\text{out}}/\partial a$ and $\partial P_{\text{out}}/\partial n_c$, are computed using equations (22) and (30), assuming $P_{\text{in}} = 1$ mW, $L_0 = L_e$ and $\sin(\phi_D) = -1$. The detection limits $\Delta n_{c,\text{min}}$ are then computed using equations (27) and (32) assuming $\Delta P_{\text{out, min}} = 10$ nW.

| Mode | Waveguide ($a = 0$) | Bulk sensitivities ($a = 0$) |
|------|---------------------|-----------------------------|
|      | $n_{\text{eff}}$ (MPA) | $L_e$ (RIU$^{-1}$) | $\partial n_{\text{eff}}/\partial n_c$ (RIU$^{-1}$) | $\partial k_{\text{eff}}/\partial n_c$ (RIU$^{-1}$) | $H$ (RIU$^{-1}$) | $\partial P_{\text{out}}/\partial n_c$ (mW RIU$^{-1}$) | $\Delta n_{c,\text{min}}$ (RIU) |
| SPP  | single interface     |                             |                             |                             |                             |                             |                             |
| $s_b$, MIM ($t = 20$ nm) | 1.3637 | 0.18 | 24.1 | 1.09 | 6.81 $\times 10^{-3}$ | 339 | 13.2 | 7.6 $\times 10^{-7}$ |
| $s_b$, MIM ($t = 20$ nm) | 2.5397 | 2.62 | 1.7 | 2.06 | 4.72 $\times 10^{-3}$ | 50 | 1.7 | 5.9 $\times 10^{-6}$ |
| $s_b$, IMI, 1 adlayer ($t = 20$ nm) | 1.3310 | 6.03 $\times 10^{-3}$ | 720.2 | 1.02 | 2.94 $\times 10^{-4}$ | 10861 | 367.5 | 2.7 $\times 10^{-8}$ |
| $s_b$, membrane wg ($t = 15$ nm, $d = 20$ nm) | 1.3356 | 7.78 $\times 10^{-3}$ | 558.2 |                             |                             |                             |                             |
|      | Surface sensitivities ($a = 3$ nm) |                             |                             |                             |                             |                             |                             |
|      | $\partial n_{\text{eff}}/\partial a$ (nm$^{-1}$) | $\partial k_{\text{eff}}/\partial a$ (nm$^{-1}$) | $G$ (nm$^{-1}$) | $\partial P_{\text{out}}/\partial a$ (mW nm$^{-1}$) | $\Delta \Gamma_{\text{min}}$ (pg mm$^{-2}$) |
| SPP  | single interface     |                             |                             |                             |                             |                             |                             |
| $s_b$, MIM ($t = 20$ nm) | 7.88 $\times 10^{-4}$ | 3.42 $\times 10^{-5}$ | 0.28 | 9.5 | 0.99 |
| $s_b$, IMI, 2 adlayers ($t = 20$ nm) | 3.39 $\times 10^{-2}$ | 8.23 $\times 10^{-4}$ | 0.83 | 28 | 0.33 |
| $s_b$, IMI, 1 adlayer ($t = 20$ nm) | 3.02 $\times 10^{-4}$ | 2.21 $\times 10^{-6}$ | 3.22 | 109 | 0.086 |
| $s_b$, membrane wg ($t = 15$ nm, $d = 20$ nm) | 1.44 $\times 10^{-4}$ | 1.23 $\times 10^{-6}$ | 1.53 | 52 | 0.18 |

Au (e.g. [104]), which are necessary for interfacing the sensor to the (bio)chemical world, is also a decisive consideration.

9. Concluding remarks

The performance of a generic single-output MZI implemented with attenuating waveguides was considered initially. The MZI sensitivity was defined in terms of its output power (the only measurable), as the change in output power relative to the change in adlayer thickness for the case of surface sensing ($\partial P_{\text{out}}/\partial a$), and as the change in output power relative to the change in

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Table 2. Summary of waveguide designs and operating parameters at $\lambda_0 = 1310$ nm. The MZI sensitivities, $\partial P_{\text{out}}/\partial a$ and $\partial P_{\text{out}}/\partial n_c$, are computed using equations (22) and (30), respectively, assuming $P_{\text{in}} = 1$ mW, $L_0 = L_e$ and $\sin(\phi_D) = -1$. The detection limits $\Delta \Gamma_{\text{min}}$ and $\Delta n_{\text{c,min}}$ are then computed using equations (27) and (32) assuming $\Delta P_{\text{out,min}} = 10$ nW.

| Mode | Waveguide ($a = 0$) | Bulk sensitivities ($a = 0$) |
|------|---------------------|-------------------------------|
|      | $n_{\text{eff}}$ (MPA) | $L_e$ (RIU)$^{-1}$ | $\partial n_{\text{eff}}/\partial n_c$ (RIU)$^{-1}$ | $\partial \tilde{k}_{\text{eff}}/\partial n_c$ (RIU)$^{-1}$ | $H$ (RIU)$^{-1}$ | $\partial P_{\text{out}}/\partial n_c$ (mW RIU)$^{-1}$ | $\Delta n_{\text{c,min}}$ (RIU) |
| SPP  | single interface     | 1.3294 | 5.24 x 10$^{-2}$ | 82.9 | 1.03 | 2.89 x 10$^{-3}$ | 819 | 27.7 | 3.6 x 10$^{-7}$ |
| $s_b$, MIM (t = 20 nm) | 2.4044 | 1.73 | 2.5 | 1.872 | 3.81 x 10$^{-2}$ | 45 | 1.5 | 6.6 x 10$^{-6}$ |
| $s_b$, IMI, 2 adlayers (t = 20 nm) | 1.3182 | 2.13 x 10$^{-3}$ | 2039 | 1.01 | 9.33 x 10$^{-5}$ | 19754 | 668.4 | 1.5 x 10$^{-8}$ |
| $s_b$, IMI, 1 adlayer (t = 20 nm) | 1.3182 | 2.13 x 10$^{-3}$ | 2039 | |
| $s_b$, membrane wg (t = 15 nm, d = 20 nm) | 1.3201 | 2.80 x 10$^{-3}$ | 1551.1 | |

| Mode | Surface sensitivities ($a = 3$ nm) |
|------|----------------------------------|
|      | $\partial n_{\text{eff}}/\partial a$ (nm)$^{-1}$ | $\partial \tilde{k}_{\text{eff}}/\partial a$ (nm)$^{-1}$ | $G$ (nm)$^{-1}$ | $\partial P_{\text{out}}/\partial a$ (µW nm)$^{-1}$ | $\Delta \Gamma_{\text{min}}$ (pg mm)$^{-2}$ |
| SPP  | single interface                 | 2.94 x 10$^{-4}$ | 1.41 x 10$^{-5}$ | 0.23 | 7.9 | 1.24 |
| $s_b$, MIM (t = 20 nm) | 3.18 x 10$^{-2}$ | 6.65 x 10$^{-4}$ | 0.77 | 26 | 0.38 |
| $s_b$, IMI, 2 adlayers (t = 20 nm) | 1.25 x 10$^{-4}$ | 8.42 x 10$^{-7}$ | 2.44 | 83 | 0.12 |
| $s_b$, IMI, 1 adlayer (t = 20 nm) | 5.93 x 10$^{-5}$ | 5.10 x 10$^{-7}$ | 1.16 | 39 | 0.25 |
| $s_b$, membrane wg (t = 15 nm, d = 20 nm) | 9.10 x 10$^{-5}$ | -6.0 x 10$^{-7}$ | 1.35 | 46 | 0.21 |

bulk refractive index for the case of bulk sensing ($\partial P_{\text{out}}/\partial n_c$). It was noted that the attenuation causes a power penalty relative to lossless waveguides, but more importantly, the attenuation means that an optimal sensing length exists, maximizing the sensitivities of the MZI; the optimal sensing length was found to be equal to the propagation length $L_e$ of the mode. The existence of an optimal sensing length is in sharp contrast to the lossless case, where increasing the sensing length indefinitely leads to a continuous increase in MZI sensitivity. Indeed, an attenuating MZI having an infinite sensing length must necessarily have zero sensitivity since there is no power output. However, as the waveguide attenuation vanishes, the propagation length, optimal sensing length and MZI sensitivity increase, and the lossless performance is eventually recovered.

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Inspection of the MZI sensitivities for bulk and surface sensing revealed that they were proportional to two parameters, defined as $H$ and $G$, respectively, each incorporating the operating wavelength, the waveguide sensitivity and its attenuation. More particularly, $H$ is given by the ratio of the effective index bulk sensitivity to its normalized attenuation $\left(\frac{\partial n_{\text{eff}}}{\partial n_c}/k_{\text{eff}}\right)$, and $G$ is given by the ratio of the effective index surface sensitivity to its normalized attenuation $\left(\frac{\partial n_{\text{eff}}}{\partial a}/k_{\text{eff}}\right)$. Maximizing $H$ or $G$ maximizes the corresponding MZI sensitivity, leading to preferred waveguide designs and preferred operating wavelengths.

It was also pointed out that the bulk and surface sensing detection limits ($\Delta n_{c,\text{min}}$ and $\Delta \Gamma_{\text{min}}$) were inversely proportional to the MZI sensitivity and thus to $G$ or $H$.

The analysis of the generic attenuating MZI is independent of the waveguide used for its implementation, so the results and conclusions are relevant to all waveguides (SPP or dielectric). Other MZI variants, for example incorporating an independent reference signal, or having two or three outputs, or incorporating phase modulation, would need to be investigated afresh in terms of its measurable(s). Finally, noise, which was ignored, may alter the conclusions.

The potential of SPP waveguides for bulk and surface (bio)chemical sensing was then assessed, anticipating use in a single-output MZI sensor. Several 1D SPP waveguides were modelled with the understanding that a 2D variant retaining the essential features of the 1D structure (similar perpendicular mode field distribution, propagation constant, sensitivities, ...) would be used to implement the sensor. Au and H$_2$O were assumed for the metal and dielectric regions, and a thin adlayer having an index representative of biological matter was used to determine the surface sensitivities. The SPP of the single interface with one adlayer along the Au surface, and the symmetric mode ($s_b$) of the symmetric MIM and IMI each with two adlayers (one along each Au surface), were modelled. The IMI in this context ends up being a free-standing Au film in H$_2$O, which does not seem too practical, so two variants were added to the set: one replaces the bottom H$_2$O region with a hypothetical index-matched substrate and the other introduces a thin Si$_3$N$_4$ free-standing membrane below the Au film as support. In both variants, only one adlayer is present along the top Au surface.

Approximate analytic expressions were derived and summarized for the single interface, from which trends and links were deduced. For instance, it was found that the $G$ and $H$ parameters in this case were proportional to recently introduced waveguide figures of merit $M_2$ and $M_3$, respectively.

The propagation constant, sensitivities and $G$ and $H$ parameters were then computed for all of the waveguides over their dimensional range and over the operating wavelength range of $600 \leq \lambda_0 \leq 1600$ nm. It was found that the bulk and surface sensitivities in the thin MIM can be $3 \times$ and $100 \times$ larger than those in the single interface, respectively, whereas the bulk and surface sensitivities in the thin IMI can be slightly smaller and $5 \times$ smaller, respectively, than in the single interface. On the other hand, $G$ in the thin IMI can be $10 \times$ larger than in the single interface, whereas $G$ in the MIM is about $3 \times$ larger. $H$ in the thin IMI can be $10 \times$ larger than in the single interface, whereas $H$ in the thin MIM is about $10 \times$ smaller.

An MZI implemented with the single interface should have detection limits for bulk and surface sensing that are comparable to conventional SPR, and an IMI-based MZI could improve on these by at least an order of magnitude. The MIM-based MZI also offers an improvement over the single interface, but for surface sensing only. Indeed, it is interesting that the IMI and the MIM both offer an improvement for surface sensing, despite the fact that they are at opposite ends of the confinement and dimensional scales: an IMI-based MZI would be millimetres long, and an MIM-based MZI would be microns long.

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Operating wavelengths that maximize $G$ or $H$ have also been identified: $G$ is largest near the short wavelength side of the Drude region ($\sim 850$ nm) in all structures. $H$ is also largest near this wavelength for the MIM and IMI, but is largest deeper into the infrared for the single interface.

It is hoped that the numerical results were collected and presented in a sufficiently clear form to be useful to other researchers investigating other integrated optic (or altogether different) sensing structures with these SPP waveguides, or close variants thereof.

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