Modeling chiral sculptured thin films as platforms for surface–plasmonic–polaritonic optical sensing

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Abstract

Biomimetic nanoengineered metamaterials called chiral sculptured thin films (CSTFs) are attractive platforms for optical sensing because their porosity, morphology and optical properties can be tailored to order. Furthermore, their ability to support more than one surface-plasmon-polariton (SPP) wave at a planar interface with a metal offers functionality beyond that associated with conventional SPP–based sensors. An empirical model was constructed to describe SPP–wave propagation guided by the planar interface of a CSTF — infiltrated with a fluid which supposedly contains analytes to be detected — and a metal. The inverse Bruggeman homogenization formalism was first used to determine the nanoscale model parameters of the CSTF. These parameters then served as inputs to the forward Bruggeman homogenization formalism to determine the reference relative permittivity dyadic of the infiltrated CSTF. By solving the corresponding boundary-value problem for a modified Kretschmann configuration, the characteristics of the multiple SPP modes at the planar interface were investigated as functions of the refractive index of the fluid infiltrating the CSTF and the rise angle of the CSTF. The SPP sensitivities thereby revealed bode well for the implementation of fluid–infiltrated CSTFs as SPP–based optical sensors.

Keywords: Bruggeman homogenization formalism, surface plasmon polariton, chiral sculptured thin film

1 Introduction

Engineered biomimicry till date has two major components: whereas bioinspiration leads to the same outcome as a biological activity, biomimetics is the reproduction of a natural functionality by copying relevant attributes of a biological organism. Bioinspiration is exemplified by aeroplanes that emulate the flight of birds and insects, biomimetics by the hook-and-loop fasteners whose structure emulates that of the burrs of some plants.

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Another example of biomimetics also has roots in botany [1, 2, 3]. Cholesterol is found in biological cells containing a nucleus. Benzoic acid is the principal component of a resin obtained from the bark of certain species of trees. The discovery of distinct temperature-dependent color effects in cholesteryl benzoate ($C_{34}H_{50}O_2$), an ester of cholesterol and benzoic acid, was reported in 1888 by Friedrich Reinitzer, an Austrian botanist [4, 5]. Within 15 years, the terms liquid crystal and flowing crystal for cholesteryl benzoate and similar materials had become well established [2, 3] due to the relentless research efforts of Otto Lehmann, with terminological clarity subsequently established in 1922 by Georges Friedel [6].

Cholesteric liquid crystals (CLCs) comprise aciculate molecules dispersed on a stack of closely spaced parallel planes. The molecules lying on a specific plane have an average orientation. This orientation progressively changes from plane to plane, the ensemble of orientations describing a helix in the thickness direction. The helix can be either left handed or right handed, just like curling tendrils that facilitate mutation by creepers and vines [7]. Given the presence of cholesterol in plant cells, the helical structure with striking optical consequences has been found in leaves of certain species [8]. When circularly polarized light of free-space wavelength in a certain regime falls normally on a CLC, the reflectance is high if the handednesses of the CLC and the incident light are the same, but is low otherwise—a phenomenon that has found much use over the last three decades [9, 10]. Parenthetically but remarkably, a coarse version of the cholesteric structure had been deduced two decades prior to Reinitzer’s discovery by Ernst Reusch [11], with surprising optical effects revealed a century later [12, 13]. However, Reusch’s work was inspired not by a botanical specimen but a mineral (mica).

Much before CLCs had not yet reached their current technological prominence, in 1959 Young and Kowal [14] reported the fabrication of what they called a “helically evaporated film” or a “helically deposited film”. Furthermore, they stated that this film “would resemble a Sole filter of the fan type, were the retardation plates of the Sole [sic] filter to approach zero thickness (and infinite number) while maintaining the total thickness of the filter constant.” A year later, Dawson and Young [15] related the structure of the Sole filter of the fan type to that of “Reusch rotators”, with their own films to be obtained therefrom “by allowing the number of elements to approach infinity”. They had thus engineered a solid-state analog of CLCs.

Although hugely significant, the 1959 paper of Young and Kowal [14] became obscure, gathering just 5 citations until and including 1996, according to the Web of Science™, out of a total of 85 at the time of this writing. The underlying concept was resuscitated in the early 1990s, with the emergence of sculptured thin films (STFs) [16, 17], which led to renewed interest in the 1959 paper.

STFs exemplify nanoengineered biomimetic metamaterials. An STF is an assembly of parallel nanowires whose bent and twisted shapes are engineered via dynamic manipulation during a physical vapor deposition process involving the production and aggregation of 1–to–3-nm clusters of atoms [18]. As STFs can be multifunctional [17, 18, 19], it is appropriate to call them metamaterials [20, 21].

The nanoscale control over the morphology of STFs has meant that their optical properties can be controlled on subwavelength scales [18]. Among a bevy of optical applications reported, the most prominent invokes the circular Bragg phenomenon exhibited by a chiral STF (CSTF). Comprising helical nanowires, CSTFs have been designed and verified to function as circular polarization filters [22] of the wideband [23, 24], narrowband [25], and multiband [24] varieties. Spectral shifts of narrowband filters due to fluid infiltration have also been modeled [26] and demonstrated [27, 28] for optical sensing. Very significantly, the planar interface of a CSTF and a metal has been theoretically shown [29, 30] to support the propagation of multiple surface-plasmon-polariton (SPP) waves—all of the same frequency but different phase speed, attenuation rate, and field configuration—and preliminary experimental evidence [31] is promising.

From a quantum mechanical viewpoint, an SPP is a quasi-particle which travels along the interface of a metal and a dielectric material, arising from the interaction of photons in the dielectric material and electrons in the metal [32]. A train of SPPs constitutes a SPP wave in classical language. Since SPP waves are acutely sensitive to the morphological and constitutive properties of the materials on either side of the interface to which they are localized, they have been widely exploited in chemical and biological sensing applications [33]. Indeed, SPP–based biosensors are at the forefront of optical label-free and real-time detection of analytes.

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3The 1960 paper of Dawson and Young [15] fared worse in achieving a celebrity status: it will probably receive its 7th citation in the Web of Science when this paper is published.
related to medical diagnostics, environmental monitoring and food safety [34, 35, 36, 37].

Essentially, an SPP–based sensor detects changes in refractive index arising in the partnering dielectric material, as follows. Suppose that a light wave excites an SPP wave which propagates along the interface of a metal and a dielectric material. The evanescent field of the SPP wave probes the dielectric material. Small changes in the refractive index of the dielectric material — for example, brought about by analyte molecules binding with biorecognition elements immobilized in the vicinity of the interface — results in detectable changes to the light wave coupled to the SPP wave, such as changes in angle and wavelength of excitation, intensity and phase [33, 36].

A key design feature of SPP–based sensors is the porosity of the partnering dielectric material, which allows analyte molecules to access the vicinity of the metal/dielectric interface. Therefore, STFs, whose porosity can be engineered to a high degree by controlling the physical vapor deposition process used for their manufacture [18], are promising candidates as platforms for SPP–based sensors. Recently, we reported on the sensitivity of SPP waves excited at the interface of a metal and a certain type of STF known as a columnar thin film (CTF), to changes in the refractive index of a fluid which infiltrates the void regions of the CTF [38]. Our numerical studies revealed that SPP waves propagate at a lower phase speed and with a shorter propagation length, if the infiltrating fluid has a larger refractive index. Furthermore, the angle of incidence required to excite an SPP wave in a Kretschmann configuration [33, 36] increases as the refractive index of the fluid increases.

We consider in the following sections the potential for a metal/CSTF interface to act as a platform for a SPP–based sensor. A CSTF may be regarded as a periodically nonhomogeneous continuum, which displays orthorhombic symmetry locally but is structurally chiral from a global perspective [18, Chap. 9]. The periodic nonhomogeneity of CSTFs engenders an especially interesting property not shared by homogeneous materials [39]: a CSTF can support more than one mode of SPP-wave propagation at its interface with a metal [29, 30, 31], thereby opening up the possibility of simultaneous detection of more than one type of analyte molecule. We note that CSTFs also offer an alternative sensing route, based on spectral shifts of the circular Bragg phenomenon brought about by the infiltrating fluid containing analytes [26], that could conceivably be harnessed in parallel with SPP–based detection to further extend the efficacy of the optical biosensing device.

In the notation adopted, vectors and column vectors are in boldface with the later enclosed in square brackets. Dyadics and matrixes are double underlined with the later enclosed in square brackets; a superscript ‘\(T\)’ denotes the transpose. The Cartesian unit vectors are written as \(\mathbf{u}_x\), \(\mathbf{u}_y\) and \(\mathbf{u}_z\). The free-space wavenumber, the free-space wavelength, and the intrinsic impedance of free space are given by \(k_0 = \omega \sqrt{\varepsilon_0 \mu_0}\), \(\lambda_0 = \frac{2\pi}{k_0}\) and \(\eta_0 = \sqrt{\mu_0 / \varepsilon_0}\), respectively, with \(\mu_0\) and \(\varepsilon_0\) being the permeability and permittivity of free space. An \(\exp(-i\omega t)\) time-dependence is implicit, with \(\omega\) denoting the angular frequency and \(i = \sqrt{-1}\).

2 Theory

As a realistic setup for launching SPP waves along the planar interface of a metal film and a CSTF, we consider the modification [40] to the standard Kretschmann configuration [41] illustrated schematically in Fig. 1. The regions \(z \leq 0\) and \(z \geq L_\Sigma\) are assumed to be occupied by homogeneous, isotropic, nondissipative, dielectric materials with relative permittivity scalars \(\varepsilon_d\) and \(\varepsilon_f = n_\ell^2\), respectively. A CSTF — infiltrated by a fluid of refractive index \(n_\ell\) — occupies the laminar region \(L_m \leq z \leq L_\Sigma\), while the laminar region \(0 \leq z \leq L_m\) is occupied by a metal with relative permittivity \(\varepsilon_m\).

2.1 Constitutive and morphological parameters of CSTF

A CSTF comprises an array of parallel helical nanowires [18]. It may be grown on a planar substrate, lying parallel to the plane \(z = 0\) say, by the deposition of an evaporated bulk material. On rotating the substrate about the \(z\) axis at a uniform angular speed throughout the deposition process, helical nanowires grow along the \(z\) direction, with the rise angle of each nanowire, relative to the \(xy\) plane, being denoted by \(\chi\).
The deposited material is assumed to be an isotropic dielectric material of refractive index \( n_s \). Significantly, \( n_s \) can be different from the refractive index of the bulk material that was evaporated [42, 43, 44].

The helical shape of each nanowire of a CSTF can be viewed as a string of highly elongated ellipsoidal inclusions, wound end-to-end around the \( z \) axis [45, 46]. The shape dyadic

\[
\mathbf{u}_n \mathbf{u}_n + \gamma_r \mathbf{u}_r \mathbf{u}_r + \gamma_b \mathbf{u}_b \mathbf{u}_b, \tag{1}
\]

wherein the normal, tangential, and binormal basis vectors are given as

\[
\mathbf{u}_n = -u_x \sin \chi + u_z \cos \chi, \quad \mathbf{u}_r = u_x \cos \chi + u_z \sin \chi, \quad \mathbf{u}_b = -u_y,
\]

prescribes the surface of each ellipsoidal inclusion. An aciculate shape is imposed on the inclusions by selecting the shape parameters \( \gamma_b \gg 1 \) and \( \gamma_r \gg 1 \). Increasing \( \gamma_r \) beyond 10 does not give rise to significant effects for slender inclusions [46]. Accordingly, for the numerical results which follow in §3, \( \gamma_r = 15 \) was chosen.

The proportion of a CSTF’s total volume occupied by helical nanowires is \( f \in (0, 1) \); equivalently, the volume fraction of the CSTF not occupied by nanowires is \( 1 - f \).

The relative permittivity dyadic

\[
\varepsilon_{STF}^s = \varepsilon_s \left( \frac{\pi(z - L_m)}{\Omega} \right) \cdot S_y(\chi) \cdot \varepsilon_{STF}^{ref} \cdot S_y(\chi) \cdot \varepsilon_{STF}^r \left( \frac{\pi(z - L_m)}{\Omega} \right), \quad L_m \leq z \leq L_S, \tag{3}
\]

characterizes the CSTF at length scales much greater than the nanoscale. Herein the handedness parameter \( h = +1(-1) \) for a structurally right (left)-handed CSTF; the rotation dyadics

\[
S_y(\chi) = u_y u_y + (u_x u_x + u_z u_z) \cos \chi + (u_x u_z - u_z u_x) \sin \chi;
\]

\[
S_y(\sigma) = u_z u_z + \left( u_x u_x + u_y u_y \right) \cos \sigma + (u_y u_x - u_x u_y) \sin \sigma
\]

and \( 2\Omega \) is the structural period. We take \( \ell_{STF} = (L_S - L_m)/2\Omega \) to be a positive–valued integer; i.e., the CSTF contains a whole number of structural periods. The reference relative permittivity dyadic \( \varepsilon_{STF}^{ref} \) has the orthorhombic form

\[
\varepsilon_{STF}^{ref} = \epsilon_{av} \mathbf{u}_n \mathbf{u}_n + \epsilon_{bo} \mathbf{u}_r \mathbf{u}_r + \epsilon_{co} \mathbf{u}_b \mathbf{u}_b, \tag{5}
\]

where \( \nu = 1 \) for a CSTF in which the void regions are vacuous (i.e., an uninfiltrated CSTF) and \( \nu = 2 \) for a CSTF in which the void regions are filled with a fluid of refractive index \( n_t \).

In principle, the relative permittivity parameters \( \{\epsilon_{a1}, \epsilon_{b1}, \epsilon_{c1}\} \) of an uninfiltrated CSTF are measurable. However, in the absence of suitable measured data for CSTFs, recent numerical studies have used values of \( \{\epsilon_{a1}, \epsilon_{b1}, \epsilon_{c1}\} \) measured for related CTFs. The nanoscale model parameters \( \{n_s, f, \gamma_b\} \) — which are not readily determined by experimental means — can be determined from a knowledge of \( \{\epsilon_{a1}, \epsilon_{b1}, \epsilon_{c1}\} \) by applying the inverse Bruggeman homogenization formalism [47]. Once \( \{n_s, f, \gamma_b\} \) have been found, they can be combined with \( \{n_t, \gamma_r\} \) in order to determine the relative permittivity parameters \( \{\epsilon_{a2}, \epsilon_{b2}, \epsilon_{c2}\} \) for the infiltrated CSTF, by applying the Bruggeman homogenization formalism in its usual forward sense [26, 46].

### 2.2 Boundary-value problem

The essence of the sensing mechanism in the modified Kretschmann configuration is described by the following boundary-value problem [40]. An arbitrarily polarized plane wave is launched in the half-space \( z < 0 \) towards the metal layer. We suppose that its wavevector lies in the \( xz \) plane, making an angle \( \theta_{inc} \in [0, \pi/2] \) relative to the \( +z \) axis. This incident plane-wave gives rise to a reflected plane wave in the half-space \( z < 0 \) and a transmitted plane wave in the half-space \( z > L_S \). Thus, the total electric field phasor in the half-space \( z < 0 \) may be expressed as

\[
E(r) = \left[ a_s u_y + a_p p_+(\theta_{inc}) \right] \exp(ikr) \exp(ik_0 \sqrt{\epsilon_d} z \cos \theta_{inc})
\]

\[
+ \left[ r_s u_y + r_p p_-(\theta_{inc}) \right] \exp(ikr) \exp(-ik_0 \sqrt{\epsilon_d} z \cos \theta_{inc}), \quad z < 0, \tag{6}
\]
while that in the half-space $z > L_\Sigma$ may be expressed as

$$E(r) = [t_s \mathbf{u}_y + t_p \mathbf{p}_+ (\theta_{tr})] \exp (i \omega x) \exp [ik_0 n_\ell (z - L_\Sigma) \cos \theta_{tr}], \quad z > L_\Sigma,$$

wherein $\mathbf{p}_+ (\theta) = \mp \mathbf{u}_z \cos \theta + \mathbf{u}_z \sin \theta$, $\kappa = k_0 \sqrt{\epsilon \delta} \sin \theta_{inc}$, and the angle of transmission $\theta_{tr}$ satisfies the law of Ibn Sahl [18] as follows:

$$\sqrt{\epsilon \delta} \sin \theta_{inc} = n_\ell \sin \theta_{tr}.$$  

(8)

By solving the related boundary-value problem, the complex-valued reflection and transmission amplitudes, namely $r_s, r_p, t_s$ and $t_p$, are related to the corresponding amplitudes $a_s$ and $a_p$ of the $s$- and $p$-polarized components of the incident plane wave. This standard procedure yields the algebraic relation [29]

$$\begin{bmatrix} t_s \\ t_p \\ 0 \\ 0 \end{bmatrix} = \left[ \begin{array}{cccc} K(n_\ell^2, \theta_{tr})^{-1} \cdot [Q]^{tstf} \cdot \exp \left( i \begin{bmatrix} P_m \end{bmatrix} L_m \right) \cdot [K(\epsilon, \theta_{inc})] \end{array} \right] \begin{bmatrix} a_s \\ a_p \\ r_s \\ r_p \end{bmatrix},$$

(9)

wherein the $4 \times 4$ matrices

$$[K(\epsilon, \theta)] = \begin{bmatrix} 0 & -\cos \theta & 0 & \cos \theta \\ 1 & 0 & 1 & 0 \\ -\sqrt{\epsilon} \cos \theta / \eta_0 & 0 & (\sqrt{\epsilon} \cos \theta) / \eta_0 & 0 \\ 0 & -\sqrt{\epsilon} / \eta_0 & 0 & -\sqrt{\epsilon} / \eta_0 \end{bmatrix},$$

(10)

$$[P_m] = \begin{bmatrix} 0 & 0 & 0 & \omega \mu_0 - (\kappa^2 / \omega \epsilon_0 \mu_m) \\ 0 & 0 & -\omega \epsilon_0 \mu_m + (\kappa^2 / \omega \mu_0) & 0 \\ 0 & \omega \epsilon_0 \mu_m & 0 & 0 \\ \omega \mu_0 & 0 & 0 & 0 \end{bmatrix}.$$

(11)

The transfer matrix $[Q]$ of one structural period of the CSTF and its evaluation are comprehensively described elsewhere [18, 29]. From (9), we may write

$$\begin{bmatrix} r_s \\ r_p \end{bmatrix} = \begin{bmatrix} r_{ss} & r_{sp} \\ r_{ps} & r_{pp} \end{bmatrix} \begin{bmatrix} a_s \\ a_p \end{bmatrix}, \quad \begin{bmatrix} t_s \\ t_p \end{bmatrix} = \begin{bmatrix} t_{ss} & t_{sp} \\ t_{ps} & t_{pp} \end{bmatrix} \begin{bmatrix} a_s \\ a_p \end{bmatrix},$$

(12)

thereby introducing the reflection coefficients $r_{ss,sp,ps,pp}$ and transmission coefficients $t_{ss,sp,ps,pp}$. The square magnitude of a reflection coefficient delivers the corresponding reflectance; i.e.,

$$R_{\alpha \beta} = |r_{\alpha \beta}|^2, \quad \alpha \in \{s, p\}, \quad \beta \in \{s, p\}.$$  

(13)

The four transmittances are defined as follows:

$$T_{\alpha \beta} = \frac{n_\ell \Re [\cos \theta_{tr}]}{\sqrt{\epsilon \delta} \cos \theta_{inc}} |r_{\alpha \beta}|^2, \quad \alpha \in \{s, p\}, \quad \beta \in \{s, p\}.$$  

(14)

The absorbances for $p$- and $s$-polarization states of the incident plane wave, namely

$$A_p = 1 - (R_{pp} + R_{sp} + T_{pp} + T_{sp}),$$

$$A_s = 1 - (R_{ss} + R_{ps} + T_{ss} + T_{ps}),$$

(15)

play a key role in identifying SPP waves. A sharp high peak in the graph of absorbance versus $\theta_{inc}$, arising at $\theta_{inc} = \tilde{\theta}_{inc}^{(r)}$ say, is a signature of SPP excitation at the $z = L_m$ interface provided that

(i) $\tilde{\theta}_{inc}^{(r)}$ is insensitive to changes in the CSTF’s thickness and

(ii) all eigenvalues of the matrix $[\tilde{Q}]$, defined by [48, 49]

$$[\tilde{Q}] = \exp \left\{ i 2 \Omega [Q] \right\},$$

(16)

evaluated at $\tilde{\theta}_{inc}^{(r)}$ have non-zero imaginary parts.
3 Numerical results and discussion

The helical morphology of a CSTF is nanoengineered by obliquely directing a collimated vapor flux in high vacuum towards a planar substrate that rotates about a fixed axis at a constant rate [18]. The angle $\chi_v$ between the average direction of the vapor flux and the substrate plane determines the dielectric properties of the CSTF. In view of the absence of suitable experimental data for CSTFs, for our numerical studies we chose the relative permittivity parameters

$$\begin{align*}
\epsilon_{a1} &= \left[ 1.0443 + 2.7394 \left( \frac{2\chi_v}{\pi} \right) - 1.3697 \left( \frac{2\chi_v}{\pi} \right)^2 \right]^2 \\
\epsilon_{b1} &= \left[ 1.6765 + 1.5649 \left( \frac{2\chi_v}{\pi} \right) - 0.7825 \left( \frac{2\chi_v}{\pi} \right)^2 \right]^2 \\
\epsilon_{c1} &= \left[ 1.3586 + 2.1109 \left( \frac{2\chi_v}{\pi} \right) - 1.0554 \left( \frac{2\chi_v}{\pi} \right)^2 \right]^2
\end{align*}$$

(17)

which were determined by experimental measurements at a free-space wavelength of 633 nm on a CTF made from patina® titanium oxide [50, 51]. Values for the corresponding nanoscale model parameters $\{n_x, f, \gamma_b\}$, as computed using the inverse Bruggeman homogenization formalism [47], are provided in Table 1 for the vapor flux angles $\chi_v = 15^\circ$, $30^\circ$, and $60^\circ$. Furthermore, we set $h = +1$ and normalized the CSTF’s structural half-period $\Omega$ relative to $\Omega_0 = 197$ nm. The metal was taken to be aluminum with relative permittivity $\epsilon_d = -56 + 21i$ at $\lambda_0 = 633$ nm, and the thickness of the metal film $L_m = 15$ nm. The relative permittivity $\epsilon_m$ is 6.656 which is typical of zinc selenide.

The parameter values chosen are consistent with those chosen for an earlier study involving an uninfiltrated CSTF ($n_x = 1$) [29], thereby allowing direct comparisons to be made. Extrapolating from this earlier study, we expect there to be up to five modes of SPP-wave propagation at the planar metal/CSTF interface, depending upon the value of the ratio $\Omega_0/\Omega$. The manifestation of these SPP modes in graphs of absorbance versus angle of incidence was tracked from the earlier study [29] to the present study by continuously varying the refractive index $n_x$. Further confirmation of the SPP status of the absorbance peaks was provided by checking that the corresponding eigenvalues of the transfer matrix $[\hat{Q}]$ have non-zero imaginary parts, per [48, 49].

We begin our presentation of numerical results with the graphs of absorbance for incident light of $s$- and $p$-polarization states versus $\theta_{inc}$ in Fig. 2. Here $n_x = 1.2$, $l_{stf} = 2$, $\Omega_0/\Omega = 1.5$ and $\chi_v = 20^\circ$. The $A_p$ peaks at $\theta_{inc} = 52.7^\circ$ and $33.3^\circ$ represent the SPP modes 1 and 2, respectively (in the terminology of Polo and Lakhtakia [29]).

The non-SPP $A_p$ peaks and the $A_s$ peaks which appear in Fig. 2 indicate waveguide modes [52] which must depend upon the thickness of the CSTF. To demonstrate this, in Fig. 3 the graphs of Fig. 2 are reproduced except that here $l_{stf} = 3$. We see that the $A_p$ peaks corresponding to the SPP modes 1 and 2 remain fixed in position but the other $A_p$ and $A_s$ peaks have moved relative to their positions in Fig. 2.

There are no $A_s$ peaks corresponding to the two SPP $A_p$ peaks in Figs. 2 and 3. However, by considerably increasing the thickness of the CSTF, evidence of a $A_s$ SPP emerges. Fig. 4 is as Figs. 2 and 3 except that here $l_{stf} = 20$ and we focus on the angle of incidence range $32.5^\circ < \theta_{inc} < 38.5^\circ$. We see that a $A_s$ peak has emerged at $\theta_{inc} = 33.8^\circ$; i.e., at the same $\theta_{inc}$ position as the mode 2 SPP $A_p$ peak. A corresponding $A_s$ peak was reported for the case of an uninfiltirated CSTF [29]. The large thickness needed is due to the slow decay rate of the electromagnetic fields in the thickness direction [30].

The SPP modes 3–5 only exist only for values of $\Omega_0/\Omega$ lower than that considered in Figs. 2–4. In Fig. 5 the $A_p$ peaks corresponding to SPP modes 3 and 4 can be seen at $\theta_{inc} = 46.1^\circ$ and $\theta_{inc} = 43.3^\circ$, respectively,
for $\Omega_0/\Omega = 0.2$ in the plot of absorbance versus angle of incidence. Also, the $A_p$ peak corresponding to SPP mode 5 can be seen at $\theta_{inc} = 44.3^\circ$ for $\Omega_0/\Omega = 0.13$ in the plot of absorbance versus $\theta_{inc}$ in Fig. 6. As in Figs. 2–4, the non–SPP peaks which appear in Figs. 5 and 6 represent waveguide modes, as has been confirmed by additional computations (not presented here) for different CSTF thicknesses.

The values of $\theta_{inc}$ at which the SPP peaks appear in the graphs of absorbance are sensitive to both the refractive index $n_\ell$ of the fluid infiltrating the CSTF and the vapor incidence angle $\chi_v$. The value of $\theta_{inc}$ at which the absorbance peak arises for the SPP mode $\tau$ — let us denote this value as $\tilde{\theta}^{(\tau)}_{inc}$ at a specific value of $n_\ell$ — is plotted versus $n_\ell \in (1,1.5)$ and $\chi_v \in (15^\circ, 60^\circ)$ for $\tau = 1$, and versus $n_\ell \in (1,1.5)$ and $\chi_v \in (15^\circ, 30^\circ)$ for $\tau = 2$, in Fig. 7. Here $l_{stf} = 2$ and $\Omega_0/\Omega = 0.6$. It may be observed that $\tilde{\theta}^{(1)}_{inc}$ uniformly increases as $n_\ell$ increases and as $\chi_v$ increases. A similar trend is exhibited by $\tilde{\theta}^{(2)}_{inc}$. The corresponding plots for $\tilde{\rho}^{(3)}_{inc}$, $\tilde{\rho}^{(4)}_{inc}$ and $\tilde{\rho}^{(5)}_{inc}$ (not provided here) also follow the same general trends as those displayed in Fig. 7. We note that for $\chi_v \lessgtr 65^\circ$ and $n_\ell \lessgtr 1.3$, the SPP mode 1 vanishes as $\tilde{\theta}^{(1)}_{inc}$ approaches $90^\circ$. Furthermore, as compared to the SPP mode 1, the SPP mode 2 exists only for a smaller $\chi_v$-range. Indeed, as compared to the SPP mode 2, the SPP modes 3–5 exist for even smaller $\chi_v$-ranges. The general trends displayed in Fig. 7 are the same as those reported for an analogous study based on an infiltrated CTF [38].

For optical-sensing applications, a feature of practical significance is the shape of the SPP peaks in graphs of absorbance versus angle of incidence. For the SPP mode 1 represented in Fig. 7, the sharpness of this $A_p$ peak, as gauged by the second derivative $d^2 \left( |A_p|^2 \right) / d\theta_{inc}^2$ evaluated at $\theta_{inc} = \tilde{\theta}^{(1)}_{inc}$, is plotted in Fig. 8 against $n_\ell \in (1,1.5)$ and $\chi_v \in (15^\circ, 60^\circ)$. The sharpness of the selected peak decreases as $n_\ell$ increases from 1.0 and as $\chi_v$ increases from 15$^\circ$. The most dramatic changes in sharpness occur at when both $n_\ell$ and $\chi_v$ have low values. The trends in Fig. 8 for the SPP mode 1 are qualitatively similar to those reported for an analogous study based on an infiltrated CTF [38], over the range $\chi_v \in (15^\circ, 60^\circ)$. Also provided in Fig. 8 is the corresponding plot of $\log \left[ d^2 \left( |A_p|^2 \right) / d\theta_{inc}^2 \right]$ evaluated at $\theta_{inc} = \tilde{\theta}^{(2)}_{inc}$. The $A_p$ peak for the SPP mode 2 is clearly much sharper than the peak for the SPP mode 1, across the entire range of $n_\ell$ values and $\chi_v$ values considered. Similarly, the $A_p$ peaks for the SPP modes 3–5 were found to be much sharper than the peak for the SPP mode 1.

Let us now further consider the sensitivity of the SPP peaks in the graphs of absorbance versus angle of incidence to both $n_\ell$ and $\chi_v$. As in Figs. 7 and 8, we fix $l_{stf} = 2$ and $\Omega_0/\Omega = 0.6$. The figure of merit (in degree/RIU$^4$)

$$\rho^{(\tau)} = \frac{\tilde{\theta}^{(\tau)}_{inc}(n_\ell) - \tilde{\theta}^{(\tau)}_{inc}(1,0)}{n_\ell - 1.0}, \quad \tau \in \{1, 2, 3, 4, 5\}, \quad (19)$$

wherein $\tilde{\theta}^{(\tau)}_{inc}$ is expressed as a function of $n_\ell$, is a measure of sensitivity for the SPP mode $\tau$. Graphs of $\rho^{(1)}$ versus $n_\ell \in (1.0, 1.5)$ are provided in Fig. 9 for $\chi_v \in \{15^\circ, 30^\circ, 60^\circ\}$. Also, $\rho^{(2)}$ is plotted versus $n_\ell \in (1.0, 1.5)$ for $\chi_v \in \{15^\circ, 21^\circ, 30^\circ\}$. Generally, $\tilde{\theta}^{(1)}_{inc}(n_\ell)$ is most sensitive to changes in $n_\ell$ when $\chi_v$ is small and $n_\ell$ is large, in keeping with an analogous study based on an infiltrated CTF [38]. A similar trend is followed by $\tilde{\theta}^{(2)}_{inc}(n_\ell)$ — and by $\tilde{\theta}^{(\tau)}_{inc}(n_\ell)$, $\tau \in \{3, 4, 5\}$, which are not represented in Fig. 9. Furthermore, the magnitudes of $\rho^{(\tau)}$ for all $\tau \in \{1, 2, 3, 4, 5\}$ are broadly similar. There are quantitative differences between the present CTF scenario for the SPP mode 1 and the analogous CTF scenario for the sole SPP mode: the sensitivities for $\chi_v = 15^\circ$ are greater for the latter scenario whereas the sensitivities for $\chi_v = 60^\circ$ are greater for the former scenario.

An alternative measure of sensitivity is provided by the refractive-index sensitivity [33]

$$\rho^{(\tau)}_{RI} = \frac{d \tilde{\theta}^{(\tau)}_{inc}(n_\ell)}{dn_\ell}, \quad \tau \in \{1, 2, 3, 4, 5\}, \quad (20)$$

also expressed in degree/RIU. While $\rho^{(\tau)}$ is the analog of the voltage-current ratio $V/I$ in electrical circuitry, $\rho^{(\tau)}_{RI}$ is the analog of the dynamic resistance $dV/dI$. In Fig. 10, $\rho^{(1)}_{RI}$ and $\rho^{(2)}_{RI}$ are plotted against $n_\ell$ with all

[33] Refractive-index unit
CSTF parameters being the same as those used for Fig. 9. Generally, $\tilde{\theta}_{\text{inc}}^{(\ell)}(n_\ell)$ is dynamically most sensitive to changes in $n_\ell$ when $n_\ell$ is large. This general trend with respect to $n_\ell$ for the SPP mode 1 is the opposite to that reported for an analogous study based on an infiltrated CTF [38]. The influence of $\chi_v$ on the dynamic sensitivity is less clear cut, as it depends upon the value of $n_\ell$ and which SPP mode is being considered. The magnitudes of $\rho_{RI}^{(1)}$ and $\rho_{RI}^{(2)}$—and $\rho_{RI}^{(3)}$, $\rho_{RI}^{(4)}$ and $\rho_{RI}^{(5)}$ not represented in Fig. 10—are broadly similar.

4 Closing remarks

Numerical simulations with an empirical model have revealed that the excitation of multiple SPP waves guided by the planar interface of a metal film and a CSTF is acutely sensitive to both the refractive index of a fluid infiltrating the CSTF and the morphology of the CSTF itself. Thus, the potential for a SPP–based CSTF optical sensor has been demonstrated. The SPP sensitivities reported here are, by and large, similar to those reported for an analogous study based on an infiltrated CTF [38]. However, in contrast to just one modality—angular shift of just one SPP mode—offered by CTFs, CSTFs offer multiple modalities. These are of two types: (i) angular shifts of more than SPP mode, and (ii) spectral shift of the circular Bragg phenomenon [26]. More than one of these modalities can be implemented simultaneously.

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Table 1: The dimensionless quantities $\gamma_b$, $f$, and $n_s$ computed using the inverse Bruggeman homogenization formalism for a titanium–oxide CTF with $\chi_v = 15^\circ$, 30$^\circ$ and 60$^\circ$.

| $\chi_v$ | $\gamma_b$ | $f$  | $n_s$ |
|----------|------------|------|-------|
| 15$^\circ$ | 2.2793     | 0.3614 | 3.2510 |
| 30$^\circ$ | 1.8381     | 0.5039 | 3.0517 |
| 60$^\circ$ | 1.4054     | 0.6956 | 2.9105 |
Figure 1: A plane wave incident at angle $\theta_{\text{inc}}$ on a metal–coated CSTF in the modified Kretschmann configuration, giving rise to a reflected plane wave and a transmitted plane wave.

Figure 2: Absorbances for $s$– (red, dashed curve) and $p$–incident (blue, solid curve) polarization plotted versus $\theta_{\text{inc}}$ (in degree) for the case where $n_t = 1.2$, $l_{\text{stf}} = 2$, $\Omega_0/\Omega = 1.5$ and $\chi_v = 20^\circ$. The peaks corresponding the SPP modes 1 and 2 are identified by $\star$ (in green).
Figure 3: As Fig. 2 except that $l_{stf} = 3$.

Figure 4: As Fig. 2 except that $l_{stf} = 20$ and only the SPP mode 2 is represented.

Figure 5: As Fig. 2 except that $n_\ell = 1.1$, $\Omega_0/\Omega = 0.2$ and the SPP modes 3 and 4 are represented.
Figure 6: As Fig. 2 except that $n_\ell = 1.1$, $\Omega_0/\Omega = 0.13$ and the SPP mode 5 is represented.

Figure 7: The angles $\tilde{\theta}_{\text{inc}}^{(1)}$ and $\tilde{\theta}_{\text{inc}}^{(2)}$ (in degree) plotted versus $n_\ell$ (in RIU) and $\chi_v$ (in degree). Here $l_{stf} = 2$ and $\Omega_0/\Omega = 0.6$. 
Figure 8: As Fig. 7 except that the quantities plotted are $d^2 (\frac{|A_p|^2}{d\theta_{inc}^2})$ at $\theta_{inc} = \tilde{\theta}_{inc}^{(1)}$ evaluated at $\theta_{inc} = \tilde{\theta}_{inc}^{(1)}$ and $\log \left[ d^2 (\frac{|A_p|^2}{d\theta_{inc}^2}) \right]$ evaluated at $\theta_{inc} = \tilde{\theta}_{inc}^{(2)}$.

Figure 9: The figures of merit $\rho^{(1)}$ and $\rho^{(2)}$ (in degree/RIU) plotted against $n_\ell$ for $\chi_v = 15^\circ$ (red, thick solid curves) and $30^\circ$ (green, dashed curves). The plots of $\rho^{(1)}$ for $\chi_v = 60^\circ$ and $\rho^{(2)}$ for $\chi_v = 21^\circ$ are also provided (blue, broken dashed curves).
Figure 10: As Fig. 9 except that the quantities plotted are the refractive-index sensitivities $\rho^{(1)}_{RI}$ and $\rho^{(2)}_{RI}$ (in degree/RIU).