Polarized spectral combs probe optical fiber surface plasmons

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Abstract: The high-order cladding modes of conventional single mode fiber come in semi-degenerate pairs corresponding to mostly radially or mostly azimuthally polarized light. Using tilted fiber Bragg gratings to excite these mode families separately, we show how plasmonic coupling to a thin gold coating on the surface of the fiber modifies the effective indices of the modes differently according to polarization and to mode order. In particular, we show the existence of a single “apolarized” grating resonance, with equal effective index for all input polarization states. This special resonance provides direct evidence of the excitation of a surface plasmon on the metal surface but also an absolute wavelength reference that allows for the precise localization of the most sensitive resonances in refractometric and biochemical sensing applications. Two plasmon interrogation methods are proposed, based on wavelength and amplitude measurements. Finally, we use a biotin-streptavidin biomolecular recognition experiment to demonstrate that differential spectral transmission measurements of a fine comb of cladding mode resonances in the vicinity of the apolarized resonance provide the most accurate method to extract information from plasmon-assisted Tilted fiber Bragg gratings, down to pM concentrations and at least 10⁻⁵ refractive index changes.

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1. Introduction

Surface Plasmon resonance (SPR) – the excitation by light of surface Plasmon waves propagating at the interface between a noble metal (most often gold or silver) and a dielectric (a glass prism for instance) – finds many applications in (bio)chemical sensing [1–3]. Thanks to their micrometric scale dimensions, optical fiber SPR sensors enable practical applications not possible with the bulky Kretschmann prism configuration, such as remote operation in very small volumes of the order of microliters. To excite SPR in optical fiber-based plasmonic sensors, light initially confined in the fiber core has to be locally outcoupled and brought into contact with the surrounding medium. Practically, this can be achieved either mechanically by polishing (or etching) the cladding so as to expose the evanescent wave to the surrounding medium or by using radiative fiber gratings (refractive index modulations imprinted in the fiber core thanks to its intrinsic photosensitivity) such as long period gratings (LPGs) or tilted fiber Bragg gratings (TFBGs). Both types of gratings couple the core mode to cladding modes that propagate towards the cladding-surrounding medium interface and are therefore sensitive to surrounding refractive index (SRI) changes [4–7]. In this work, we make use of TFBGs that maintain all the advantages of low loss optical fibers and allow for intrinsically temperature-insensitive measurements [8], a particularly important feature in high sensitivity SRI measurements. The purpose of the present paper is to demonstrate how the high finesse comb of resonances observed in the transmission spectra of TFBGs can be used to probe the effective index of surface plasmons most efficiently through differential amplitude measurements (instead of wavelength shifts).

A standard fiber Bragg grating (FBG) consists of a refractive index modulation in the core of an optical fiber that is imprinted perpendicular to the propagation axis. An FBG acts a selective mirror in wavelength, reflecting some wavelengths around a characteristic one, so-called Bragg wavelength. The latter is sensitive to temperature and mechanical strain but, as it results from light coupled in the fiber core, it is not influenced by SRI changes. A TFBG corresponds to a core refractive index modulation angled by a few degrees with respect to the perpendicular to the propagation axis. This induces two kinds of couplings: the self-backward coupling of the core mode at the Bragg wavelength and numerous couplings between the core mode and backward-going cladding modes at resonance wavelengths below the Bragg wavelength. Contrary to the Bragg resonance, the cladding mode resonances are modified by SRI changes [9–11], because both their effective index and mode field distribution depend on SRI. These two effects appear as grating resonance wavelength shifts and amplitude changes.

When a nanometric gold coating is deposited all around the fiber section, these effects are magnified by plasmonic enhancement for modes that have effective indices close to that of the SPR at the outer boundary of the gold coating [12] thus opening the path to sensitive biodetection [13]. In practice, SPR generation can only occur when the light propagating in the cladding is polarized radially to the fiber surface and a fortunate property of TFBGs is that they can excite spectrally distinct cladding modes with such polarization state. We have shown previously that when the input core mode is linearly polarized in the plane of the tilt direction (P-polarization), the electric field of the higher order cladding modes coupled by the TFBG is polarized mostly radially at the cladding interface and thus oriented in the correct direction to excite surface plasmon waves on the metal film [8]. On the other hand when the input core mode is linearly polarized out the plane of the tilt direction (S-polarization), the
The electric field of higher order cladding modes is polarized mostly azimuthally (i.e. tangentially to the metal) at the cladding interface and thus cannot couple energy to the surface plasmon waves. Therefore, when spectrally neighboring cladding mode resonances corresponding to orthogonal input linear polarization have effective indices close to a surface plasmon resonance, only one of the pair will be attenuated by transferring energy to the SPR.

SRI sensing over a large range is possible by tracking the wavelength shift of the most attenuated cladding mode resonance, which yields SRI sensitivity of ~500 nm/refractive index unit (RIU) in the range between 1.32 and 1.42. However, in most applications of SPR refractometry, the purpose is not absolute sensing over large SRI ranges but rather the detection of very small changes, due for instance to biomaterials attachment on a localized area of the gold surface. In this case, the most strongly coupled cladding mode resonances cannot be used for detection because they are too attenuated and broadened by the plasmon to reveal fine spectral and amplitude shifts: it was found experimentally that the amplitude evolution of a selected mode located slightly away from the SPR center can be exploited in practice [6,13]. It was also demonstrated recently that the differential behavior between two orthogonally polarized TFBG transmitted spectra can be used for demodulation purposes. We showed that narrowband resonant features (also located at wavelengths just below the SPR center) present a wavelength shift that enables refractive index measurements with a resolution better than $10^{-5}$ RIU, achieved through a quality factor of $10^5$ and signal-to-noise ratio greater than 50 dB [12,14]. This particular finding was supported by a recent theoretical analysis that showed how the presence of the gold layer shields azimuthally polarized cladding modes from the outside environment while allowing radially polarized ones to tunnel through and excite plasmons [15].

It is therefore well established that the transmission spectra of metal-coated TFBGs provide rich data sets from which highly sensitive refractometric (or biochemical binding) information can be retrieved. What is missing is theoretical foundation and data mining strategy to extract the most meaningful data for each particular sensing situation. In the work reported below, we show why and how spectrally adjacent but orthogonally polarized cladding mode resonances behave differently when the SRI varies. In particular, we identify an “apolarized” resonance, for which the effective refractive index remains constant whatever the input state of polarization. This apolarized resonance: 1) only appears in metal coated TFBGs, 2) is always located on the short wavelength side of the SPR maximum attenuation and 3) follows the SPR maximum location for “large” SRI changes corresponding to different sensing environments. Therefore, for each new biochemical sensing experiment, the straightforward determination of the apolarized resonance provides a narrowband spectral probe that is automatically located at the optimum spectral location for maximum sensitivity. Furthermore, since the apolarized resonance is located in the middle of the steep shoulder of the SPR envelope, its amplitude will change drastically even for SPR spectral shifts that would be hard to detect from the wavelength changes alone. We therefore suggest a two-step sensor interrogation scheme that consists of the identification of the apolarized reference and the subsequent measurement of its wavelength shift in response to biochemical binding experiments. Also, this peculiar resonance gives a reference to unambiguously locate the most effective resonance in terms of amplitude change for refractometric and biochemical sensing applications. Of course, the Bragg wavelength continues to serve its purpose as a global wavelength and power reference to factor out temperature, strain, or power source fluctuations from experiments.

Finally, a biomolecular recognition experiment is conducted with the biotin-streptavidin couple to confirm that monitoring selected cladding mode resonances in the vicinity of the apolarized one provides the most accurate method to extract information from SPR-TFBGs.
2. Experiments

Our experiments were carried out on 1 cm-long TFBGs manufactured into hydrogen-loaded standard single-mode optical fiber by means of a continuous-wave frequency-doubled Argon-ion laser emitting at 244 nm and a 1095 nm period uniform phase mask. The phase mask was tilted by 6° in the plane perpendicular to the incident UV beam. The latter was scanned along the phase mask at a velocity of 10 μm/s and with a mean power of 60 mW. Right after the inscription process, the gratings were annealed at 100 °C for 12 hours to stabilize their physical properties. A 30 nm gold coating was then deposited on the TFBGs using a standard sputtering process. The gold thickness was monitored in real time thanks to a Quartz microbalance placed in the sputtering chamber. A rotating clamp was installed in the sputtering chamber to ensure the uniformity of the gold layer around the fiber outer section.

TFBG transmitted amplitude spectra were recorded by an optical vector analyzer from LUNA Technologies, which was chosen for both its high measurement resolution (1.25 pm) and fast scanning rate (~1 sec to cover the full TFBG spectrum). A linear polarizer was placed upstream of the TFBG to control and orient the state of polarization (SOP) of the light launched into the plasmonic device. Care was taken to avoid polarization instabilities (short fiber lengths were used, strong curvatures were avoided, etc.).

Figure 1 displays the transmitted amplitude spectrum of an SPR-TFBG immersed in salted water (refractive index measured close to 1.356 at 589 nm), which was recorded with a linear input SOP optimized to maximize coupling to the SPR, corresponding to the P polarization mode, as further explained in the following. The Bragg wavelength appears at the right end side, centered at 1602 nm. In the following, the Bragg wavelength is used to remove any effect from surrounding temperature changes by monitoring its shift. This intrinsic feature is very interesting in practice as a change of 0.1 °C induces an SRI change of $10^{-5}$, thus comparable to the resolution of our sensing platform.

![Fig. 1. Amplitude transmitted spectrum of an SPR-TFBG immersed in salted water.](image)

Figure 2 displays the two orthogonally polarized spectra (so-called P and S polarization modes, referring to the linear polarization of the input beam relative to the tilt plane) that yield antagonist behaviors in liquids, as explained in [12]. The two spectra consist of quasi-periodic finely spaced combs of resonances that correspond to the hundreds of modes that can propagate in the fiber cladding.

The black curve corresponds to the P mode spectral comb and exhibits the typical SPR signature around 1541 nm, which is due to the maximum phase matching of the cladding mode to the surface plasmon mode of the gold water interface, according to [6]. It is obvious from this figure that S and P modes come in pairs and in order to facilitate the following discussion, the mode resonances are labeled as a function of their position with respect to the
most important one in the P spectrum (mode 0 in our numbering). Further clarification about the mode labeled 0 will be provided in the following.

Fig. 2. Transmitted amplitude spectra for two orthogonal SOPs (S and P polarization modes) of a TFBG immersed in salted water (SRI = 1.338).

2.1 Dependence of the cladding mode resonances on the input SOP

While the SPR mode is also the most sensitive to the SRI, it is nearly totally attenuated (in fact only revealed by its “absence” from the spectrum). As a consequence, other modes, just off the SPR peak, were exploited in previous studies. In [13], P mode transmitted spectra were measured and the equivalent of the mode labeled 2 in Fig. 2 was tracked for biosensing purposes. In [12], SRI measurements exploited the polarization dependent loss (PDL) spectrum (corresponding to the absolute value of the difference between the P and S modes). There, the + 1 mode was tracked, yielding SRI measurements accurate to $10^{-5}$ RIU.

Figures 3 and 4 present the evolutions of the amplitude and wavelength of selected peaks for input SOPs ranging between 0° and 180°, by steps of 2.4° when an SPR-TFBG is immersed in water. Figure 3 reveals that, for short wavelengths (mode $-2$), the behavior is similar to that of bare TFBGs in air, with the P mode wavelength longer than the S one. Then, getting closer to the SPR mode and for each cladding mode resonances pair, the P mode wavelength increases less than the S one. The crossing point occurs for the 0 mode, which we name “apolarized” resonance because it has the same wavelength for all the polarization states, even though it consists of the overlap of two different cladding modes. By the way, as the PDL for a TFBG corresponds to the absolute value of the difference between the S and P modes, the 0 mode is characterized by a strong attenuation in the PDL spectrum [12]. Past the 0 mode, the P resonances appear on the short wavelength side of the S ones. This peculiar behavior results from the fact that the P mode begins to localize in the gold sheath as it approaches the SPR, which lowers its effective refractive index (due to the small value of the gold refractive index). The S mode is tangentially polarized at the gold boundary and hardly penetrates it, therefore it does not feel this effective index decrease.

Beyond the crossing, the + 2 P mode now lags significantly behind the S mode, which points out to a strong localization of the P mode field in the gold layer and the strong influence of the SPR (and hence of the SRI) on the mode resonance. The next modes show an even stronger lag of the P mode but the associated resonance also becomes somewhat wider, because of the loss of energy to the metal. In the case of the + 4 mode, the wavelength
separation becomes so great that the S and P modes are completely dissociated. And further analysis of the modes in the vicinity of the SPR becomes meaningless.

Figure 3 shows similar graphs for modes further away on the red side of the SPR. As the cladding mode energy becomes more confined in the glass, the S and P modes are now strongly dissociated (nearly 0.5 nm of wavelength spacing between them) and the only difference is that the P modes are lossier as they penetrate a little more in the metal.

As detailed in the next section, the differential behavior of the cladding mode resonances as a function of the input SOP determines why they also behave distinctly in response to slight SRI changes.

2.2 Dependence of the cladding mode resonances on the SRI

Figure 5 presents the amplitude spectrum of an SPR-TFBG (P mode) immersed in salted water (refractive index measured close to 1.356 at 589 nm). By dilution, the refractive index value has been progressively modified over a range of 3.6 $10^{-4}$ RIU. Figure 5 also depicts the cladding mode resonance peak positions for all the investigated SRI values. As expected from
the characterization in polarization, cladding mode resonances behave differently in response to SRI changes.

Figure 5 displays a detailed view of the spectra around 4 selected cladding mode resonances (modes $-2$, $0$, $+2$ and $+20$, respectively). The differential behavior between cladding mode resonances now appears very clearly. 0 and $+2$ modes present the most continuous wavelength shift as a function of the SRI value, but the $+2$ mode, in addition to a larger wavelength shift, also presents a strong variation of its amplitude (more than 1 dB here). As illustrated by the $-2$ mode, cladding mode resonances below the 0 mode do not track SRI modifications with precision. The same conclusion stands for cladding mode resonances above the SPR that remain insensitive to slight SRI changes ($+20$ mode for instance), up to $\Delta \text{SRI} = 3.0 \times 10^{-4}$. Beyond that value, resonances away from the SPR center (and as close as the $-2$ resonance on the short side) begin to be affected because the SPR envelope shifts more significantly.

Based on these observations, Fig. 7 displays the wavelength shifts and amplitude variations of selected cladding mode resonances as a function of SRI changes up to $\Delta \text{SRI} = 3.0 \times 10^{-4}$. Here, relative evolutions are depicted, i.e. they are normalized with respect to the data collected for the first investigated SRI value. In terms of wavelength shift, modes around the SPR are the most sensitive and the 0 mode exhibits the most interesting behavior as it increases quasi-linearly. The changes remain quite small and somewhat irregular however, with a maximum sensitivity equal to 47 nm/RIU (the experiments reported in [6,12, 15] were obtained with slightly thicker gold films yielding higher sensitivities). The evolution of amplitudes shown in Fig. 7(b) is significantly more interesting and regular. The $+2$ mode is by far the most sensitive to use, with a total variation reaching $-1.1$ dB (a significant fraction of the initial peak-to-peak amplitude of the resonance, equal to 12 dB yielding a sensitivity equal to $-3600 \text{ dB/RIU}$). We also added the amplitude changes of the S mode adjacent to the $+2$ P mode to Fig. 7(b), thereby confirming that in addition to being significantly less sensitive in wavelength shift (as shown previously in [15]) S resonances are also insensitive in amplitude. Of course the exact numerical magnitude of these results vary depending on the exact experimental configuration (mostly on the initial SRI, gold thickness, grating length and tilt) but the main qualitative features observed here are inherent to the TFBG-SPR device: the
existence of the apolarized resonance and the identification of another pair of resonances with the largest maximum differential amplitude sensitivity between the S and P SOP that is located about mid-way between the apolarized resonance and the SPR center.

Fig. 6. Selected cladding mode resonance shifts for an SRI change of $3.6 \times 10^{-4}$ RIU.

Fig. 7. Experimental relative wavelength shift (left) and amplitude variation (right) of selected cladding mode resonances for an SRI change of $3.6 \times 10^{-4}$ RIU.

3. Numerically simulated evolutions

To corroborate the experimental trends, numerical simulations were conducted using a finite-difference complex mode solver in cylindrical coordinates (FIMMWAVE from Photon Design Inc.). Simulations yield the complex effective refractive indices and the mode fields of the SPR-TFBG used above. It is worth mentioning that the purpose of the simulations is to confirm the existence of the 0 mode and not to calculate a full TFBG spectrum. The latter is
by far more complex to obtain, as it requires the effective refractive indices and mode fields of at least 10 nearly degenerate cladding mode orders (5 for each polarization state) for each resonance, and the subsequent computation of the coupling coefficients and integration of multimodal coupled mode solutions. For each mode computed by the solver, we use the grating phase matching condition to estimate the corresponding resonance wavelength. This is obtained by considering 1.447 for the effective refractive index of the core mode and 552 nm for the grating period. Again, modes were labeled as a function of their position with respect to the apolarized mode pair. Figure 8 depicts the mode loss (imaginary part of the effective refractive index) of the modes as a function of the corresponding resonance wavelength (associated with the real part of the effective refractive index) for a standard single mode fiber covered by a 30 nm gold layer and immersed in different liquids (at these wavelengths the gold layer refractive index used by the solver is 0.56-11.4). Four configurations were simulated for liquids characterized by a refractive index at 1550 nm equal to 1.32, 1.34, 1.36 and 1.38, respectively. Taking into account the dispersion of the salted solution used in the experiments, the refractive index value of 1.34 at 1550 nm matches exactly the results of Fig. 5 to Fig. 7. The SPR mode can be readily identified as the one for which the mode loss is the highest. In complete agreement with experimental results, S modes (TE_{0n} and HE_{1n}) farthest from the SPR center appear on the short wavelength side of their P mode adjacent resonances (TM_{0n} and EH_{1n}). For mode 0, the wavelength positions of the TM_{0n} and HE_{1n} mode (1.32, 1.34 and 1.36 cases) or TE_{0n} and HE_{1n} (1.38 case) nearly coincide. These results also show that the exact occurrence of the mode 0 depends on the relative position of the spectral comb with respect to the SPR envelope and on the Vernier effect (the mode density increases at higher wavelengths). Proceeding forward along the wavelength scale, S modes then appear on the long wavelength side of the P modes. The cross over point, where the P mode shifts to the left side of the S mode is highlighted in the top section of each subplot, which shows the location of the S resonance relative to the P resonance of each pair, as a function of the S mode resonance wavelength.

Starting from 1.34, we then modified the surrounding refractive index value in steps of 3 \times 10^{-4} up to a range of 1.2 \times 10^{-3} RIU. Figure 9 depicts the relative wavelength shifts and relative mode loss variations for selected resonances. The results confirm the already established finding [15] that the SPR mode is the most sensitive with a wavelength shift computed equal to 194 nm/RIU. It is also clear that in absolute terms, the wavelength shift approach is inherently limited in ultimate accuracy: for SRI changes of 3.0 \times 10^{-4} in conditions such as those used experimentally here all the modes shift by less than 25 pm, except for the SPR mode whose exact wavelength position is impossible to measure experimentally. So it is likely that the demonstrated minimum detectable level of 10^{-5} reported in [12] (based on statistically determined wavelength noise levels of +/- 3 pm) is as good as possible for wavelength-based approaches. On the other hand, amplitude changes of the order of 1 dB are much easier to measure, especially at absolute power levels between -10 and -25 dBm with modern telecommunication-grade instrumentation (this is why the use of low loss standard single mode fiber at near infrared wavelengths presents such a great advantage for SPR instrumentation). In fact the instrument we use has a dynamic range of 80 dB and absolute insertion loss accuracy of 0.05 dB.
Fig. 8. Simulated mode loss as a function of the resonance wavelength for a standard single mode fiber covered by a 30 nm gold layer immersed in liquids of refractive index equal to 1.32 (a), 1.34 (b), 1.36 (c) and 1.38 (d). Modes corresponding to P polarization are black and blue, S polarization modes are green and red. The wavelength separation between S and P modes as a function of S wavelengths is displayed in the top of each subplot.

Fig. 9. Simulated relative wavelength shift (left) and relative mode loss variation (right) of selected cladding mode resonances for an SRI change of 1.2 $10^{-3}$ RIU.

We are not going to claim minimum detectable levels for the amplitude interrogation technique presented, because we have not yet made this characterization. Instead, and based
on the observations made above, we use a biotin-streptavidin recognition experiment to demonstrate the capability of the 0 mode (in wavelength) and the + 2 mode (in amplitude) to resolve biomolecular modifications at the sensor outer surface down to concentrations of 2pM.

4. Quantitative detection of macromolecular interactions

To obtain a self-assembled monolayer of biotin on the outer surface of the sensors, TFBGs were first thoroughly rinsed with ethanol. They were then immersed in a solution of biotinylated thiols (mercaptoundecyl-hexaethyleneoxy biotin amide) dispersed in ethanol. Thiol incubations were done during 12 hours at room temperature in a 1 mm thick capillary tube sealed at both ends to prevent solvent evaporation. At the end of the incubation, the functionalized gold-coated TFBGs were removed from the tube and again rinsed with ethanol, prior to their use for streptavidin quantification.

Biotinylated SPR-TFBGs were then immersed in different streptavidin solutions with concentrations ranging between $10^{-11}$ and $5 \times 10^{-4}$ g/ml. The sensor response caused by macromolecular interactions is evaluated relative to a reference measurement done in phosphate buffer saline (PBS). It is worth to mention that the refractive indices of solutions were measured with a refractometer (Reichert AR200) at 589 nm. Every solution is characterized by a refractive index that matches the one of PBS, with an uncertainty of $10^{-4}$ RIU corresponding to the resolution of the refractometer. This control ensures that the obtained data can be attributed to the binding of streptavidin molecules and not to undesired refractive index variations.

Figure 10 depicts the evolution of the wavelength and of the amplitude of two relevant cladding mode resonances relative to their value in the reference spectrum as a function of the streptavidin concentration. It reveals that the lowest streptavidin concentration that our sensor is able to detect is of the order of $10^{-10}$ g/ml, corresponding to 2 pM. The measured wavelengths are corrected with respect to the slight Bragg wavelength shift (2 pm maximum) attributed to temperature changes. The wavelength shift of the 0 mode reaches nearly 50 pm at $5 \times 10^{-4}$ g/ml. Amplitude variations of the + 2 mode reach 1.1 dB. For both evolutions, the saturation is obtained around $2 \times 10^{-4}$ g/ml. Therefore, we conclude that at this concentration all the functionalized surface is covered by streptavidin molecules.

5. Conclusion

In this paper, we have analyzed theoretically and experimentally the modifications of TFBG cladding mode resonances in gold coated optical fibers induced by changes in the surrounding refractive index. This analysis has provided a clear method to identify which of the
parameters of TFBG spectra measured with linearly polarized input light provide the most sensitivity to SRI changes, and by extension to bio-molecular binding events on the surface of the gold coated fibers.

In particular, we have shown that in a complete TFBG transmission spectrum there exists a single “apolarized” resonance whose wavelength does not change when the input state of polarization rotates. In addition to the confirmation that coupling to a surface plasmon on the outer layer of the gold coating is in fact occurring when this resonance is present, it provides a reference point that can be used to locate the most sensitive resonances for refractometric and biochemical measurements by either wavelength shift or amplitude shift methods. For wavelength interrogation, the apolarized resonance provides the best signal to noise ratio and sensitivity, but this technique remains limited to detectable levels near $10^{-5}$ in refractive index change because the Q-factor (wavelength/width) of the resonances we use is $10^5$ (for 1 cm long gratings). On the other hand, the amplitude of a resonance located mid-way between the apolarized resonance and the SPR center (i.e. on the steepest part of the shoulder of the SPR envelope) changes drastically. These measurements need to be carried out in P polarization, in order for the cladding mode fields to penetrate the gold layer and sense the outer medium, but if additional (simultaneous or not) measurements are carried out in S polarization they can be used to reference out any light power level fluctuations and environmental changes other than those occurring right at the fiber surface.

We thus demonstrated how two well defined quasi-periodic frequency combs (the S and P polarized TFBG transmission spectra) can be used to extract a maximum of information from plasmon-mediated interactions on metal coated optical fibers.

As a demonstration of these methods, we measured the binding of proteins on a fiber down to 2 pM concentrations. It is worth noting that in previous experiments involving biomolecular binding [13] the amplitude method had been chosen empirically (by trial and error on very large data sets) as the one providing the highest sensitivity. The work presented here provides a sound theoretical basis for this finding. Finally, it is clear from the results shown that when the changes to be measured are larger, the behavior of the resonances changes and the methods presented can no longer be applied because the envelope of the SPR interaction shifts too much. For such cases, envelope detection methods such as those provided in [14], also based on differential mode pairs, provide very accurate measurements as demonstrated by the high regression coefficients of the linear fits obtained.

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