High resolution fiber optic surface plasmon resonance sensors with single-sided gold coatings

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Abstract: The surface plasmon resonance (SPR) performance of gold coated tilted fiber Bragg gratings (TFBG) at near infrared wavelengths is evaluated as a function of the angle between the tilt plane orientation and the direction of single- and double-sided, nominally 50 nm-thick gold metal depositions. Scanning electron microscope images show that the coating are highly non-uniform around the fiber circumference, varying between near zero and 50 nm. In spite of these variations, the experimental results show that the spectral signature of the TFBG-SPR sensors is similar to that of simulations based on perfectly uniform coatings, provided that the depositions are suitably oriented along the tilt plane direction. Furthermore, it is shown that even a (properly oriented) single-sided coating (over only half of the fiber circumference) is sufficient to provide a theoretically perfect SPR response with a bandwidth under 5 nm, and 90% attenuation. Finally, using a pair of adjacent TFBG resonances within the SPR response envelope, a power detection scheme is used to demonstrate a limit of detection of $3 \times 10^{-6}$ refractive index units.

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1. Introduction
The use of surface plasmon resonance (SPR) effects enhances the sensitivity of fiber-optic sensors for surrounding refractive index (SRI) sensing, and by extension for chemical or biochemical sensing [1,2]. Amongst the wide variety of fiber-optic SPR sensors developed so far, most use metal coatings on geometry-modified fibers, similar in working principle to the prism-based Kretschmann-Raether configuration [3–9]. Although such fiber-optic SPR sensors can achieve extremely high SRI sensitivities and have been shown in recent experiments to achieve impressive detection results [10,11], the general use of working wavelengths the visible region, figures of merits (FOM) lower than 100 (due to the relatively broad spectral width of the resonance), and, in most cases, the lack of polarization control capability have limited their applications to relatively few laboratory demonstrations. As a path towards broader acceptance of this technology, near infrared fiber-optic SPR structures built using commercial telecommunication optical fibers and fiber gratings have been proposed and demonstrated [12,13]. Such devices benefit from several advantages: the SPR sensitivity increases with wavelength [2], fiber gratings provide narrowband spectral resonances that increase the FOM by two orders of magnitude [1,14], and, in the case of tilted fiber Bragg gratings (TFBGs), the capability to generate radially polarized light around the fiber circumference (i.e. TM-polarized across the metal coating) to allow for the efficient and selective excitation of surface plasmons [15]. In recent years, the comprehensive working principle, the interrogations techniques, and the optimization methods of SRI sensitivity, for
the TFBG-based SPR sensor have been investigated in our group [12–20]. As a result, a number of chemical [21] and biomedical [22–26] sensing applications were successfully demonstrated with the TFBG-based SPR platform, as well as probes for the optical properties of nanoscale metal films [27,28].

In all of the cases mentioned so far however, there remains a technological difficulty to overcome as it is generally considered difficult to deposit a metal film of the required thickness (near 50 nm for gold) and uniformity around the fiber circumference. The impact of thickness non-uniformities is to broaden and weaken the SPR response, with a corresponding negative impact on the figure of merit and limit of detection [29]. The most common methods to deposit the required coatings use physical vapor deposition techniques, such as evaporation (electron beam or thermal) and sputtering, both of which are directional (thicker in the region of the fiber facing the metal source). In order to achieve uniform coatings by these techniques, rotating mechanisms can be used to hold fibers in the deposition tool [5,26], or successive depositions carried out after manually rotating the fiber [22]. In the latter case, the gold coatings are still not absolutely uniform around the TFBG surface with thicker areas facing the vapor deposition directions.

In the results to be presented here, we demonstrate experimentally the surprising fact that in the case of TFBG-SPR sensors, a properly oriented, single-sided, single step evaporated gold coating with continuously varying thickness provides a SPR response with sensitivity, amplitude and spectral width that is as good as and in some ways better than devices with nominally uniform coatings, and demonstrate a measured limit of detection of $3.6 \times 10^{-6}$ RIU (Refractive Index Units) with a single-sided coated fiber.

2. Simulations

Fig. 1. Simulated TM-polarized transmission spectra of a 10° TFBG coated by 50 nm gold film in pure water. The electric field distributions of the four hybrid plasmonic vector cladding modes located within the SPR bandwidth are simulated with a four-layer optical fiber model (inset). The grating planes are tilted in the $y$-$z$ plane, resulting in horizontal refractive index fringes in the fiber core only.

The presence of TFBGs inside fiber cores causes a well-defined asymmetry along the direction of the tilt plane that impacts the mode shape and polarization of the cladding modes generated by the grating [15]. Figure 1 shows a simulated transmission spectrum of a 10° TFBG coated by a uniform gold film with a thickness of 50 nm immersed in water (SRI of 1.3155), under conditions where cladding modes are radially polarized, i.e. TM-polarized.
The fiber used was a standard SMF-28 (Corning) telecom fiber (core radius = 4.1 μm, cladding radius = 62.5 μm, cladding index = 1.4440, and core index = 1.4509) and the refractive index of the 50 nm thick gold coating was 0.58−i11. The gold thickness was chosen to be 50 nm based on the study of the thickness dependence of the TFBG-SPR response reported in Ref [14]. The simulation was carried out using standard coupled mode theory [30]. There is a sharp decrease in the amplitude of cladding mode resonances in the vicinity of ~1543 nm, for which the mode effective indices have a value near 1.3262 (calculated from the phase matching relation for this grating, Eq. (1) [1,13,14]). This confirms that those cladding modes have transferred energy to a lossy plasmon wave at the gold-water interface because the real part of the effective index of a surface plasmon wave at such boundary is equal to 1.3249 (from Eq. (2), reproduced from Ref [2]).

\[ \lambda_{cl} = (n_{eff}^{cl} + n_{eff}^{co}) \Lambda \]  

(1)

where \( \lambda_{cl} \) is the cladding mode resonance wavelength, \( n_{eff}^{cl} \) is the effective index of the cladding modes (to be found), and \( \Lambda = 556.4 \) nm is the grating period (projected along the fiber axis).

\[ n_{eff}^{sp} = \sqrt{\frac{\varepsilon_m \varepsilon_d}{\varepsilon_m + \varepsilon_d}} \]  

(2)

where \( n_{eff}^{sp} \) is the effective index of a surface plasmon at the interface between two media, \( \varepsilon_m \) is the relative permittivity of gold ((0.58−i11)^2), and \( \varepsilon_d \) is the relative permittivity of water (1.3155). The most attenuated cladding modes were identified from the simulation to be: the EH1,47, EH2,46, EH1,46, and EH2,45 modes (only the vector modes with azimuthal orders of 1 and 2 were considered due to their maximum coupling coefficients for 10° TFBGs [15]), and their electric field distributions in the fiber cross section are shown in the lower panel of Fig. 1. Now because the tilt of the grating is in the \( y-z \) plane, the locations of the maxima in the mode field patterns around the fiber circumference are locked relative to the tilt plane. In particular, it can be seen that EH1,47 and EH1,46 modes have their strongest radial fields at the top and bottom of the fiber cross sections, i.e. along the \( y \) axis, and near zero fields along the horizontal axis. This situation indicates that if a non-uniform metal coating is to be used, it should be preferentially oriented such that the most uniform sections are co-located with the maxima of the cladding mode radial fields, i.e. on the top and bottom of the fiber. In other words a two-step deposition should work best, provided the depositions are located in the correct direction relative to the tilt plane orientation.

3. Fabrication and characterization

The TFBGs used in this work were inscribed in hydrogen-loaded CORNING SMF-28 fibers with a pulsed KrF excimer laser using the phase-mask method [20]. The conditions of the hydrogenation process of the SMF-28 fibers include a pressure of 15.2 MPa, a temperature of 20 °C, and a duration of 14 days. The length and the tilt angle were chosen at 10 mm and 10° to excite a large number of high-order cladding modes with strong evanescent fields for SPR measurements. The Bragg wavelength is around 1610 nm, resulting from a phase mask period of 1112 nm. Before the gold depositions, the orientations of the grating planes of the fabricated TFBGs were determined from the radiation patterns of red light at 632.8 nm launched in the grating [29]. Figure 2 shows a typical radiation pattern of the red light propagating through a 10° TFBG, where the two brightest spots in the vertical direction indicate the orientation of the tilt grating plane, which is then marked on the fiber jacket in the vicinity of the grating.
Fig. 2. Radiation pattern of core guided red light from 10° TFBG (weak circular feature near the center of the figure). The vertical white arrows indicate the brightest areas along the radiation pattern, and consequently the tilt plane orientation (also shown in the side (x-z) and end view (x-y) models of the tilted grating).

Fig. 3. (a) SEM image of cross section of a gold-coated TFBG fabricated by the conventional two-step deposition method (electron-beam gold evaporation). Four zoom-in SEM images were taken from the four different cladding boundary areas with roughly orthogonal directions. (b) Relative position between the orientation of TFBG planes and the locations of the gold coating maxima and minima (aligned with the orthogonal dashed lines). The angle \( \theta \) indicates the relative angle between the grating fringe crossings in the core and the orientation of the deposition.

The metal deposition proceeds as follows: After being cleaned in a piranha solution (an 8:1:1 mixture of deionized (DI) water, Ammonium Hydroxide, and Hydrogen Peroxide), the marked TFBGs were mounted at specific angles to the tilt plane on a fiber holding fixture in
an electron-beam physical vapor deposition system (Balzers BA 510). Depositions were carried out at room temperature and under vacuum ($10^{-6}$ Torr). Several devices were prepared with different orientations of the deposited films relative to the tilt plane. In many cases, two gold deposition steps were conducted consecutively with the fiber holder being rotated by exactly 180° between the two deposition runs, with a mass-equivalent thickness of 50 nm for each run and a deposition rate of ~6 nm/min.

Scanning electron microscope (SEM) images of the complete cross section and zoomed views of four orthogonal cladding edges of a two-step, gold-coated TFBG are shown in Fig. 3. It is very obvious that two opposite edges have much thicker gold films while the orthogonal two directions have much thinner coatings, resulting in an overall approximate elliptical thickness profile with a major axis at an angle of $\theta$ to the grating tilt orientation (as shown in Fig. 3(b)). In the experiments, four kinds of TFBG samples coated by non-uniform gold films with elliptical thickness profiles at $\theta$ angles of 0°, 30°, 60°, and 90° relative to the tilt plane were fabricated. In addition, two kinds of single-sided-coated TFBG samples with $\theta$ equal to 0° and 90° were also fabricated for experimental comparisons.

4. Results and discussion

4.1 $\theta$-dependent TFBG-SPR responses

![Fig. 4. Experimental TM-polarized transmission spectra of TFBGs coated by non-uniform gold films with the different orientations respect to the direction of tilted grating planes. The red TFBG spectra shown in (a) and (d) indicate the single-sided gold-coated cases for the $\theta$ angles of 0° and 90°, respectively (spectra offset for clarity). The vertical grey bars indicate the corresponding positions of the SPR resonances. The blue arrows indicate the cut-off cladding mode. The Bragg wavelengths of the TFBGs are all around 1610.1 nm.](image)
Polarized spectra of gold-coated TFBGs with coating angles were measured with an experimental setup that includes a broadband source (BBS) (JDSU BBS1560), a polarization controller (PC) (JDSU PR2000), and an optical spectrum analyzer (OSA) (ANDO AQ6317B). Figure 4 shows the measured transmission spectra for the TM-polarized cladding modes of TFBG-SPR sensors in DI water at \( \theta \) angles of 0°, 30°, 60°, and 90°. Each deposition condition was reproduced twice with essentially identical results, as expected from the stability observed over years of experimentations with double sided coatings that were approximately oriented [13,15–19,23,24].

Two additional spectra are shown for single-sided gold coatings at 0° and 90° orientations. The SPR position (defined by the most attenuated cladding mode resonance) is indicated by grey shading in all spectra. The 30° and 60° cases correspond more or less to the original results of the polarized TFBG-SPR (and many other subsequent papers), where two-step depositions were used but randomly oriented relative to the tilt plane. The first notable feature from these spectra is that the SPR resonance wavelength blue-shifts with increasing \( \theta \) angle. Based on the simulation results of the TFBG-SPR wavelength dependence on thickness (in the range between 10 and 100 nm) for uniform gold coatings, the results shown here indicate that the overlap between the hybrid cladding mode profile and the thickest gold layer increases as \( \theta \) approaches 90° because an increase of thickness does cause a SPR blue shift (as well as a narrowing of the plasmonic attenuation region in simulations [14]. Indeed, for a perfectly oriented two-step coating the width of the spectral region where cladding modes become hybrid (and thus attenuated) is narrower for the experimental spectrum (~2 nm, in Fig. 4(d)) than in the simulation made with a uniform ideal coating of the same thickness (~5 nm, in Fig. 1). The result at 0° also indicate that a “perfect” error in coating orientation relative to the tilt plane results in very poor SPR performance (very broad featureless attenuation of cladding mode resonances) thus explaining the fairly large differences that were observed in the past between TFBG-SPR devices with randomly oriented two-step coatings.

Finally and quite surprisingly, the narrowest SPR attenuation envelopes were obtained with the single step one-sided coatings (\( \theta = 90° \) (single), Fig. 4(d)), while the orthogonal orientation results in a spectrum that is essentially identical to that of an uncoated grating in water (\( \theta = 0° \) (single), Fig. 4(a)), examples of which can be found in Ref [31]. While the narrower SPR bandwidth of the optimized coating contributes to a better figure of merit of the SPR device for sensing, an additional feature of this particular result deserves some attention. It turns out that only half the cladding mode profile interacts with a single sided coating, leaving the other half of the symmetric mode profile extending its evanescent field across a metal free fiber surface. Because of this, the optimized single step spectra also show the sharp decrease in cladding mode amplitudes observed at shorter wavelengths, indicative of the loss of guidance (mode cut-off point) [31]. This simultaneous presence of the SPR-active hybrid plasmonic modes and well identified cut-off modes provides two independent measurements of the same quantity (the SRI) in a single device. While spectral signatures for SPR and mode cut-off have been observed previously in TFBGs with uniform but too thin coatings, it was at the expense of the figure of merit because of the broadening of the SPR occurring in such cases and the fact that the cut-off modes still had to tunnel across some metal to reach the surrounding medium [26].

### 4.2 SRI sensitivity

Here, the SRI sensitivities of the two TFBGs with optimized double- and single-sided gold coatings at \( \theta = 90° \) are compared experimentally. Figure 5 shows the TM-polarized (SPR active) spectra of the two TFBG samples immersed in salt-water solutions with different mass concentrations of salt from 0% to 26% (the corresponding refractive indices range from 1.315 to 1.360 [31]). Recalling that dB scales are used in Fig. 4, there does appear to be a small difference between the amplitudes of the SPR modes of the single and double coating, with the latter being more attenuated (i.e. their amplitudes are smaller by a few dB), as expected.
By extracting the wavelength shifts of the most attenuated resonance in each spectrum (i.e. the SPR resonance), the measured SRI sensitivities are 589.8 nm/RIU and 566.8 nm/RIU for the double-sided coating and single sided coating cases, as shown in the insets of Fig. 5(a) and 5(b), respectively. It is quite remarkable that the single-sided gold-coated TFBG sample is just as good as the double-sided one, thus providing a much simpler fabrication process compared with double-sided ones (or uniform depositions using rotating fiber holders [26]). It is well-known that the high SRI sensitivity for the TFBG-SPR structure results from the large evanescent field of the hybrid plasmon wave propagating along the gold coating surface, which largely enhances the SRI-dependence of the cladding modes located within the SPR position [2]. In theory, the single-sided gold coating on TFBG surface can only excite the SPP wave on half cladding surface, which should decrease the SRI-dependence of the effective indices of those cladding modes.

Fig. 5. SPR spectra of TFBGs with double- (a) and single-sided (b) gold coatings for the case of $\theta = 90^\circ$ under different SRIs (spectra offset for clarity). The green arrows indicate the SPR wavelengths. The insets show the extracted SRI-induced SPR wavelength shifts and the corresponding SRI sensitivities.
Finally, the single-sided TFBG-SPR is used to test for the limit of detection, using a differential technique that is uniquely available in such devices because of the presence of the dense comb of resonances that sample the SPR spectral envelope of the gold film. It can be seen on Fig. 6 that there are two cladding resonances in the middle of the SPR spectral range that show clear opposite amplitude changes as the SPR resonance shifts with SRI. These differential amplitude shifts can be used to measure the SPR response, yielding a high sensitivity up to $3.34 \times 10^4$ dB/RIU. A separate measurement of the standard deviation of the resonance amplitude measurement, using resonances far from the SPR as reference, yielded a sensor resolution of 0.05 dB, resulting in a limit of detection of $3 \times 10^{-6}$ RIU.

4. Conclusion

We have demonstrated that the optimum excitation of hybrid plasmonic modes in the cladding of metal coated optical fibers using a TFBG is obtained when the orientation of the deposited metal coating on the fiber surface is aligned with the grating tilt plane. SPR signatures with extremely narrow line-width (less than 5 nm) and large attenuation depth (~10 dB relative to neighboring resonances) can be obtained when the orientation of TFBG tilt plane is parallel with that of the gold film. Most importantly, it was demonstrated that this kind of optimum response is obtained even for single sided coatings, thereby reducing the complexity of fiber SPR device fabrication considerably by removing the requirement for controlling the metal coating uniformity with nanometer precision around the fiber circumference. Finally, the single-sided TFBG-SPR device was used to demonstrate a new SPR interrogation method based on the differential amplitude changes of mode resonances located on either side of the SPR maximum. Thanks to the density of the spectral resonances and the sharpness of the SPR effect in these devices, this interrogation technique yielded a statistically significant limit of detection of $3 \times 10^{-6}$ RIU. The work present here provides a simple and reliable method to fabricate high sensitivity fiber-SPR sensors without constraints on the nanoscale metal coating uniformity.

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