Experimental determination of the sensitivity of Bloch Surface Waves based sensors

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Abstract: Detection of glucose in water solution for several different concentrations has been performed with the purpose to determine the sensitivity of Near Infrared Bloch Surface Waves (λ = 1.55μm) upon refractive index variations of the outer medium. TE-polarized electromagnetic surface waves are excited by a prism on a silicon nitride multilayer, according to the Kretschmann configuration. The real-time reflectance changes induced by discrete variations in glucose concentration has been revealed and analyzed. Without using any particular averaging strategy during the measurements, we pushed the device detection limit down to a glucose concentration of 2.5mg/dL, corresponding to a minimum detectable refractive index variation of the water solution as low as $3.8 \cdot 10^{-6}$.

OCIS codes: (240.6690) Surface waves; 280.4788 Optical sensing and sensors; (350.4238) Nanophotonics and photonic crystals.

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1. Introduction

Bloch Surface Waves (BSW) are electromagnetic modes propagating at the interface between a homogeneous medium and a truncated periodic structure like a dielectric monodimensional photonic crystal (1DPC) [1]. BSW dispersion curves are located within forbidden bands of the 1DPC, beyond the light line of the homogenous medium. This results in a exponential decay of the field envelope inside the periodic structure and an exponential decay of the field in the homogeneous medium because of total internal reflection. BSW on dielectric 1DPCs are equivalent to surface plasmon polaritons (SPP) on thin metal films and share some common characteristics. However, since dielectrics are characterized by much lower extinction than metals, BSW resonances appear much narrower than those observed for SPP [2].

The use of BSW is particularly promising in the optical sensing area [3], where the capability to confine and guide light in micro- and nano-structures can be exploited to enhance sensitivity by increasing the effective light-matter interaction. Beside the conventional Surface Plasmon Resonance (SPR)-like detection scheme [4,5], other improved detection configurations based on enhancement phenomena upon BSW coupling have been demonstrated [6,7].

In this work we report on the design, fabrication and characterisation of hydrogenated amorphous silicon nitride ||a-$Si_{1-x}N_x$:H| 1DPC for the optical detection of glucose mediated by BSW. As compared to other techniques, optical sensing is particularly attractive since it does not need ionizing radiation, generally it does not require consumable reagents, and the response is rather fast. Recent advances in optical glucose detection rely on transduction mechanisms based on Raman spectroscopy [8], fluorescence [9], diffraction [10], and resonance in integrated photonic devices [11]. Among these techniques, BSW-based detection represents a powerful alternative showing very high sensitivity.

2. Materials and Methods

a-$Si_{1-x}N_x$:H 1DPCs were grown by plasma-enhanced CVD on glass slides. Depending on the nitrogen content, a-$Si_{1-x}N_x$:H can have a tunable refractive index and optical gaps over a wide energy range [12]. These features, together with the nanometric control of the growth process, make such alloy very appealing for the realization of optical structures such as Bragg reflectors, microcavities [13] and multilayers sustaining BSW [14]. The composition of the a-$Si_{1-x}N_x$:H layers was controlled by operating on the ammonia fraction in a SiH_4 + NH_3

plasma, allowing the fabrication of high quality periodic stratified structures. More particularly, a- stratified structure made of $Si_{1-x}N_x$:H with variable average stoichiometry can benefit of a refractive index range from 1.7 (for a-Si_3N_4:H) up to 3.5 (for pure a-Si:H) in the low absorption energy region.. This characteristics allows the versatile use of silicon nitride alloys in the fabrication of 1DPC operating with BSW either in the near infrared or in the visible energy range [7]. In the former case, the highest refractive index contrasts can be obtained. In the latter, a suitable compromise between moderate high refractive index of sub-stoichiometric a-$Si_{1-x}N_x$:H with respect to a low layer absorbance can be reached. Moreover, the plasma assisted CVD process concerning with SiH_4 + NH_3 gas mixtures allows to obtain excellent interface abruptness, taking care that at each heterointerface, the rf discharge is interrupted and the deposition chamber evacuated, avoiding any intermixing effect. In the last decade, such a process allowed to obtain high quality nanometric multilayers exploited in light emitting devices [15,16].

The multilayer design is optimized for operating in contact with a water environment and allowing TE polarized BSW only. The structure consists of a 10-period stack of high (n_H = 2.19 at λ = 1530 nm) and low (n_L = 1.72 at λ = 1530 nm) refractive index layers having thicknesses d_H = 265 nm and d_L = 318 nm, respectively. A flow cell (volume 1.4 cm^3) is contacted to the top surface of the multilayer as shown in Fig. 1. The fluids used in the experiments are injected in the cell by inlet and drain hoses connected to syringes.

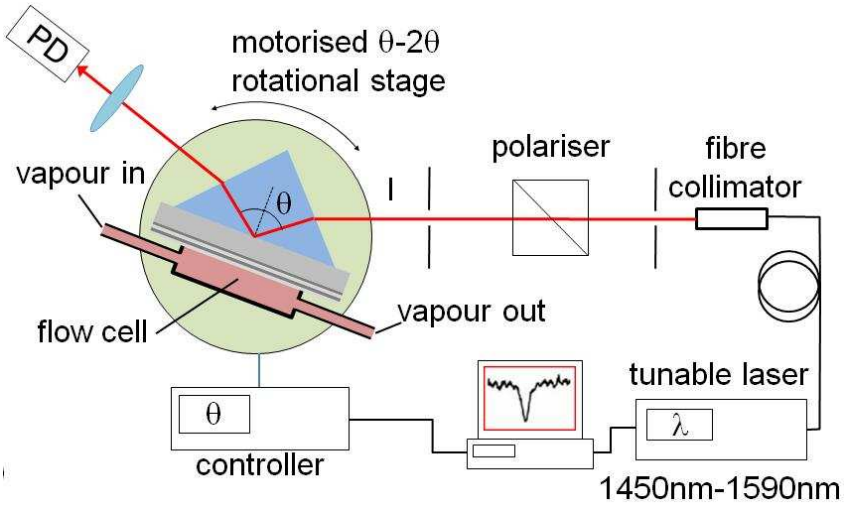

Fig. 1. Experimental setup for BSW coupling in the Kretschmann configuration. The illumination beam has low divergence (~0.038 deg.) and a linear polarization parallel to the 1DPC interfaces (TE). A flow cell is contacted to the top surface of the 1DPC.

A preliminary experimental map of the BSW dispersion curve in the wavelength range $\lambda \in$[1450 nm,1590 nm] was performed in the Kretschmann–Raether configuration, by means of the setup shown in Fig. 1, where a collimated and TE-polarized beam from a fibered, tunable laser diode, is focused on the sample through the input facet of a 45 deg BK7 glass coupling prism (n_{BK7} = 1.501). θ is the angle between the normal to the 1DPC planar interfaces and the direction of the incident beam at the prism/multilayer interface, ranging in the interval $\theta \in$[37 deg., 70 deg.]. A lens (focal length 50 mm) collects and focuses the reflected light onto a photodiode. The collection lens and the photodiode are rotated according to the angular position of the sample, in a θ–2θ fashion. Reflectance profiles, either angularly or spectrally resolved, are obtained by sweeping either θ or λ over the given ranges.

2. Results and Discussion

In Fig. 2a we show the measurements of the BSW resonances obtained for several increasing glucose concentrations in the water solution contained in the flow cell. A BSW is coupled at a fixed incidence angle θ_0 = 63.6deg starting from the condition of pure water. The glucose concentration is then increased starting from C = 10 mg/dL up to C = 10^4 mg/dL (0.01% to 10% vol.) in discrete steps. The reflectance values were best-fitted with Lorentzian curves, allowing the dip position and width to be determined. It is found that the spectral position of the BSW resonance is affected by the increase of the refractive index of the homogeneous medium, resulting in a positive shift $\Delta\lambda = \lambda - \lambda_0$ from the initial position of the dip at λ_0 = 1470 nm (pure water), as also observed in other BSW based sensing experiments [17]. We point out that the width of the resonances lays around 5nm, well below the standard width observed for SPP (tens of nm).

The $\Delta\lambda$ shift extracted from the measurements shown in Fig. 2a is plotted in Fig. 2b as a function of the corresponding glucose concentration. The variation of the refractive index of the solution $\Delta n(C) = n_C - n_0$, where n_C and n_0 are the refractive indices of the solution with glucose concentration C and of pure water respectively, was estimated by using the relation $\Delta n(C) = \alpha \cdot C$, where $\alpha = 1.515 \cdot 10^{-6}$dL/mg [18]. $\Delta n(C)$ is also reported on the top abscissa of Fig. 2b. From the linear fit of the experimental values, solid line in Fig. 2b (red on-line), we obtained an estimation of the sensitivity $\Delta\lambda/\Delta n = (1.105 \pm 0.040) \cdot 10^3$ nm/RIU. Such value is larger than the sensitivity of SPR sensors operating in the visible range and one order of magnitude smaller than their sensitivity in the near infrared (NIR-SPR) [19].

The sensitivity we measured is in good agreement with the expectations from the following approximated model. Given that the propagation constant of a monochromatic BSW at λ_0 excited at the interface 1DPC/solution in Kretschmann configuration can be written as $\beta = 2\pi/\lambda_0 \cdot n_{BK7}/n_C \cdot \sin\theta$, the overall variation $\Delta\beta$ at a fixed θ_0 is:

$$\Delta\beta = -2\pi \sin\theta_0 \cdot n_{BK7} \cdot \left(\frac{\Delta\lambda}{\lambda_o^2 n_0} + \frac{\Delta n}{\lambda_0 n_0^2} \right) \tag{1}$$

If we assume that variations $\Delta\beta$ produced by the perturbation $\Delta n(C)$ can be fully compensated by a corresponding $\Delta\lambda$, while keeping θ_0 fixed, after imposing $\Delta\beta = 0$, we obtain:

$$\frac{\Delta\lambda}{\lambda_0} = -\frac{\Delta n(C)}{n_0} \tag{2}$$

From this approximated linear model, we find a slope $|\Delta\lambda/\Delta n| = \lambda_0/n_0 = 1.1053 \cdot 10^3$ nm/RIU, which represents the detection sensitivity, in very good agreement with the linear best fit of the experimental data. Such result is also in agreement with rigorous calculations carried out by the transfer matrix method [4] and compared to the experimental measurements in Fig. 2b (dashed line, blue on-line). We observe that, for such calculations, the dependence $\Delta\lambda$ on Δn departs from linearity for larger λ. This is expected since the sensitivity is proportional to the first derivative of the BSW dispersion curve [20] that is not linear over a broader spectral range (see, e.g. Ref [20].).

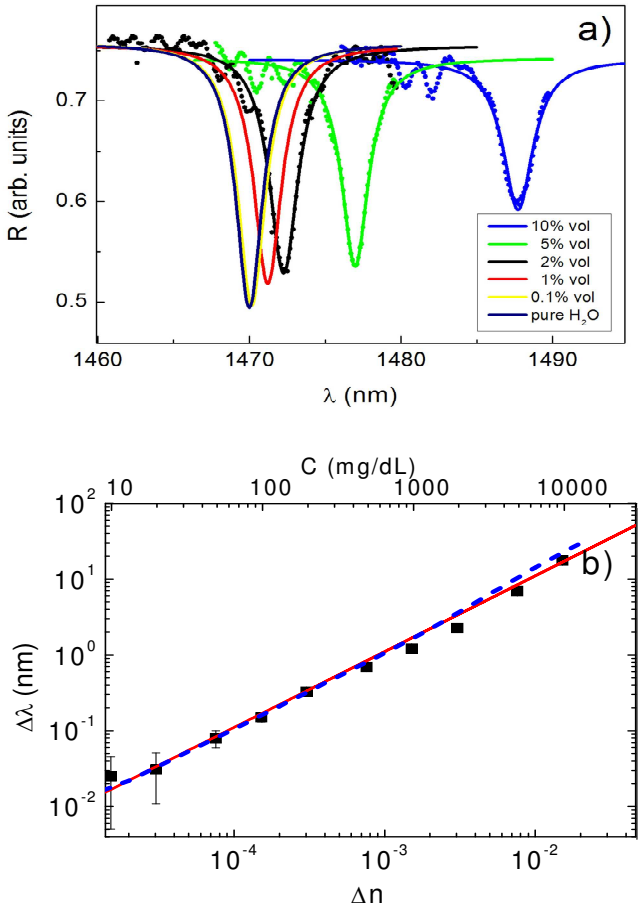

Fig. 2. (a) Best-fit of the spectrally resolved reflectance profiles R(λ) at fixed angle θ_0 showing the red-shift of the BSW resonance for increasing glucose concentrations. For the sake of clarity, the experimental points are shown only for the three largest concentrations. (b) Spectral shift Δλ of the BSW resonance as a function of the glucose concentration C and of the corresponding refractive index variation of the solution. The linear fit of the experimental data (solid, red on-line) and the calculated Δλ (dashed, blue on-line) are shown for comparison.

Since in a compact device it is not feasible and cost effective to integrate a finely tunable laser source providing a large enough spectral resolution for appreciating the small Δλ occurring at very low Δn, it would be desirable to operate at a single wavelength. A single wavelength detection scheme can be accomplished by measuring the real-time reflectance variation ΔR(t) at a fixed working point, at incidence angle θ_{WP} and wavelength λ_{WP}, as the glucose solution is being injected in the flow chamber. Such approach is usually named amplitude measurement and has been widely used, in particular in SPR imaging. Starting from the initial condition with the flow chamber filled with pure water, we chose values for θ_{WP} and λ_{WP} corresponding to the flex (side of increasing λ) of the BSW dip, where R(θ,λ) shows the steepest slope. Glucose solution is then injected at a specific concentration, and a time trace ΔR(t) is recorded. The flow cell is subsequently filled with pure water to clean it and to check for complete recovery of the signal. By analyzing each of the ΔR(t) profiles at specific glucose concentrations, a comprehensive plot ΔR(C) can be obtained, as presented in Fig. 3.

ΔR(C) shows a linear trend versus the glucose concentration, within the range of 5-500 mg/dL. For larger concentrations the shift of the BSW resonance gets larger than the resonance width, see Fig. 2a, and the device is driven out of linearity. The resolution, i.e. the lowest detectable glucose concentration, can be calculated by extrapolating the fit in Fig. 3 to the minimum measurable reflectivity change ($0.5°/_{00}$) evaluated by analysing the detection noise, whose behaviour is shown in the inset of Fig. 3 and that is mainly due to thermal fluctuations of n_C. We obtain C_{MIN} = 2.5 mg/dL (equivalent to a concentration of 0.0025% vol.) corresponding to a refractive index variation of the water solution as low as $3.8 \cdot 10^{-6}$.

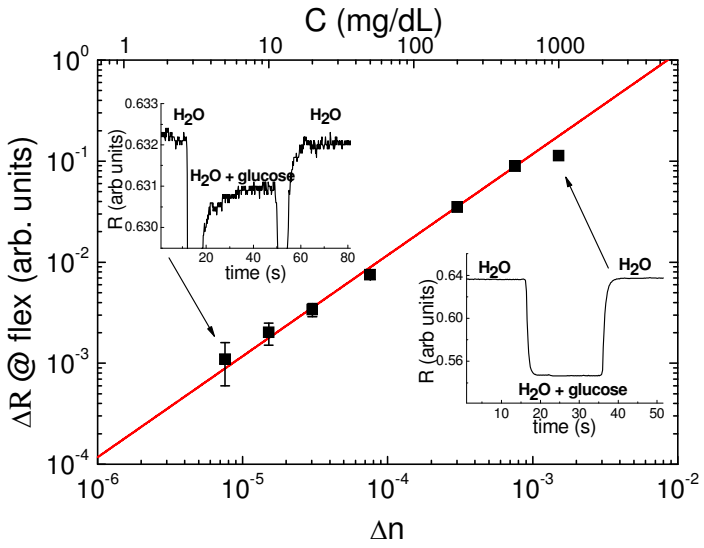

Fig. 3. Measured reflectance variations ΔR(t) as a function of the glucose concentration C in water solution. Insets: temporal ΔR(t) traces recorded during sequential injection cycles water/glucose solution/water for the two concentrations corresponding to the indicated experimental points. The continuous line shows the best linear fit of the experimental data.

This noticeable resolution is better than that obtained by NIR-SPR and amplitude measurements (10^{-5}) [21], despite the smaller sensitivity, owing to the remarkable narrowness of the BSW dips. If we consider the simplicity of the 1DPC and the detection technique, which does not make use of any averaging procedure, the measured Δn value is nicely small as compared to referenced results obtained either by SPR [19] or by means of other photonic approaches such as disk micro-resonators, providing a sensitivity to glucose corresponding to $\Delta n \geq 10^{-4}$ [11].

3. Conclusions

In conclusion, we presented an experimental work illustrating the potential of the use of a BSW-based detection technique applied to aqueous glucose solutions down to a concentration of $0.25 \cdot 10^{-4}$vol. corresponding to a minimum detectable refractive index variation as low as $3.8 \cdot 10^{-6}$. Our results are better than the best findings in the state-of-the-art for sensors working in the amplitude measurement configuration, thus providing an experimental evidence of the expected advantages of BSW over other optical detection techniques. It is worth to underline that BSWs are particularly sensitive to small changes of refractive index near the free surface of the multilayer structure. In the discussed case, bulk variations of the aqueous surrounding medium are analyzed in order to check the sensitivity of BSW-mediated sensing. In order to exploit BSW for multicomponent analysis, a careful surface functionalization aimed to the

specific biomolecule binding is required. Specific functionalization protocols, as those recently developed on silicon based surfaces for proteins, peptides and DNA strands [22,23], can be fruitfully used for providing chemically selective BSW based photonic biosensors.

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