Reflectance-based two-dimensional TiO₂ photonic crystal liquid sensors

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We propose and experimentally demonstrate a reflectance-based photonic crystal (PC) liquid sensor. The PC is made of two-dimensional TiO₂ nanopillar arrays. Such a reflectance-based structure with large functional area not only simplifies the optical guiding but also enhances the sensor signal. A linear shift of reflectance peaks is found for liquids with refractive indices varying from 1.333 to 1.390 at wavelength near 1.5 μm . Excellent agreement between measured values and the generated reflectance model at a fixed wavelength is obtained, indicating the high potential of these PC-based liquid sensors for biological and environmental applications. © 2012 Optical Society of America

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Photonic crystals (PC) can work as real-time, accurate, and highly sensitive biosensors and liquid sensors [1]. Previously reported PC devices, such as waveguides [2] and microcavities [3–5] were mainly operated using in-plane methods by measuring the transmittance. However, packaging complexity and high loss often limit their practical use. To overcome this limitation, we consider using the reflection mode instead of the often-used transmission mode.

To illustrate the sensing ability of PCs, the photonic band structures of two-dimensional (2D) triangular PC pillar arrays were first studied by the planewave expansion method [6]. Considering the presence and positions of band gaps and the corresponding geometrical features [7], TiO₂ (refractive index, RI or n , of 2.4) was chosen as the nanopillar material. An example with transverse-electric (TE) and transverse magnetic (TM) mode bands is shown in Fig. 1(a). The radius to lattice constant ratio (r/a) was 0.28 and the RI of the environmental medium was equal to 1.35. Two TM photonic band gaps were found at the dimensionless frequency [6] of around 0.35 (first band gap) and around 0.60 (second band gap), while no TE band gap was observed. Light with frequencies within such gaps cannot propagate through the PC structures, leading to low transmission or high reflection. To extensively investigate the effect of varying RI on the PC arrays, the dimensionless frequencies and corresponding wavelengths of mid-band-gaps are plotted versus RI from 1.30 to 1.50 in Fig. 1(b). The positive linear correlation suggests red shift as RI increases, which is interesting for sensing applications.

Figure 2(a) shows the schematic drawing of the proposed PC liquid sensor based on reflection detection. Liquids to be detected are guided into the chamber, which consists of two SiO₂ confining layers and a PC array in between. Polarized light with a certain wavelength range is projected through the transparent top window layer into the PC array at a specific incident angle. Once the light enters the chamber, two PC-related propagation behaviors occur [series A and B in Fig. 2(a)]. Light with frequencies outside the photonic band gaps can propagate transversely along the liquid-immersed PC array (series A), while light with frequencies within the

photonic band gaps cannot propagate through such PC array (series B). Thus high reflecting signals (reflectance peaks) can be observed in these frequency (or corresponding wavelength) regions. Since the positions of reflectance peaks are related to the RI changes, liquids with different RIs can be identified.

Furthermore, because the effective index of the liquid-immersed PC array (from 1.57 to 1.72 for liquid with RI between 1.30 and 1.50) is higher than the two SiO₂ confinement layers (RI: 1.45), higher energy localization tends to occur for the transversely propagating light. In addition, considering the high optical absorption of some liquids (for example, water-based solutions) in the infrared region, the reflection contrast between the two propagation behaviors is even more significant.

The main advantages of reflection-based sensing method compared to the in-plane transmittance approaches are: (1) It is much easier to couple/guide light into the liquid chambers as there is no need of the additional narrow waveguides. Furthermore, in the transmittance method, the inevitable junctions of the packaging seals and the waveguides often lead to signal losses. (2) As incident light is directly projected on the sensor structure, much larger sensing areas can be exploited with this

Fig. 1. (Color online) (a) Simulated photonic band structures of TiO₂ nanopillar arrays surrounded in an environmental medium with refractive index of 1.35 and (b) linear changes of the mid-band-gaps versus environmental refractive index.

Fig. 2. (Color online) (a) Schematic drawing of the reflectance-based PC liquid sensor; (b) SEM image of the uniform and smooth TiO_2 nanopillars array. Pillar height is 950 nm, pillar radius is 225 nm, and chamber volume is ~ 4 nL.

configuration; thus more signals can be collected with the same light beam density.

In order to study and verify the sensing ability of the proposed structure, devices with TiO_2 photonic crystal arrays were fabricated. A recently presented technique called “AARDE” based on atomic layer deposition of TiO_2 was used to form the accurately sized and highly ordered nanopillar arrays, without direct etching [8]. The fabricated 2D triangular photonic crystal structures have an area of 2 mm by 2 mm. The lattice constant was $0.8 \mu\text{m}$ and the radius of the TiO_2 pillars was 225 nm, resulting in an r/a of 0.28. The fabricated TiO_2 nanopillars were inspected with a Philips XL50 SEM [Fig. 2(b)]. It can be seen that the TiO_2 nanopillar array was very uniform and perfectly ordered.

Sugar solutions have a stable and linear relationship between RI and concentration for concentration under 40% [9]. Therefore, sugar solutions with concentration from 5% to 35% (RI from 1.3403 to 1.3902, taken from [9]) together with deionized water (RI: 1.333) and isopropanol (RI: 1.378) were employed to test our PC liquid sensor. The liquid droplets were dipped onto the device chamber with ultraclean nozzles. A 300 micrometer thick glass slide was then placed on top of the device as a cap window for light transmission and to prevent liquids from evaporating. Reflectance measurements were performed at incident angles between 45° to 75° with TM-polarized beams. In agreement with simulation, the samples aligned to the Γ -M direction presented the best density of states. After each measurement, the same device

Fig. 3. (Color online) Wide range reflection spectra of the PC liquid sensor for the 25% and 35% sugar solutions.

Fig. 4. (Color online) Normalized reflectance curves of different concentration sugar solutions tested with the PC liquid sensor. Inset: one example showing the Gaussian fit of the reflectance peak for determining the exact peak position.

and cap window were rinsed, dried and re-used for the next test.

Figure 3 shows the raw reflection spectra of two sugar concentrations (25% and 35%) from 450 nm to 1688 nm using a Woollam M2000 ellipsometer at 60° . Sharp peaks around 1500 nm as expected are observed. These peaks experienced red shift as the concentration (also for RI) increased. Moreover, except for the unique sharp peaks, the two spectra shared similar trends in the studied wavelength range. A total loss of 5.2 dB was measured.

High-resolution reflectance spectra (Fig. 4) around the peak region were obtained under the same measurement conditions using a PerkinElmer Lambda 950 UV/VIS spectrometer with a clamp as holder. The reflectance variation observed in Fig. 3 was significantly reduced by this mechanical clamping. Single and red shifting peaks were observed, which is in good agreement with the simulations and the data of the Woollam ellipsometer. Because of the large functional area of the reflection-based PC sensor, strong and smooth profiles of the peaks were observed, suggesting that the PC-induced reflection dominates in such region. This is a clear improvement compared to transmittance-based PC sensors, [3,4] where irregular shapes and less smooth peaks are usually found due to the weak signal. To characterize the peaks, Gaussian fittings were carried out using a baseline-included model:

$$y = a \exp[-(x - \lambda)^2 / (2w^2)] + b(x - \lambda) + y_0, \quad (1)$$

where λ represents the center of a peak, namely the peak position; a , b , w , and y_0 are constants to be determined. All of the peaks were well fitted with such model. The inset of Fig. 4 shows one of the fitted samples.

The peak positions of the measured sugar solutions together with the ones for water and isopropanol are plotted versus their refractive indices in Fig. 5. The relationship can be expressed by a linear fit:

$$\lambda = S \times n + \lambda_0, \quad (2)$$

where λ is the peak position, S is the wavelength sensitivity, and λ_0 is constant. A high correlation coefficient of

Fig. 5. (Color online) Measured peak positions of 5%–35% sugar solutions together with water and isopropanol versus their refractive indices. The linear fitting slope indicates a sensitivity of 441.6 nm/RIU.

0.99915 was obtained for the sugar solutions. Thus liquids with different RIs can be distinguished with the “wavelength-variant” method by monitoring the peak shift along the wavelength range. The wavelength sensitivity S was 441.6 nm per refractive index unit (RIU).

Besides the sensing methods using wavelength shifts, fixed-wavelength laser detections are also commonly used in real-time applications. With the above-verified linearity between the wavelength and RI, together with the complete Gaussian profile of the peaks, the reflectance of various RIs at a certain wavelength can be determined by substituting expression (2) for λ in Eq. (1):

$$f(n) = a \exp\{-[x_0 - (S \times n + \lambda_0)]^2 / (2w^2)\} + b[x_0 - (S \times n + \lambda_0)] + y_0, \quad (3)$$

where x_0 is the fixed wavelength of the laser source and n is the only variant. If the one-to-one correspondence between reflectance and refractive index exists, models set by Eq. (3) can provide another accurate approach for sensing.

To demonstrate, a model of reflectance versus RI from 1.325 to 1.395 was derived from the Gaussian profile for one of the concentrations studied (35% in this case, arbitrarily chosen). The fixed wavelength x_0 was set to 1495 nm. To compare with this calculation, the measured values of all of the concentrations at 1495 nm, which are indicated with the crosses on the dashed line in Fig. 4, were read out and re-plotted in Fig. 6. An excellent match between the measured values and the model curve was found, which verifies the accuracy of the model. The large reflectance discrimination among the measured points and the one-to-one correspondence in the concerned RI region (or concentrations) definitely allow an easy liquid identification. Thus, PC liquid sensors can be achieved by such “reflectance-variant” method at fixed wavelength. The fact that just one calibration is sufficient for generating the accurate model with a wide range of RIs makes the PC sensors very attractive for practical use.

Fig. 6. (Color online) Model curve derived from Gaussian profile for the 35% sugar solution is plotted together with the measured reflectance data at wavelength of 1495 nm.

In summary, we have proposed and demonstrated a reflectance-based PC liquid sensor using a 2D TiO₂ nanopillar array. Such reflectance-based structure simplifies the optical guiding, lowers the coupling losses, and enhances the sensor signal with its large functional area. Because of the high linearity of the wavelength peak shifting along the RI changes and the complete Gaussian profile of the reflectance peaks, both of the wavelength- and reflectance-variant approaches can provide very promising sensing properties. The flexible fabrication scheme for compact integration, the fast and precise real-time sensing capability, and the easy handling for practical use show great potential of these reflectance-based PC liquid sensors for multipurpose applications such as biosensing, hazard control and quality monitoring of beverages and fuel productions.

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