Refractive index sensing with an InGaAsP photonic crystal membrane cavity by means of photoluminescence

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We report the experimental investigation of several types of InGaAsP photonic crystal membrane cavities with embedded InAs quantum dots for use as refractive index sensors. The cavities are read out by photoluminescence, avoiding the need to couple them to waveguides. The sensitivities are determined from infiltration of the cavities with sugar-water solutions with known refractive index. The sensitivities increase markedly when the resonance frequency approaches the air band and correlate inversely with the quality factor. A maximum sensitivity of 425 nm per refractive index unit is found, a value among the highest reported so far. Preliminary surface functionalization for use as a biosensor is also reported.

Introduction

A photonic crystal (PhC) membrane cavity sensor is a system that detects the change in the cavity’s resonant wavelength when the ambient refractive index is changed. Recent publications in this field show that PhC membrane cavities promise to function as highly sensitive (bio)chemical sensors [1-5]. Because our quantum dot incorporated structures allow us to do photoluminescence experiments, we are able to fabricate and characterize a large number of cavities on the same chip. In this work, we report the systematic investigation of the sensitivity of five different cavity designs, by infiltrating them with water and different sugar-water solutions.

Fabrication of the cavities and experimental set-up

A 220 nm thick InGaAsP quaternary layer, containing one monolayer of self-assembled InAs quantum dots (density 300/µm²), is grown on an InP buffer layer by metal-organic chemical vapor deposition and capped by an additional InP layer. Hexagonal lattice photonic crystal patterns are defined in a 350 nm thick ZEP 520 resist layer by 30 keV electron beam lithography. Next, the pattern is transferred into an underlying 400 nm thick SiNₓ layer by reactive ion etching with CHF₃, after which it is transferred into the InP/InGaAsP/InP layer stack by inductively coupled plasma etching (Cl₂/Ar/H₂). The final step is a selective wet chemical etching step using a HCl:H₂O = 4:1 solution to undercut the InGaAsP layer and leave a free standing InGaAsP membrane.

Figure 1: Scanning electron microscope (SEM) images of the fabricated InGaAsP photonic crystal membrane cavities (a) modified H0, (b) modified H1, and (c) modified L3.
Figure 1 shows SEM images of the fabricated InGaAsP PhC membrane cavities referred to as (a) modified H0, (b) modified H1, and (c) modified L3 in which no hole, one hole and a line of three holes are left out of the pattern respectively. The cavity’s nearest neighbor holes are modified by reducing their radius and/or shifting them outwards in order to have different modes with high quality factors. We will refer to the designs as H0, H0s&r, H1s&r and L3s.

Room temperature photoluminescence (PL) experiments are conducted using a continuous wave diode laser ($\lambda = 660$ nm) at low power to avoid any heating complications. The laser spot is focused on one of the cavities by a 50x objective with a numerical aperture of 0.5 and the same objective is used to collect the light emitted by the cavity. The collected signal is dispersed in a 50 cm monochromator and detected by an InGaAs diode array cooled by liquid nitrogen.

**Sensitivity measurements**

Figure 2 shows the PL spectra obtained from a modified H1 cavity before and after water infiltration. After infiltrating the PhC with water, the resonant wavelengths are red-shifted due to the increase in the ambient refractive index $\Delta n = 0.3330$. The resonance peaks of the (theoretically) doubly degenerate quadrupole mode (Q1 and Q2) are shifted by approximately 55 nm and the peak of the hexapole mode by 70 nm. The red-shifts $\Delta \lambda$ are used to determine the sensitivity, given by $S = \Delta \lambda / \Delta n$. From the PL spectra of 80 cavities (all five designs included), we observe sensitivities between 100 and 425 nm/RIU. The data show that the modified H1s&r cavities are more sensitive than the other designs and Figure 2 shows that there is a strong mode dependence. The maximum sensitivities from our experiments are shown in Table 1. They are among the highest reported in literature so far [2,4], and compare favorably with cavities with central holes [3] or slots [5] made specifically to maximize sensitivity.

![Figure 2: PL spectra before and after water infiltration of a modified H1 cavity.](image)

The analysis of the sensitivities of 80 cavities shows two important trends that hold for all five designs but which we will illustrate for the H0s&r cavities in Figures 3(a) and (b). Figure 3(a) shows that the sensitivity scales as a function of the normalized frequency ($a / \lambda$), irrespective of the lithographic parameters. The sensitivity increases when the cavity modes are lithographically tuned from the dielectric band ($a / \lambda = 0.29$) to the air band ($a / \lambda = 0.37$) since the modes closer to the air band have more overlap of the electromagnetic field with the PhC holes [6]. Therefore, they are more sensitive to the changes in refractive index inside the holes.

<table>
<thead>
<tr>
<th>Cavity design</th>
<th>Sensitivity (nm/RIU)</th>
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<tbody>
<tr>
<td>H0 reduction only</td>
<td>390</td>
</tr>
<tr>
<td>H0 shift and reduction</td>
<td>390</td>
</tr>
<tr>
<td>H1 reduction only</td>
<td>310</td>
</tr>
<tr>
<td>H1 shift and reduction</td>
<td>425</td>
</tr>
<tr>
<td>L3 shift only</td>
<td>110</td>
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</tbody>
</table>
The detection limit is defined as the minimum change in refractive index that can be accurately detected. It is determined by the ratio of the resolution of the sensor and the sensitivity and is given by \[
\text{SNR}^{0.25} \cdot Q \cdot S
\] where \( \text{SNR} \) is the signal-to-noise ratio and \( Q \) the quality factor [7]. Therefore it is necessary to maximize \( \text{SNR} \), \( S \) and \( Q \) if we want to optimize (i.e. minimize) the detection limit. However, Figure 3(b) shows that the sensitivity tends to decrease as the quality factor increases. This inverse correlation is again the result from the variation in overlap of the electromagnetic field with the holes.

Figure 3: (a) The sensitivity as a function of the normalized frequency for one mode of the H0_{as} cavities for three different lattice constants, the shaded regions represent the dielectric and air band (b) The sensitivity as a function of the quality factor (measured in air) for the H0_{as} cavities.

To further investigate the sensitivity and the detection limit, a modified H1_{asr} cavity is infiltrated with sugar-water solutions with different concentrations of sugar with known refractive index. In Figure 4(a) the PL spectra measured in air and in the sugar-water solutions are shown. For this cavity the sensitivity is 240 nm/RIU but the quality factor is only \( \sim 1500 \) which leaves room for improving the detection limit. Figure 4(b) shows the dependence of the resonant wavelength on the refractive index. The sensitivity is not entirely linear but increases slightly with refractive index. The 3D FDTD simulations (CrystalWave) show that there is a discrepancy between our experiments and simulations, which is attributed to incomplete filling of the holes and the area underneath the membrane.

Figure 4: (a) The hexapole resonance peaks for an H1_{asr} cavity for measurements in air and in the sugar-water solutions (expressed in % weight/weight), (b) The experimental and simulated resonance wavelengths of the same H1_{asr} cavity for the different refractive indices.

**Functionalization of the membrane**

In order to test possible biosensing applications we functionalize the InGaAsP PhC membranes with proteins to carry out a model assay. We investigate the PL data of 18
Refractive index sensing with an InGaAsP photonic crystal membrane cavity by means of... cavities of five different designs. We choose for the protein bovine serum albumin functionalized with biotin molecules (bBSA, Pierce Biotechnology) to be our probe molecule. The bBSA is bound to the membrane surface during an one-hour incubation in a 0.14 mM citrate buffer (pH 6.8), after which the membrane is rinsed with Milli-Q water and blown dry with N2. We consistently observe a red-shift of the order of 0.5 nm, about a factor four smaller than calculated for a monolayer coverage of bBSA. In the second step the target molecule streptavidin (Pierce Biotechnology) is bound to the biotin molecules during an one-hour incubation in a 20 mM potassium phosphate buffer (pH 6.5), after which the membrane is rinsed and blown dry with N2. Surprisingly we do not observe an additional red-shift of the resonances, but a blue-shift that may be even below the original resonance wavelengths. This strongly suggests that the streptavidin solution not only removes the bBSA, but even some of the semiconductor material. Most probably, the blue-shift is caused by the potassium phosphate buffer etching the membrane which implies that the choice of the buffers used is of crucial importance for the design of a PhC biosensor.

Conclusions
In this work we investigated the sensitivity of five different types of cavities to changes in the ambient refractive index. The maximum sensitivity value observed was 425 nm/RIU for a modified H1s&e cavity. We have shown that the sensitivity depends on the cavity type and on the mode position inside the band gap; the modes closer to the air band are more sensitivity to changes in the refractive index. An inverse correlation between the sensitivity and the quality factor is observed, which is of importance for the detection limit. Additionally we have shown that functionalizing the InGaAsP membrane is possible but this and the detection of target molecules needs to be optimized.

References