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Integration of Sub-Wavelength Nanofluidics on Suspended Photonic Crystal Sensors

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ABSTRACT

In this paper, we introduce a novel sensor scheme which merges nano-photonics and nano-fluidics on a single platform through the use of free-standing photonic crystals (PhCs). PhCs offer great freedom to manipulate the spatial extent and the spectral characteristics of the electromagnetic fields. Also, nanoholes in PhCs provide a natural platform to transport solutions. By harnessing these nano-scale openings, we theoretically and experimentally demonstrate that both fluidics and light can be manipulated at sub-wavelength scales. In this scheme, the free standing PhCs are sealed in a chamber such that only the nano-scale hole arrays enable the flow between the top and the bottom channels. The nanohole arrays are used as sensing structures as well as nanofluidic channels. Compared to the conventional fluidic channels, we can actively steer the convective flow through the nanohole openings for effective delivery of the analytes to the sensor surface. This scheme also helps to overcome the surface tension of highly viscous solution and guarantees that the sensor can be totally immersed in solution. We apply this method to detect refractive index changes in aqueous solutions. Bulk measurements indicate that active delivery of the convective flow results in better performance. The sensitivity of the sensor reaches 510 nm/RIU for resonance located around 850 nm with a line-width of ~10 nm in solution. Experimental results are matched very well with numerical simulations. We also show that cross-polarization measurements can be employed to further improve the detection limit by increasing the signal-to-noise ratio.

Keywords: sub-wavelength, nano-photonics, nano-fluidics, photonic crystals, biological sensor.

INTRODUCTION

Within the last decade, several highly sensitive optical label-free nano-sensors have been introduced [1]. To have automated and portable lab-on-a-chip systems for bio-screening and bio-detection, researchers are integrating nano-resonator sensors with micro-fluidic circuits [2, 3]. These integrated systems offer significant advantages. They can enable label-free biodetection and can perform in real time. Also, they can be massively multiplexed for parallel screening of biological assays. Furthermore, the sensitivity can be increased all the way to the single particle detection limit. However, recent theoretical and numerical calculations indicate that we have to take into account the effects of various fluidic integration schemes as they can fundamentally limit the sensor performances [4]. For nano-sensors embedded in conventional microfluidic channels, the detection limit is often determined by the mass transport limitations [5]. Microfluidic channels support laminar flow profile, which is parallel to the surface and the flow rate becomes weaker close to the channel edge. In such case, most of the analytes flow through the channel without having a chance to get close enough to the ligands for capturing. For those who flow near the surface, once they are captured, so called depletion zones are formed near the sensor surface [6, 7]. These are the regions where the analytes transport diffusively. It could seriously limit the performance of the nanosensors, particularly when they are working at low concentration. Consequently, if no method is introduced to actively direct the convective flow towards the surface of the nano-micro size sensors, analytes at low concentrations may need week-to-years to diffuse due to mass transport limitations imposed by the depletion zones.

In this paper, we introduce a novel sensing scheme which merges nano-fluidics and nano-photonics on the same platform. It combines two individual micro-fluidic channels with a nanohole array based suspended PhC sensor. The nanohole array structures offer great freedom to manipulate the spatial extent and the spectral characteristics of the
electromagnetic fields. They are also natural platforms to transport the liquid at the nano-scales: a unique opportunity that has not been explored yet. The nanohole arrays are active both optically and fluidically. They are used for sensing by resonantly transmitting the light. As the same time, they are used as fluidic nanochannels to actively steer the convective flow to the surface. We present both theoretical and experimental analysis of the fluidics and the photonic components of our integrated system. As we show below, the proposed scheme offers enhancements in the sensor performance and the sensitivity.

**PRINCIPLE OF THE DEVICE**

Fig. 1a illustrates our proposed platform for active control of convective flow. The scheme is unique. It has two fluidic channels with multiple inlets/outlets. The two channels are connected by the free standing PhCs structure. This is contrary to the conventional approach in which the convective flow stream passes over the sensor (Fig. 1b). Fig. 1c and d show the steady state velocity distribution for the actively (proposed) and the passively (conventional) controlled convective flow schemes. Flow profiles around PhC regions are shown in details (insets). The simulations are done in two-dimensions using incompressible isothermal fluid flow, in which Navier-Stokes equations are solved numerically using finite element method in COMSOLTM.

For the passively controlled scheme (Fig. 1d), laminar flow profile is formed as the viscous forces in the fluid dominate over the inertial forces. The convective flow rate is fast close to the center of the channel but becomes very slow near the edges. This implies that in an immunoassay based sensing the depletion zones will extend further from the sensor surface causing ever slower analyte transport. One can increase the convective flow rate to shrink the depletion zones. However, such a passive (indirect) control only results in moderate improvements in mass transport rates [7]. The alternative approach we propose here is to steer the convective flow directly towards the sensing surface. As shown in Fig. 1c, the convective flow is still very strong around the sensing surface and the turbulences are generated around the holes. Such a directed flow can strongly improve the delivery of the analytes to the sensor surface. This scheme also helps to overcome the surface tension of highly viscous solution and guarantees that the sensor can be totally immersed in solution.

**DEVICE DESIGN AND FABRICATION**

In order to implement the proposed scheme, we use PhC structures on free standing membranes. PhCs offer unique opportunities to tailor the spatial extent of the electromagnetic field and control the strength of the light-matter...
interaction. In this work, we exploit guided resonances supported by square lattice silicon nitride (SiN) PhC slabs (inset in Fig. 3a). The periodic index contrast of the structures enables their excitation with a plane-wave illumination at normal incidence. This means, compared to other nanostructures, we do not need critical coupling schemes [8-10]. The ease of resonance excitation is particularly advantageous for sensor multiplexing applications.

Fig. 3. PhC sensor design: (a) Transmission spectra calculated by 3D-FDTD simulations are shown when the PhC slab is emerged in three different media: air (blue), water (red) and an IPA-chloroform mixture (green), respectively. Inset shows the schematic view of the design. Parameters for the structure are: \( r = 270 \text{ nm} \), \( a = 600 \text{ nm} \) and \( d=90 \text{nm} \). (b) Electromagnetic intensity distribution of the 1st mode when the structure is in air. Top and cross section views are shown, respectively.

Fig. 3a shows the transmission spectra of a specific design calculated by three dimensional finite-difference time-domain (3D-FDTD) method in three different media. For each case, two resonances are observed within the given spectral range. The intensity distribution of the 1\(^{st}\) order mode when the structure is in air is shown in Fig. 3b. Portion of the field extends into the holes, which is crucial in increasing the field overlap with the surrounding media for higher sensitivity. The spectral location of these resonances is highly sensitive to the refractive index changes occurring within the surroundings of PhC slabs. We evaluate its bulk sensitivity (in units of nm/RIU) by calculating the shift of the resonance position in wavelength versus the refractive index change in the surrounding environment.

Fig. 3. (a) Fabrication scheme. (b) Optical image of a membrane (c) SEM top view of the structure. (d) SEM cross-view of the structure tilted at 40°.

One important consideration here is the mechanical strength of the membranes as they need to stand the relatively high pressures generated by the perpendicular convective flow. Mechanically highly robust Low Pressure Chemical Vapor Deposition (LPCVD) silicon nitride (SiNx) films are excellent choice. Fig. 3a summarizes the fabrication steps.
We start with silicon wafer coated with LPCVD SiN films on both sides. Photolithography and RIE etching are used to open apertures on the back side. Then, we perform KOH etching, which selectively removes silicon. These steps result in suspended membrane as shown in Fig. 3b. On the membranes, PhCs are fabricated by performing e-beam lithography (EBL) and dry-etching. Finally oxygen plasma is used to clean the residues.

The optimized PhC structures are fabricated and scanning electron microscopy (SEM) images are shown in Fig. 3c and 3d. The diameter and the periodicity are 540 nm and 605 nm, respectively. The thickness of the membrane is measured to be 90nm. These numbers are quite close to the optimized design with \( r/a = 0.45 \) and \( d/a = 0.15 \). Although the membrane area (~40,000 \( \mu \text{m}^2 \)) is much larger compared to its thickness, the membranes are observed to survive long hours of operation under flow pressure.

**EXPERIMENTAL RESULT**

To carry out the flow tests, the structures are integrated in a chamber with two inlets/outlets both on the top and the bottom channels fabricated in polydimethylsiloxane (PDMS). To implement the laminar flow scheme (Fig. 1b), we blocked the inlet/outlet of the bottom channel. To steer the convective flow actively towards the sensing surface, we blocked one of the openings of both channels (Fig. 1a). Fig 4 illustrates active control of the flow. The solution is introduced from the bottom channel using the bottom inlet and steered perpendicular toward the surface. The video images are captured by an upright microscope, are shown in Fig. 4. At first the bottom channel is almost filled with the solution. Then the solution starts to go through the nanohole openings. The solution spreads over the surface. Finally, the structure is totally immersed in the solution. No damage or breakage of the membrane due to the applied pressure is observed.

![Fig. 4 Video images of the actively directed perpendicular convective flow:](image)

To experimentally evaluate the sensing response of the different flow schemes, transmission spectra of PhCs are obtained by launching a collimated and unpolarized light at normal incidence. The transmitted signal is collected with an objective lens and coupled into a spectrometer for spectral analysis. The comparison of the transmitted spectra is shown in Fig. 5a. Blue curve is the transmission spectrum taken in air, which clearly shows the excitation of the lowest and the next higher order modes at 667 nm and 610 nm, respectively. The red and the green curves are the responses in the solution (DI-water) for both flow schemes. When the convective flow is parallel to the surface (green curve), no leakage to the bottom surface is observed. On the other hand, when the convective flow is actively directed through the openings, PhC membrane is totally immersed in DI-water. This results in a larger refractive index change and more than 40 nm additional resonance shift. Fig. 5b and c shows the simulation results overlaid directly with the experimental measurements without any shifting. Near perfect match between the resonance locations and the line-widths are observed for both modes. The widths of the resonance peaks are significantly narrower when the structure is immersed in solution. This is due to the reduction of the index contrast within the slab resulting in less efficient coupling with the radiation continuum. With reduced index contrast (which could be due to immersion in solution or reduction of hole size [1](#)), guided resonances asymptotically turns into fully confined slab modes with infinite Q factors and narrow line-widths.
Bulk sensitivity of the PhCs is tested by successively applying five different solutions through the directed flow scheme: DI-water, acetone, IPA and two IPA-chloroform mixtures with refractive indices of 1, 1.33, 1.356, 1.377, 1.401 and 1.424, respectively. The measurements are performed by slowly pumping the solution to the chamber at 50 $\mu$L/s pumping rate. Prior to each measurement, we make sure the former solution is entirely replaced by the new one. As shown in Fig. 6a, with increasing refractive index the resonances red-shift and the line-widths become narrower. The linewidth of the resonance in DI-water is measured to be $\sim$10 nm. Fig. 6b shows the shift in resonance wavelength versus the refractive index of the liquid. The agreement between the experimental data and the theoretically predicted shifts is excellent. The experimentally measured sensitivity of the sensor achieves 510 nm/RIU for operation near 850 nm in wavelength.

As we know, low concentration of analytes would result in very small resonance peak shifts. In such cases, it is crucial to have narrow resonances with large signal-to-noise ratios. This can be achieved by using cross-polarization measurements [12]. The incident light is transmitted by PhC slabs through two different pathways [11]. One of them is the direct pathway, where a portion of the electromagnetic field goes straight through the slab. The other is the indirect pathway, where the remaining portion couples into the guided resonances before leaking into the radiation modes. These two pathways interfere with each other and result in resonances with sharp Fano-type asymmetric line-shapes. If we launch a polarized light and collect the signal after an analyzer oriented perpendicular to the polarizer, only the scattering from the guided resonances contributes. This results in dramatic suppression of the background and isolation of the resonances with large signal-to-noise ratios. In addition, the cross-polarization measurements result in purely...
Lorentzian-shape resonance profiles with narrower line-widths. Fig. 7a compares the cross-polarization spectrum (red) with the regular one (blue). The spectra are taken when the structure is in air. Three Lorentzian functions are used to fit the experimentally measured spectrum. The addition of them (red dashed curve in Fig. 7b) matches very well with the blue curve. For the 1st resonance (the one at the longer wavelengths), the cross-polarization measurements are fitted very well with two Lorentzians. There could be several reasons for this splitting, such as non-zero NA of the incoming light. However, in solution, the fundamental mode shows no splitting and has a much narrower linewidth.

Fig. 7. (a) Cross polarization spectrum (blue) and the regular unpolarized measurement (red). (b) Green curves correspond to the fitting of the resonance feature in the spectrum with single Lorentzian functions. Their summation is denoted in red dashed line and overlaid with the experimental result (blue curve).

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REFERENCE