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Analysis of a photonic crystal cavity based on absorbent layer for sensing applications

Jean Dahdah,* Nadège Courjal, and Fadi I. Baida

Institut FEMTO-ST, CNRS UMR 6174, Département d'Optique P.M. Duffieux, Université de Franche-Comté, 16 Route de Gray, 25030 Besançon Cedex, France

**Corresponding author: jdahdah@univ-fcomte.fr*

We present a photonic crystal (PhC) cavity based on a single hole defect filled with a sensitive absorbent layer for sensing applications. A preliminary study performed with the plane wave expansion method shows that the resonance peak of the cavity mode is 0.5 nm shifted for a 1 nm thick sensitive layer. A Lorentz dispersion model implemented in a two-dimensional-finite difference time domain homemade code shows that the absorption of the layer can be exploited for enhancing the sensitivity of the sensor. With the proposed geometry, we find that a variation in the refractive index of 10^{-7} leads to a variation in the transmittivity of 23% at the resonance peak. This study is proposed for the development of a compact benzene sensor on a MgO doped lithium niobate PhC.

1. INTRODUCTION

Photonic crystals (PhCs) have been studied in the past years as attractive platforms for sensing applications [1–9]. Introducing a point defect in the otherwise perfect PhCs allows the electric field to be strongly confined around the defect region for a specific frequency. This leads to narrow transmission peaks inside the photonic bandgap. The high confinement of the optical mode in the small modal volume leads to a high quality factor [10,11] resulting in a high sensitivity of the microcavities toward local changes in their environment (temperature, pressure, refractive index, etc.). In the last decade, many studies have demonstrated, both experimentally and theoretically, the attractive potential of microcavities for detecting the presence of specific molecules. These sensors rely on a microcavity coated with a functional layer that reacts specifically with a given analyte. The physico-chemical reaction induces a change in the refractive index [1,8] together with a variation in the layer's thickness, which causes a frequency shift of the resonance peak [2,5].

In this paper, we present a single hole defect (SHD) microcavity functionalized by a sensitive layer having an absorption peak at the resonance wavelength. The presence of the analyte is thus detected by a variation in the output intensity that is not only due to the variations in thickness and refractive index, but also to the modification of absorption. We are going to show that this configuration helps in increasing the sensitivity of the sensor.

The theoretical study of the microcavities coated by a thin sensitive layer can be performed by the finite difference time domain (FDTD) method, but the calculation is highly time consuming due to the small thickness of the layer. Here, we propose a preliminary approach based on plane wave expansion (PWE) associated with the super-cell method, which enables a significant gain of time cal-

culatation for establishing the optimal parameters of the cavity. A 2D-FDTD homemade code [12] is exploited only in a second time, in order to take into account the absorption of the sensitive layer and to evaluate its contribution to the sensor's sensitivity.

2. SHD CAVITY DESIGN

The proposed structure is schematically depicted in Fig. 1(a). It consists of a biperiodic triangular lattice of circular air holes, in which a defect is introduced by reducing the center pore radius. A third medium is introduced as a ring-shaped intermediate layer around all the holes' surfaces. This medium corresponds to a sensitive layer that can react selectively with a specific analyte. The reaction induces a change in the properties of the layer (refractive index, absorption, and thickness) that can be measured by a variation in the light intensity transmitted at the cavity mode resonance frequency. In view of enhancing the intensity variation, the parameters of the cavity are determined to make the resonance peak of the cavity coincide with the absorption peak of the sensitive layer.

Here we propose to study a MgO:LiNbO₃ substrate coated with a porphyrin layer. The choice of the lithium niobate substrate is motivated by the possibility of tuning the transmission response by an external effect. A dynamic control of the operating point can for instance be provided by an electro-optic effect. Consequently, the resonance peak of the cavity can be adjusted electrically to correspond to the maximum of absorption of the porphyrin layer, which occurs at $\lambda=419.5$ nm. The LiNbO₃ substrate is doped with MgO to avoid the optical damages due to the photorefractive effect at visible wavelengths [13]. The possibility of functionalizing lithium niobate substrates has recently been demonstrated in [14]: the same method can be applied for grafting porphyrin layers

Fig. 1. (a) SHD studied cavity, where dark gray corresponds to substrate ($\epsilon_b=5.319$), light gray indicates the sensitive layer ($\epsilon_c=2.775$), and white areas are air holes ($\epsilon_a=1$). (b) Zoom-in made around the defect region where r is the defect radius, d is the thickness of the layer, R is the air hole radius, and a is the period of the PhC.

on lithium niobate substrates. The porphyrin layer plays then the role of a probe for benzene molecules.

The dielectric permittivity of the MgO:LiNbO_3 substrate is $\epsilon_b=5.3190$ at $\lambda \approx 420$ nm. The dielectric permittivity of the porphyrin layer is $\epsilon_c=2.7225$, and its thickness is denoted as d as shown in Fig. 1(b). Its absorption properties are described in the third section.

This choice of benzene sensor was motivated by numerous reports that denounce its high concentrations in habitations and the consequent impacts on health. Indeed, benzene is myelo-toxic and it is a leukemia-inducing compound. So, a compact sensor can be very attractive for evaluating personal exposures to benzene pollutants. It should be emphasized that the presented configuration is not limited to one application. For instance, the porphyrin layer could be replaced by a metallophthalocyanine layer if volatile organic compound vapors were targeted. In this case, the operating wavelength would be 600 nm [15].

First, we have developed a PWE based code which takes into account the presence of a thin layer coating the holes. The first step was to determine the band structure of the 2D triangular lattice PhC. The band structure is determined by the Fourier coefficients of the reciprocal dielectric functions [16], which in our case are given by

$$\kappa(0) = f_1 \left(\frac{1}{\epsilon_a} - \frac{1}{\epsilon_c} \right) + f_2 \left(\frac{1}{\epsilon_c} - \frac{1}{\epsilon_b} \right) + \frac{1}{\epsilon_b}, \quad (1)$$

$$\begin{aligned} \kappa(G) = & 2f_1 \left(\frac{1}{\epsilon_a} - \frac{1}{\epsilon_c} \right) \left(\frac{J_1(GR)}{GR} \right) + 2f_2 \left(\frac{1}{\epsilon_c} - \frac{1}{\epsilon_b} \right) \\ & \times \left(\frac{J_1(G(R+d))}{G(R+d)} \right), \end{aligned} \quad (2)$$

where $f_1 = (2\pi/\sqrt{3})(R^2/a^2)$, $f_2 = (2\pi/\sqrt{3})[(R+d)^2/a^2]$ are the filling factors, $J_1(x)$ is the Bessel function of the first kind, and G is the reciprocal lattice vector in two dimensions. The band structure shown in Fig. 2(a) corresponds to a triangular lattice of air holes coated with a thin layer ($0.1a$) of porphyrin for the TE polarization meaning that the magnetic field $\mathbf{H}$ is parallel to the hole axis. The TE polarization is chosen because there is no photonic bandgap for the TM one.

To model the SHD cavity we have developed a code based on the supercell method that takes into account the

Fig. 2. (Color online) (a) Dispersion diagram of a triangular lattice without a defect, where a photonic bandgap gives rise between $\omega a/2\pi c = 0.334$ and $\omega a/2\pi c = 0.415$. (b) Zoom-in on the photonic bandgap of the cavity where the resonant mode of the SHD cavity appears at $\omega a/2\pi c = 0.3761$.

defect center inside the hexagonal matrix of holes and the porphyrin layer around the holes. The supercell unit is defined by 9×9 holes, which is the minimum superlattice size that avoids splitting of resonances. All parameters of the SHD simulations are defined relative to the period a of the lattice. By studying the influence of the radius R of the lattice holes, the thickness d of the sensitive layer, and the radius r of the defect hole, we have determined a configuration that enables a resonant mode at the center of the bandgap while meeting the technological requirements of fabrication. This compromise was found with the configuration $d=0.1a$, $R=0.3a$, and $r=0.1a$. With such a configuration, the defect hole is totally filled by the porphyrin layer.

Figure 2(b) shows the resonant mode for the SHD cavity for TE polarization. As can be seen in the figure, the configuration leads to a normalized resonance frequency of $\omega a/(2\pi c) = 0.3761$, which implies a lattice period $a = 158$ nm for the operating wavelength $\lambda = 419.5$ nm.

The sensitivity of the structure to changes in the thickness of the porphyrin was evaluated by the PWE method combined with the supercell method as described above. The results can be seen in Fig. 3 and they show that the resonance shift increases with the thickness of the sensitive layer. More precisely, we can conclude from Fig. 3 that a variation in thickness of $0.04a$ leads to a shift in the resonance frequency of $16 \cdot 10^{-3}(c/a)$, which means that a variation in thickness of 1 nm of the sensitive layer

Fig. 3. Plot of resonance shift as function of monolayer optical thickness.

implies a shift of 0.5 nm of the resonance peak at the operating wavelength of 419.5 nm.

Though difficult to realize, such a device is nevertheless compatible with the actual machining techniques. Until now, the best reported performances on lithium niobate nanostructuration have been achieved by focused ion beam (FIB) milling. The Carz Zeiss XB1540 FIB system or FEI's newest dual beam TM (Helios Nanolab family) exhibit a resolution of 5 nm at 30 keV. These new techniques enable us to be confident about the realization of holes with a diameter of 32 nm (radius of 16 nm). Moreover, the scaling of PhC features for sensitive layers having an absorption peak >420 nm is also possible.

These preliminary results show the interest of the PWE method for estimating the optimal parameters of the cavity coated with a thin sensitive layer. They also emphasize a sensitivity that can advantageously be compared to what can be reported with other configurations (see [5] for example). However, a wavelength shift lower than 1 nm is very difficult to detect: we propose to exploit the absorption of the layer in view of enhancing the variation in the transmission response in the presence of benzene. These further developments have been investigated by the 2D-FDTD method.

3. ANALYSIS OF THE PORPHYRIN MONOLAYER

As previously explained, the aim is to make the resonance peak of the PhC microcavity coincide with the absorption peak of the porphyrin layer. The purpose is to exploit both the maximum variation of the imaginary part of the dielectric constant and the maximum variation of the refraction index, as can be deduced from the Kramers–Kronig relations.

In view of evaluating the influence of the layer's absorption on the transmission response, we propose to fit the permittivity ε_L of the porphyrin's layer with a Lorentz dispersion model that will be implemented in a FDTD code [17]. More precisely, the analytical expression of dielectric permittivity ε_L is modeled as

$$\varepsilon_L(\omega) = \varepsilon_\infty - \frac{\Delta\varepsilon\Omega_L^2}{(\omega^2 - \Omega_L^2) + i\Gamma_L\omega}, \quad (3)$$

where Ω_L is the oscillator strength, Γ_L is the spectral width of the Lorentz oscillators, $\Delta\varepsilon$ is the weighting factor

[18], and finally $\varepsilon_\infty = n_0^2$ where $n_0 = 1.65$ denotes the refractive index of the layer far from the resonance. The refractive index n and the extinction coefficient κ are deduced from the real and the imaginary parts of $\sqrt{\varepsilon_L}$, respectively:

$$n = \sqrt{\frac{1}{2} \left(\varepsilon_\infty + \sqrt{\varepsilon_\infty^2 + \frac{\Delta\varepsilon^2\Omega_c^2}{\Gamma_L^2}} \right)}, \quad (4)$$

$$\kappa = \sqrt{\frac{1}{2} \left(-\varepsilon_\infty + \sqrt{\varepsilon_\infty^2 + \frac{\Delta\varepsilon^2\Omega_c^2}{\Gamma_L^2}} \right)}. \quad (5)$$

The extinction coefficient κ can be evaluated from the experimental measurement of absorbance as described in [19]. The resulting experimental curve is reported with a solid line in Fig. 4(a). This experimental curve was fitted to the Lorentz dispersion model expressed in Eq. (5) by a least-squares method. The fitting parameters are presented in Table 1, and the resulting fitting curve is represented with a dashed line in Fig. 4(a).

The frequency dependence of the refractive index was then deduced from the Lorentz model expressed in Eq. (4) and from the fitting parameters reported in Table 1. The resulting figure of dispersion is reported in Fig. 4(b) showing a maximum index variation of only 10^{-5} .

The adsorption of gas molecules by the porphyrin layer provokes a change in the layer's absorption peak. This variation can be modeled by a variation in the weighting factor $\Delta\varepsilon$. The relationship between the weighting factor $\Delta\varepsilon$ and the concentration of gas molecules has been estimated from [19] and experimental results that were performed in our laboratory. In the absence of benzene, the weighting factor is estimated to be $\Delta\varepsilon = 8.6 \times 10^{-6}$ while it becomes $\Delta\varepsilon = 7.7 \times 10^{-5}$ in the presence of 45 ppm of benzene. Thus, a concentration of benzene of 5 ppm corresponds to a weighting factor of $\Delta\varepsilon = 8.6 \times 10^{-6}$, and it becomes $\Delta\varepsilon = 8.6 \times 10^{-5}$ in the presence of 50 ppm of benzene.

The variation in the weighting factor is reverberated on both the extinction coefficient and the refractive index, as can be seen in Eqs. (4) and (5). We have represented the influence of the weighting factor on the refractive index and on the extinction coefficient in Figs. 9(a) and 9(b), respectively. The refractive index and the extinction coefficient were evaluated at the maximum of absorption of the

Fig. 4. (a) Spectral variation of the extinction coefficient of the porphyrin layer; the solid line corresponds to the experimental data and the dashed line corresponds to the Lorentzian fitting. (b) Refraction index variation of the porphyrin layer.

Table 1. Parameters for the Lorentz Dispersion Model Described in Eq. (3)

Dispersion Model	Resonance Frequency	FWHM	Weighting Factor
Lorentz model	$\Omega_L = 4.534 \times 10^{15}$ rad/s	$\Gamma_L = 2 \times 10^{14}$ rad/s	$\Delta\varepsilon = 0.086 \times 10^{-4}$

porphyrin layer. We can conclude from Figs. 9(a) and 9(b) that the weighting factor $\Delta\varepsilon$ influences more the extinction coefficient than the refractive index. Indeed, a weighting factor of $\Delta\varepsilon = 8.6 \times 10^{-5}$ induces a variation in the refractive index of only 10^{-7} , which can be compared to the 6×10^{-4} variation in the extinction coefficient. This behavior was implemented in a 2D-FDTD code in order to evaluate the changes in the transmission response in the presence of benzene molecules.

4. FDTD SIMULATIONS AND ABSORPTION EFFECT

2D-FDTD calculations based on a homemade code were performed to analyze the resonant mode. Indeed, 2D-FDTD calculations provide an estimation of the peak behavior provided that the holes are deep enough [20] (typically deeper than six times the diameter). The SHD cavity structure data were generated using MATLAB, and the defect hole position was then integrated to define the complete PhC structure [Fig. 1(a)]. The PhC structure was modeled with an incident beam which consists of a Gaussian pulse centered at $\lambda = 419.5$ nm and having a temporal width allowing the calculation of transmission over a wide spectral range. In order to faithfully describe both the holes and the porphyrin monolayer, we used a sub-gridding technique [21]. By the way, one period of the PhC is initially discretized with $(20 \times 82) \times (20 \times 41)$ nodes. Then, an average value of the permittivity is calculated over 20×20 nodes in order to get 82×41 nodes for one period in the FDTD calculations. Simulations were made for the TE polarization and for the ΓM direction. The different transmission spectra of the cavity were calculated for wavelengths ranging from 350 to 550 nm. Figure 5(b) presents the transmission spectrum of the configuration shown in Fig. 5(a): the resonance peak appears at $\lambda = 419.5$ nm in the photonic bandgap, with a quality factor of Q around 700. Such a quality factor is not so adequate for the refractive index sensor [22], but in our case, we studied the influence of the layer's absorption on the transmittivity of the PhC and not the mode shift.

In Fig. 6 we present the influence of the mode cavity resonance versus $\Delta\varepsilon$. Both the amplitude and the position

Fig. 5. (a) Scheme of the photonic structure simulated by FDTD where the light propagates in the ΓM direction. The incident Gaussian beam has a beam waist $W_0 = 2 \mu\text{m}$. (b) Transmission spectrum of the PhC cavity, where the resonant mode appears in the photonic bandgap at $\lambda = 419.5$ nm.

of the transmission peak are affected. The transmittivity at the resonance peak is plotted as a function of the wavelength peak λ_r in Fig. 7(a) and as a function of $\Delta\varepsilon$ in Fig. 7(b). In addition, we have determined the electromagnetic field distribution using our FDTD code (Fig. 8) without and with the absorption phenomenon. Figures 8(a) and 8(d) show the square root of the magnetic field amplitude for $\Delta\varepsilon = 8.6 \times 10^{-6}$ (i.e., without benzene) and for $\Delta\varepsilon = 8.6 \times 10^{-3}$ (i.e., in the presence of 5000 ppm of benzene), respectively. As it is shown, the magnetic field is strongly confined around the defect. Figures 8(b) and 8(c) and Figs. 8(e) and 8(f) present, respectively, the square root of the electric field's amplitude for the same cases as described above. As it is well shown on these last figures, the electric field is confined in the defect region where the defect hole was totally filled by the porphyrin layer. Note that the confinement of intensity inside the cavity is almost 60 times smaller when the weighting factor is increased 1000 times. Though not realistic ($\Delta\varepsilon = 8.6 \times 10^{-3}$ corresponds to 5000 ppm of benzene), the use of such values helps emphasize the influence of absorption upon the intensity distribution.

Fig. 6. (Color online) Variation in the transmittivity at the cavity mode for different values of $\Delta\varepsilon$.

Fig. 7. (Color online) (a) and (b) present, respectively, the square root of the amplitude of the magnetic and electric fields, without considering the absorption effect. (d) and (e) present those for $\Delta\varepsilon = 8.6 \times 10^{-3}$ (corresponding to 5000 ppm of benzene). [(c), (f)] Zoom-in at the defect region in the case of the electric field distribution.

Fig. 8. (Color online) (a) Transmittivity as a function of the resonance wavelength, where a slight red shift is obtained by decreasing $\Delta\epsilon$. (b) Transmittivity through the cavity as a function of $\Delta\epsilon$. $\Delta\epsilon=8.6\times 10^{-6}$ corresponds to the porphyrin layer without benzene. $\Delta\epsilon=8.6\times 10^{-5}$ corresponds to a presence of 50 ppm of benzene and $\Delta\epsilon=8.6\times 10^{-4}$ to a presence of 500 ppm of benzene.

From these results, we can conclude that the resonance peak encounters a variation in transmittivity of 23% when the weighting factor varies from 8.6×10^{-6} (i.e., without benzene) to 8.6×10^{-5} (i.e., in the presence of 50 ppm of benzene), which corresponds to a variation in the refractive index of 10^{-7} [see Fig. 9(a)] and a variation of 6×10^{-4} for the extinction coefficient [see Fig. 9(a)]. Consequently, the sensitivity of the device is estimated to be of about 3 ppm. Indeed, by considering that the minimum detectable variation in transmittivity is of 1%, we have calculated that 2.3 ppm is the minimum concentration of benzene that can be detected. This value is strongly dependent on the performances of the sensitive layer: improving its sensitivity would enable detection of lower quantities of benzene.

Fig. 9. (a) Refractive index and (b) extinction coefficient variation function of the weighting factor $\Delta\epsilon$ calculated from Eqs. (4) and (5), respectively.

5. CONCLUSION

We have presented a PhC with a single-hole defect (SHD) functionalized by a sensitive absorbent layer for sensing applications. The advantage of this configuration relies on an enhanced sensitivity that is not only due to a variation

in the thickness and refractive index, but also to the variation in absorption in the presence of the targeted molecules.

We have implemented a supercell method that takes into account the thin sensitive layer. First, the resonance peak has been estimated to encounter a shift of 0.5 nm per 1 nm variation in the layer's thickness. Second, we have studied the behavior of the cavity by using 2D-FDTD simulations that includes a Lorentz dispersion model to take into account the layer's absorption. The sensitivity of the sensor is considerably enhanced by working at the peak of absorption of the sensitive layer. The variation in transmittivity at the resonance peak was estimated to be of 23% for an index variation of only 10^{-7} . This study was dedicated to the detection of benzene, but it can be applied to the realization of new compact dynamic sensors for biomedical or environmental applications.

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