Slow-light enhanced optical detection in liquid-infiltrated photonic crystals

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Abstract Slow-light enhanced optical detection in liquid-infiltrated photonic crystals is theoretically studied. Using a scattering-matrix approach and the Wigner–Smith delay time concept, we show that optical absorbance benefits both from slow-light phenomena as well as a high filling factor of the energy residing in the liquid. Utilizing strongly dispersive photonic crystal structures, we numerically demonstrate how liquid-infiltrated photonic crystals facilitate enhanced light-matter interactions, by potentially up to an order of magnitude. The proposed concept provides strong opportunities for improving existing miniaturized absorbance cells for optical detection in lab-on-a-chip systems.

1 Introduction

Optical techniques are finding widespread use in chemical and bio-chemical analysis, such as absorbance, fluorescence, Raman-scattering, and surface-plasmon-resonance measurements, see Mogensen et al. (2004); Hess et al. (2002); Kneipp et al. (2002); Homola et al. (1999). In particular, the Beer–Lambert absorbance measurement has become one of the classical quantitative workhorses in analytical chemistry, Skoog et al. (1997). The increasing emphasis on miniaturization of chemical analysis systems during the past decade, Janasek et al. (2006), has naturally stimulated a considerable effort in integrating microfluidics, Squires and Quake (2005); Whitesides (2006), and optics in lab-on-a-chip microsystems, Verpoorte (2003); Mortensen et al.
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Fig. 1 Panel (a) illustrates a typical macroscopic cuvette while panel (b) shows a microscope image (top-view) of an equivalent lab-on-a-chip implementation of a microfluidic channel (vertical direction) integrated with planar optical waveguides (horizontal direction). Courtesy of K. B. Mogensen and J. P. Kutter (MIC – Department of Micro and Nanotechnology, Technical University of Denmark, www.mic.dtu.dk).

(2007), and more recently this has to a large extent powered the emerging field of optofluidics, see Psaltis et al. (2006); Monat et al. (2007). However, lab-on-a-chip implementations of the above mentioned optical techniques call for new approaches going beyond a simple miniaturization of existing optical setups. The need for a new perspective is perhaps best appreciated by emphasizing the optical drawback of miniaturization. Reducing the optical path length will eventually also decrease the sensitivity significantly. In other words, the sensitivity fundamentally scales with volume hosting the interaction between light and the solution of analytes.

Panel (b) in Fig. 1 illustrates a typical lab-on-a-chip implementation of an absorbance cell. The optical path length $L$ is often reduced by several orders of magnitude compared to typical macroscopic counterparts, such as the cuvette shown in panel (a). A typical size reduction by two orders of magnitude will penalize the optical sensitivity in an inversely proportional manner, Mogensen et al. (2003).

In the above context, photonic crystals (PhC) are potential candidates for the desired new approach on the light-matter interactions as recently reviewed by Mortensen et al. (2007). The concept of PhCs refers to a class of artificial electromagnetic structures offering highly engineered dispersion properties, as first suggested by Yablonovitch (1987); John (1987). PhCs are porous structures, but with the voids arranged in a highly regular way. Thus,
for a wavelength $\lambda$ comparable to the periodicity $\Lambda$ of the PhC, the electromagnetic radiation interacts strongly with the matter. PhCs have over the years been recognized as strongly dispersive environments supporting a number of phenomena including photonic band gaps and slow-light propagation, see Joannopoulos et al. (1995); Sakoda (2005). The key-point in the present context is that liquid-infiltrated PhCs will overall inherit the unusual dispersion properties from their non-infiltrated counterparts, thus also changing the way light interacts with bio-molecules dissolved in the liquid, Mortensen et al. (2007). The effects can be quite pronounced compared to light-matter interactions in a spatially homogeneous liquid environment and in this paper we follow this route to compensate the reduced optical path.

In particular, we consider slow-light enhanced optical detection in liquid-infiltrated photonic crystals, thus potentially compensating for the cost of miniaturization and reduction in optical path length.

The enhancement factor $\gamma \equiv \alpha/\alpha_l$ for an absorbing liquid (with absorption coefficient $\alpha_l$) can be expressed as

$$\gamma = f \times \frac{c/n_l}{v_g},$$

as recently derived by Mortensen and Xiao (2007). Here, $0 < f < 1$ is a dimensionless number quantifying the relative optical overlap with the liquid. The fraction on the right-hand side expresses the ratio of the group velocity $c/n_l$ in the bare liquid to the group velocity $v_g$ in liquid-infiltrated photonic crystal, thus clearly illustrating the enhancement by slow-light propagation ($v_g \ll c$). In the following, we first derive the above expression before we with numerical simulations illustrate the slow-light enhancement in liquid-infiltrated PhCs with three different periodic structures.

2 Theory

2.1 Electromagnetic wave equation

The electromagnetic problem that we consider is governed by the electromagnetic wave equation for the electrical field,

$$\nabla \times \nabla \times |E\rangle = \varepsilon \frac{\omega^2}{c^2} |E\rangle,$$

where for the absorbing liquid the dielectric function $\varepsilon$ has a small imaginary part, $\omega$ is the angular frequency, and $c$ is the velocity of light in vacuum. In a recent paper, Mortensen and Xiao (2007) derived Eq. (1) rigorously with the aid of standard first-order electromagnetic perturbation theory

$$\delta\omega = -\frac{\omega}{2} \frac{\langle E | \delta\varepsilon | E \rangle}{\langle E | \varepsilon | E \rangle}$$

(3)
and more details may be found in our recent work, see Mortensen et al. (2007). In Eq. (3), \( \delta \omega \) is the first-order frequency shift introduced by a perturbation \( \delta \varepsilon \) of the dielectric function \( \varepsilon \) (assumed frequency independent) with the electric field being the unperturbed one. In the present context the perturbation is associated with absorption in the liquid-part of space thus giving rise to an imaginary frequency shift. In the following we emphasize this by explicitly writing the shift as \( i \delta \omega \) with \( \delta \omega \) being real. For the temporal dependence of the intensity we have \( I(t) = I_0 |e^{-i\omega t}|^2 = e^{-2i\delta \omega t} \) so that we may define an absorption rate given by \( \Gamma = 2 \delta \omega \).

2.2 Scattering-matrix approach

Here, we will emphasize the light-matter interaction time and offer an alternative derivation based on scattering matrices combined with the concept of the Wigner–Smith delay time. Following Beenakker and Brouwer (2001) we start from the scattering matrix

\[
S(\omega) = \begin{pmatrix} S_{11}(\omega) & S_{12}(\omega) \\ S_{21}(\omega) & S_{22}(\omega) \end{pmatrix} = \begin{pmatrix} r(\omega) & t'(\omega) \\ t(\omega) & r'(\omega) \end{pmatrix}
\]

(4)

where \( S_{ij} \) are the S-parameters connecting incident and out-going plane-wave (or Bloch) states to the left and the right of the scattering region. Next, let \( S_0(\omega) \) denote the scattering matrix in the absence of absorption so that for a weak absorption we have a small imaginary shift \( i \delta \omega \) in frequency and thus \( S(\omega) \approx S_0(\omega + i \delta \omega) \). Taylor expanding the right-hand side we get

\[
S(\omega) \approx S_0(\omega) + i \delta \omega \frac{\partial S_0(\omega)}{\partial \omega} = S_0(\omega) \left[ 1 - \Gamma \frac{\partial}{\partial \omega} Q_0(\omega) \right].
\]

(5)

In the second equality \( \Gamma = 2 \delta \omega \) is the absorption rate and we have furthermore used the unitarity of the scattering matrix, \( S_0^\dagger S_0 = 1 \), to introduce the Hermitian Wigner–Smith delay time matrix

\[
Q_0(\omega) = -i S_0^\dagger(\omega) \frac{\partial S_0(\omega)}{\partial \omega}.
\]

(6)

Now, we would like to calculate the transmission probability \( T \) through a single eigenmode in which case \( S \) is a two-by-two matrix. Assuming time-reversal symmetry we have \( T = |S_{12}|^2 = |S_{21}|^2 \) which we have the freedom to rewrite as

\[
T(\omega) = \frac{1}{2} \text{Tr} \{S(\omega)S^\dagger(\omega)\} + \mathcal{O}(1 - T).
\]

(7)

For near-resonance transmission, i.e. for a low reflection, we get

\[
T(\omega) \approx \frac{1}{2} \text{Tr} [1 - \Gamma Q_0(\omega)] = \frac{1}{2} \sum_{n=1}^{2} [1 - \Gamma_t n(\omega)] = 1 - \Gamma \tau
\]

(8)
where we have used the unitarity of $S_0$ together with the cyclic invariance of the trace. In the second equality we have introduced the eigenvalues $t_1 = t_2 = \tau$ of the Wigner–Smith delay time matrix where $\tau$ is often referred to as the dwell time. We may use perturbation theory, Eq. (3), to calculate the absorption rate which gives

$$\Gamma = f \times \Gamma_l, \quad f = \frac{\langle E | D \rangle}{\langle E | D \rangle}$$  \hspace{1cm} (9)

where we have introduced the displacement field $|D\rangle = \varepsilon |E\rangle$ and $\Gamma_l$ is the absorption rate of the liquid itself. For the filling factor we have $0 < f < 1$ and the integral in the numerator is restricted to the region containing the absorbing fluid while the integral in the denominator is spatially unrestricted. The presence of the filling factor $f$ is quite intuitive since only the fraction $f$ of the light residing in the fluid can be subject to absorption. Note that compared to the definition normally used by the photonic crystal community, Joannopoulos et al. (1995), our definition quantifies the relative overlap with the liquid-part of space rather than the dielectric part given by $1 - f$. Finally, we rewrite Eq. (8) by re-exponentiating, i.e. $(1 - x) \longrightarrow \exp(-x)$, so that

$$T = \exp(-\Gamma \tau) \equiv \exp(-\gamma \times \Gamma_l \tau_l)$$  \hspace{1cm} (10)

corresponding to the expected exponential damping of the resonance due to absorption. In the second equality we have introduced $\gamma \equiv \alpha / \alpha_l = (\Gamma \tau)/(\Gamma_l \tau_l)$ and using that $\tau = L / v_g$ for the periodic medium and $\tau_l = L / (c/n_l)$ in the bare liquid we finally arrive at Eq. (1).

The expression for $\gamma$ clearly demonstrates how optical absorbance benefits from slow-light phenomena. The effective enhancement of the absorbance may thus naturally be interpreted as an effective enhancement of the light-matter interaction time. These conclusions may be extended to also non-periodic systems, including enhanced absorbance in disordered systems as well as intra-cavity absorbance configurations, see Mortensen et al. (2007).

In general, fluids/liquids have a smaller refractive index than the typical solid materials comprising the photonic crystal itself. Thus, increasing the filling factor (and thereby the light-liquid interaction) implies the confinement or guidance of light in the low index regions, which is typically not the case explored in more ordinary applications of PhCs for e.g. optical communication purposes. However, a high light-liquid overlap can be achieved in some structures, e.g., slot waveguides, hollow anti-resonant reflective optical waveguides, as well as photonic-crystal structures. For liquid-infiltrated photonic crystals and photonic crystal waveguides, it is possible to achieve $v_g \ll c$ and at the same time have a filling factor of the order unity, $f \sim 1$, whereby significant enhancement factors become feasible.
Fig. 2 (a) Photonic band structure for normal incidence of either TE or TM polarized light on a Bragg stack of period $\Lambda = a_1 + a_2$ with $n_1 = 1.33$, $n_2 = 3$, $a_1 = 0.6863\lambda$, and $a_2 = 0.3137\lambda$. Photonic band gaps are indicated by yellow shading and the dashed line indicates the long-wavelength asymptotic limit, Eq. (11). (b) Corresponding enhancement factor which peaks and exceeds unity close to the photonic band-gap edges. The dashed line indicates the long-wavelength asymptotic limit, Eq. (12).

3 Numerical examples

Having derived a formal expression for the enhancement factor $\gamma$, we next apply a full-vectorial plane wave method of Johnson and Joannopoulos (2001) to calculate the eigenmodes of Eq. (2) in the non-absorbing limit and from these solutions we evaluate the enhancement factor, Eq. (1), for a number of geometries.

3.1 Bragg stack

Let us first illustrate the slow-light enhancement for the simplest possible structure which is a Bragg stack with normal incidence of electromagnetic radiation, as shown in the inset of panel (b) of Fig. 2. The structure is composed by the low-index material layers of width $a_1 = 0.6863\lambda$ being a liquid with refractive index $n_1 = 1.33$ and the high-index layers have a width $a_2 = 0.3137\lambda$ and a refractive index $n_2 = 3$. Panel (a) of Fig. 2 shows the
corresponding band structures, where photonic band gaps are indicated by yellow shading and the dashed line indicates the long-wavelength asymptotic limit to a close-to-linear dispersion

$$\omega(\kappa) \simeq \frac{ck \Lambda}{a_1 n_1 + a_2 n_2}. \quad (11)$$

When approaching the band-gap edges, the dispersion flattens. Note that the first band is a dielectric-like one, see e.g. Mortensen et al. (2007); Joannopoulos et al. (1995), meaning that most of the energy localizes in the high dielectric region. For the first band it is well-known that the flat dispersion originates from a spatial localization of the field onto the high-index layers and thus \( f \ll 1 \) near the band edges where the inverse group velocity diverges. However, in spite of the localization, the enhancement factor may still exceed unity as shown in panel (b) where the dashed line indicates the long-wavelength asymptotic limit with

$$f \simeq \frac{\langle 1 | \epsilon | 1 \rangle}{\langle 1 | \epsilon | 1 \rangle} = \frac{a_1 n_1^2}{a_1 n_1^2 + a_2 n_2^2}. \quad (12)$$
Fig. 4 (a) Photonic band structure for propagation of TE polarized light along the ΓX direction in a PhC waveguide in a triangular lattice of period Λ with holes of radius $r = 0.37\Lambda$ and $n = 3.24$. The PhC waveguide is formed by enlarging the radius ($r_1 = 0.50\Lambda$) of the air holes in a single row, which are infiltrated by a liquid with $n_l = 1.33$. The grey shading indicates the regions of finite density of states in the surrounding PhC while the yellow shading shows the waveguide mode-gap region. (b) Corresponding enhancement factor $γ$. The inset shows the structure of the photonic crystal waveguide.

For the second band, the enhancement factor still exceeds unity, which is attributed both to slow-light, as well as to the large filling factor. The filling factor is about 20% at the band edge for the second band. In order to further benefit from the slow-light enhanced light-matter interaction, we obviously have to pursue optofluidic structures supporting both low group velocity and at the same time large filling factors.

3.2 Pillar-type photonic crystal

Figure 3 shows the case for a 2D square photonic crystal with high-index dielectric rods in liquid. Compared to the Bragg stack in Fig. 2, some of the modes in this structure have both a low group velocity and at the same time a reasonable value of the filling factor $f$. In particular, the fourth band in panel (a) is quite flat. The corresponding enhancement factor $γ$ exceeds
4 for a bandwidth of order 100 nm for a pitch around $\Lambda \sim 1000\,\text{nm}$, as indicated on the right $y$-axis in panel (b).

### 3.3 Void-type photonic crystal waveguide

Recently, it was shown by Gersen et al. (2005) how photonic crystal waveguides can be used to slow down the propagation velocity of light. Here, we illustrate how this may be utilized for slow-light enhancement in a PhC waveguide structure. We consider the PhC waveguide shown in the inset in panel (b) of Fig. 4, where the PhC is two-dimensional with a triangular array of holes in an InP/GaInAsP dielectric background with a refractive index of $n = 3.24$. The radius of the air holes is $r = 0.37\,\Lambda$. In order to realize a non-solid waveguide, the PhC waveguide is formed by slightly enlarging the radius ($r_1$) of the air holes in a single row which are subsequently utilized for liquid-infiltration. In order to obtain a high filling factor, we optimize the waveguide structure and obtain $f \approx 40\%$ when $r_1 = 0.5\,\Lambda$. The enhancement factor exceeds unity near the band edge, which is attributed to both slow-light propagation as well as to the large filling factor.

### 4 Conclusion

In conclusion, we have studied the potential of using liquid-infiltrated photonic crystals to enhance optical absorbance. Using a scattering-matrix approach we have re-derived a general expression for the enhancement factor, thus confirming the interaction-time interpretation offered in previous work. Our results clearly demonstrate how optical absorbance may benefit from slow-light phenomena combined with a filling factor close to unity. With the aid of photonic crystals, it is possible to design an optical enhanced detection system which has both a slow group velocity and a high filling factor. The slow-light enhancement of the absorption, by possibly an order of magnitude, may be traded for yet smaller miniaturized systems or for increased sensitivity of existing devices.

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