Liquid crystal infiltration of InP-based planar photonic crystals

J. Martz,^{a)} R. Ferrini, F. Nüesch,^{b)} and L. Zuppiroli

Laboratoire d'Optoélectronique des Matériaux Moléculaires, École Polytechnique Fédérale, CH-1015 Lausanne, Switzerland

B. Wild, L. A. Dunbar, and R. Houdré

Institut de Photonique et d'Electronique Quantique, École Polytechnique Fédérale, CH-1015 Lausanne, Switzerland

M. Mulot^{c)} and S. Anand

Department of Microelectronics and Information Technology, Royal Institute of Technology, S-16440, Kista, Sweden

(Received 30 November 2005; accepted 3 March 2006; published online 26 May 2006)

A procedure for the infiltration of planar photonic crystals (PhCs) with liquid crystals (LCs) is presented. InP-based PhCs are infiltrated with the nematic LC-K15 in a specially designed high-vacuum chamber. The infiltration technique is validated and systematically characterized by measuring the transmission through the infiltrated PhCs at different temperatures and for different polarizations. The reproducibility and reliability of our procedure are demonstrated and a high filling efficiency is obtained. © 2006 American Institute of Physics. [DOI: 10.1063/1.2199090]

I. INTRODUCTION

In the past decade great effort has been devoted to the study of photonic crystals (PhCs), which are artificial materials that offer the possibility to control light on its wavelength scale.¹ Using this property, several applications and devices have been proposed and experimentally demonstrated in different fields from quantum optics to integrated optics.¹ Unfortunately, there are still many factors limiting the use of PhCs in real devices, such as fabrication imperfections,² losses,^{3,4} and temperature sensitivity.⁵ However, by replacing the low index element of the PhC with different materials, e.g., infiltrating the air pores with a synthetic organic material that has a tunable refractive index, can overcome some of these issues by either controlling the optical properties of the PhC on demand (*tuning*) or compensating the temperature sensitivity or the imperfections of the PhC itself (*trimming*).^{6–13} This research topic may have a huge technological impact in various application fields from integrated optics¹⁴ to detection and sensing.¹⁵

The potential of PhC infiltration with liquid crystals (LCs) has been already demonstrated for one-, two- and three-dimensional (1D, 2D, and 3D) PhCs.^{1,6–13,16} The characteristic optical anisotropy (birefringence) of LCs due to the molecule configuration and alignment allows one to easily modify their refractive index n , and thus the optical properties of the entire PhC. In this article, we will show how infiltrating InP-based planar PhC devices with LCs allows one to trim their optical response. A technique that uses a specially designed vacuum chamber is presented. The reli-

ability and reproducibility of the infiltration process are assessed by the optical characterization of the infiltrated structures.

II. SAMPLE DETAILS AND OPTICAL CHARACTERIZATION

Planar PhCs consisting of a triangular lattice of air holes were etched by chemically assisted ion beam etching (CAIBE) through a nominally undoped InP/(Ga,In) $\times$ (As,P)/InP vertical waveguide grown by metalorganic vapor phase epitaxy.^{17–19} In the wavelength range of interest (i.e., $\lambda \approx 1.5 \mu\text{m}$) the waveguide is single mode for both the transverse electric (TE) and magnetic (TM) polarization directions (i.e., electric field *perpendicular* and *parallel* to the hole axis, respectively). The effective refractive index is $n_{\text{eff}}=3.24$ (3.243) and 3.23 (3.233) for the TE and TM guided modes at the temperature $T=25^\circ\text{C}$ (40°C), respectively.⁵ Two GaAsInP quantum wells (QWs) were embedded in the core layer for optical measurements: An internal light source (ILS) technique¹⁷ was used to optically characterize the infiltrated PhC structures. The photoluminescence (PL) excited inside the QWs is used as a built-in probe beam. Part of the PL signal propagates parallel to the surface as a guided mode and interacts with the PhC structure before escaping from a cleaved edge where adequate light collection is performed.¹⁷ The absolute PhC transmission is obtained normalizing the spectrum measured after transmission through the PhC slab with respect to a spectrum collected in a non-patterned region of the sample. Since ILS measurements on a single PhC structure yield the transmission spectrum only in a narrow spectral interval ($\sim 100 \text{ nm}$), the lithographic tuning was adopted and the PhCs were replicated with different periods a . Thus, the absolute transmission through the PhC structures was measured as a function of the normalized photon energy $u=a/\lambda$. The

^{a)}Present address: Rolic Technologies Ltd, CH-4123 Allschwil, Switzerland.

^{b)}Present address: Laboratory for Functional Polymers, EMPA, CH-8600 Dübendorf, Switzerland.

^{c)}Present address: Optoelectronics Laboratory, Helsinki University of Technology, PL 3500, FIN-02015 TKK, Finland.

FIG. 1. High-vacuum chamber designed for the infiltration of photonic crystals.

sample was mounted on a Peltier stage to allow temperature tuning from 25 °C to 40 °C.⁵

The PhC structures were fabricated by electron beam lithography and chemically assisted ion beam etching.¹⁹ Two types of structures were fabricated with lattice periods $a=280\text{--}580\text{ nm}$ ($\Delta a=20\text{ nm}$): (i) eight-rows-thick Γ M-oriented PhC slabs; and (ii) Fabry-Pérot (FP) cavities consisting of two 4-rows-thick Γ M-oriented PhC mirrors separated by a spacer $W/a=1.7$. Due to the hole widening during the etching of the smallest holes (typical of CAIBE),¹⁹ the measured air filling factor f decreases from 0.50 to 0.42 when increasing a .¹⁸ That corresponds to a hole diameter $D=200\text{--}400\text{ nm}$.

III. LIQUID CRYSTAL INFILTRATION

The fabricated PhCs were infiltrated with the nematic LC-K15 from Merck that consists of only one type of molecules [4-cyano-4'-pentylbiphenyl (5CB)]. Its stability with respect to the infiltration procedure was verified by differential scanning calorimetric measurements.¹⁸ The nematic-to-isotropic phase transition temperature (clearing point) is $T_{c1}=35.1\text{ °C}$. For $\lambda \approx 1.5\text{ }\mu\text{m}$ the ordinary and extraordinary refractive indices at $T=25\text{ °C}$ are $n_o=1.516$ and $n_e=1.682$, respectively, whilst the isotropic refractive index at $T=40\text{ °C}$ is $n_i=1.575$.²⁰

The capillary forces assist the physical infiltration of porous materials with LCs.¹ Differently from the 3D- or the 2D-membrane cases, in substrate-based planar PhCs the air holes are accessible only from the top and the evacuation of gases from the pores may be difficult. Indeed it has been demonstrated^{8,12,18} that working under vacuum can help the infiltration process and minimize the effects of gas trapping either in the sample or in the LC blend. To this end a vacuum chamber specifically designed for infiltration experiments was built (see Fig. 1). The chamber is equipped to enable surface cleaning by low-power plasma, control of temperature and gas flows during the infiltration procedure, and, when necessary, chemical surface modification by molecule grafting.¹⁶

The best temperature and pressure conditions were investigated to optimize the in-filling process and to increase its reproducibility. The procedure can be divided into three steps:

(i) *Evacuation*: The vacuum chamber is pumped and heated, thus emptying the gaseous content of the holes and degassing the LC blend.

FIG. 2. Measured TE transmission spectra through (a) 8-rows-thick Γ M-oriented PhC slabs (see the inset) and (b) a Fabry-Pérot (FP) cavity between two 4-rows-thick Γ M-oriented PhC mirrors (see the inset) *with* and *without* LCs (black lines). W is the cavity width and a the PhC lattice period. The best-fit curves of the experimental spectra are shown (gray dotted lines).

(ii) *Contact*: Once a steady state is reached with a base pressure of 10^{-6} mbar , the PhC sample is ready to be infiltrated and is brought into contact with the LCs by immersing the sample into the reservoir.

(iii) *Infiltration*: The vacuum chamber is brought to atmospheric pressure. This is a key step of the infiltration process; the pressure differential between the chamber and the holes forces the LC molecules to penetrate deeply into the pores. Temperature is a fundamental parameter: during the infiltration procedure, the chamber is constantly kept at a temperature much higher than the clearing point ($T=110\text{ °C}$). This reduces the LC viscosity, thus favoring the pore infiltration, and avoids wall contamination by water residuals. Heating the chamber before PhC infiltration also allows the purification of the LCs, which is indeed essential. If LCs are not properly degassed, trapped gases may combine to generate bubbles inside the holes; heating and pumping the chamber boils out the trapped gases.

IV. EXPERIMENTS AND DATA ANALYSIS

The TE polarized transmission spectra at room temperature through the Γ M-oriented PhC slabs (see the inset) *without* and *with* LCs are shown in Fig. 2(a) (black lines). A well-defined stopgap appears in the uninfiltrated sample. Once the PhC is infiltrated, due to the reduced refractive index contrast $n_{eff}-n_{hole}$ between the semiconductor matrix and the holes, the photonic band gap shrinks and the band edges redshift: $\Delta u_{Diel}=-5 \times 10^{-3}$ and $\Delta u_{Air}=-5 \times 10^{-2}$ at the dielectric and air band edge, respectively. Moreover, in the air band the transmission increases from 0.30 *without* LCs up to 0.70 *with* LCs, thus indicating a strong reduction of the PhC losses after infiltration. In low-refractive index contrast planar PhCs perforating the vertical waveguide induces coupling of the guided wave to radiation modes into the claddings, thus resulting in out-of-plane losses.³ When infiltrating PhCs with a material whose refractive index is higher than that of the air (i.e., $n_{hole}>1$), the out-of-plane diffraction at the waveguide-hole interface is reduced. In order to confirm this, the spectra were fitted by means of a two-dimensional finite difference time domain (2D-FDTD) model assuming n_{eff} as the matrix index and n_{hole} as the hole refractive index.²¹ Out-of-plane losses were included into the 2D model by adding a phenomenological imaginary dielectric

constant ε'' in the holes.³ The measured values $f=0.42$ and 0.46 were assumed that correspond to the energy u (i.e., to the PhC period a in the lithographic tuning approach; see above)¹⁷ where the fitted band edges are located in the spectra *without* and *with* LCs, respectively.^{18,19} The independent fit parameters were ε'' and n_{hole} [the calculated spectra are shown in Fig. 2(a) (gray dotted lines)]; once the reference case $n_{hole}=1$ has been fully characterized yielding the f value for the measured PhC slabs, the absolute accuracy for the obtained ε'' and n_{hole} values is of the order of 10^{-3} and 10^{-2} , respectively. The values $n_{hole}=1$ and 1.59 ± 0.01 , and $\varepsilon''=0.045\pm0.001$ and 0.030 ± 0.001 were found for the PhCs *without* and *with* LCs, respectively, thus confirming the increase of n_{hole} and the consequent decrease of losses after infiltration.

Transmission spectra at room temperature through the FP cavities (see the inset) *without* and *with* LCs are shown in Fig. 2(b). In the latter case, because of the increase of n_{hole} , the FP resonance redshifts with respect to the uninfiltred sample: $\Delta u_{peak}=-1.38\times10^{-2}$. We remark that, although for a refractive index variation Δn in the holes $\Delta u_{peak}/\Delta n < \Delta u_{Air}/\Delta n$ (-2×10^{-2} and -8×10^{-2} , respectively), the change of the PhC optical properties is intrinsically better shown by the shift of a resonance peak with a reasonably large quality factor (i.e., ≥ 100) than by the shift of a smooth transmission band edge. The FP peaks were fitted using the Airy's formula given in Ref. 17: the calculated curves are shown in Fig. 2(b) (gray dotted lines). Due to the reduced refractive index contrast Δn , after infiltration the mirror transmission T_{4-rows} increases while its reflectivity R_{4-rows} decreases: $T_{4-rows}=0.013\pm0.005$ and 0.090 ± 0.005 and $R_{4-rows}=0.960\pm0.005$ and 0.887 ± 0.005 *without* and *with* LCs, respectively. Therefore, in agreement with the results obtained for simple slabs, the loss coefficient $L_{4-rows}=1-T_{4-rows}-R_{4-rows}$ decreases from 0.017 ± 0.005 to 0.013 ± 0.005 *without* and *with* LCs, respectively. Due to the change of both the mirror optical properties and the intracavity losses,^{18,19} the FP peak transmission (T_{peak}) increases and the cavity quality factor (Q) decreases; $T_{peak}=0.12\pm0.01$ and 0.57 ± 0.01 and $Q=310\pm5$ and 100 ± 5 *without* and *with* LCs, respectively.

A systematic characterization of the infiltration procedure was carried out by means of optical measurements. On one hand, several infiltration experiments were performed on the same sample under the same temperature and pressure conditions. The same results were found in terms of the optical properties of the PhC structures after infiltration,¹⁸ thus testifying to the great reproducibility and reliability of our in-filling technique. On the other hand, as shown in Ref. 12, the filling efficiency is a critical parameter, whose measurement is not easy. For instance, due to the particular morphology and geometry of planar PhCs, studying infiltration by means of standard scanning electron or atomic force microscopy is not straightforward. However, optical measurements have already proved to be very useful in the characterization of planar PhCs.^{3,17} Here the average infiltration efficiency η was accurately determined by measuring the TE and TM transmission spectra [see black lines in Figs. 3(a) and 3(b), respectively, and the inset] through infiltrated PhC slabs at a temperature $T=40^\circ\text{C}>T_{c1}$. Due to the birefringence of

FIG. 3. Experimental (black lines) and calculated (gray dotted lines) (a) TE and (b) TM transmission spectra through infiltrated eight-rows-thick Γ M-oriented PhC slabs (see Fig. 1). The orientation of the E field with respect to the hole axis and the light propagation direction (vector k) are sketched in the inset for both the TE and TM polarizations.

nematic LCs, the component of their dielectric constant along the electric field (and thus the effective n_{LC} value) depends on the molecule orientation. However, when working at a temperature above the clearing point, LCs are in an isotropic phase and the dielectric constant is a scalar, independent from the molecule orientation, i.e., $n_{LC}=n_i$. Following the same procedure as for the fit of the spectra at room temperature (see above), the TE and TM air band transmission edges were fitted by means of the 2D-FDTD model with $f=0.46$ and 0.48 , respectively,^{18,19} and ε'' and n_{hole} as free fitting parameters; the calculated spectra are shown in Fig. 3 (gray dotted lines). Concerning losses, while the value $\varepsilon''=0.010\pm0.001$ was obtained for the TE polarization, $\varepsilon''=0$ was taken for the TM polarization, since in this case only the fit of the energy position of the air band edge is meaningful. The fit yielded the value $n_{hole}=1.50\pm0.01$ for both polarizations, thus confirming that at $T=40^\circ\text{C}$ the infiltrated LCs are in their isotropic phase. The same result is obtained by the 2D-FDTD fit of the resonance energy in the transmission spectrum through the FP cavities. The infiltration efficiency is defined as the percentage of the PhC slab that is filled, i.e., $\eta=(n_{hole}-1)/(n_{LC}-1)$. The accuracy of η depends on the accuracy of both n_{hole} and n_{LC} ; the latter quantity is usually obtained from independent measurements with an accuracy better than 10^{-2} (e.g., see Ref. 20), so that the accuracy of η is limited by that of n_{hole} and is of the order of 2×10^{-2} . Since $n_{LC}=n_i=1.575$,²⁰ the value $\eta=0.87\pm0.02$ is obtained. The latter is an average value and it does not contain any local information on the partial or complete filling of single holes. Moreover, we remark that the hole size seems to have no influence on the infiltration process; the same efficiency is obtained for $D=250-340$ nm.

V. CONCLUSIONS

In conclusion, the trimming of the optical properties of planar PhCs by LC infiltration was demonstrated. A high-vacuum chamber specifically designed for the PhC infiltration was realized. A reproducible and reliable filling procedure was developed and systematically characterized by means of optical measurements. With respect to previous studies,¹² our technique allows the accurate control of all the infiltration parameters (i.e., temperature and pressure) and, when necessary, the modification of the surface chemical

state.¹⁶ The infiltration efficiency η was accurately determined by measuring transmission spectra in both the TE and TM polarizations and at a temperature higher than the LC clearing point.

ACKNOWLEDGMENTS

The authors would like to acknowledge Dr. J. Moosburger from Technische Physik, University of Würzburg (Würzburg, Germany) for the electron-beam lithography of the InP-based PhCs. The PhC samples were fabricated in the framework of the European project Photonic Crystal Integrated Circuits (PCIC Contract No. IST-1999-11239). The infiltration experiments were performed in the framework of the Swiss National Center of Competence in Research (NCCR) in Quantum Photonics, and the European Network of Excellence on Photonic Integrated Components and Circuits (ePIXnet Contract No. 004525).

¹*Photonic Crystals*, edited by K. Busch, S. Lölkes, R. B. Wehrspohn, and H. Föll (Wiley-VCH, Weinheim, Germany, 2004).

²M. Skorobogatiy, G. Bégin, and A. Talneau, *Opt. Express* **13**, 2487 (2005).

³R. Ferrini, R. Houdré, H. Benisty, M. Qiu, and J. Moosburger, *J. Opt. Soc. Am. B* **20**, 469 (2003).

⁴R. Ferrini, B. Lombardet, B. Wild, R. Houdré, and G.-H. Duan, *Appl. Phys. Lett.* **82**, 1009 (2003).

⁵B. Wild, R. Ferrini, R. Houdré, M. Mulot, S. Anand, and C. J. M. Smith,

Appl. Phys. Lett. **84**, 846 (2004).

⁶K. Busch and S. John, *Phys. Rev. Lett.* **83**, 967 (1999).

⁷K. Yoshino, Y. Shimoda, Y. Kawagishi, K. Nakayama, and M. Ozaki, *Appl. Phys. Lett.* **75**, 932 (1999).

⁸S. W. Leonard, J. P. Mondia, H. M. van Driel, O. Toader, S. John, K. Busch, A. Birner, U. Gösele, and V. Lehmann, *Phys. Rev. B* **61**, R2389 (2000).

⁹S. Kubo, Z.-Z. Gu, K. Takahashi, Y. Ohko, O. Sato, and A. Fujishima, *J. Am. Chem. Soc.* **124**, 10950 (2002).

¹⁰G. Mertens, T. Röder, H. Matthias, H. Marsmann, H.-S. Kitzerow, S. L. Schweizer, C. Jamois, R. B. Wehrspohn, and M. Neubert, *Appl. Phys. Lett.* **83**, 3036 (2003).

¹¹S. Gottardo, D. S. Wiersma, and W. L. Vos, *Physica B* **338**, 143 (2003).

¹²Ch. Schuller, F. Klopff, J. P. Reithmaier, M. Kamp, and A. Forchel, *Appl. Phys. Lett.* **82**, 2767 (2003).

¹³S. M. Weiss, H. Ouyang, J. Zhang, and P. M. Fauchet, *Opt. Express* **13**, 1090 (2005).

¹⁴S. F. Mingaleev, M. Schillinger, D. Hermann, and K. Busch, *Opt. Lett.* **29**, 2858 (2004).

¹⁵M. Loncar, A. Scherer, and Y. Qiu, *Appl. Phys. Lett.* **82**, 4648 (2003).

¹⁶J. Martz, L. Zuppiroli, and F. Nüesch, *Langmuir* **20**, 11428 (2004).

¹⁷R. Ferrini, D. Leuenberger, M. Mulot, M. Qiu, J. Moosburger, M. Kamp, A. Forchel, S. Anand, and R. Houdré, *IEEE J. Quantum Electron.* **38**, 786 (2002).

¹⁸J. Martz, B. Wild, R. Ferrini, L. A. Dunbar, M. Mulot, S. Anand, R. Houdré, and L. Zuppiroli, *Proc. SPIE* **5926**, 592601-1 (2005).

¹⁹M. Mulot, R. Ferrini, B. Wild, J. Moosburger, A. Forchel, R. Houdré, and S. Anand, *J. Vac. Sci. Technol. B* **22**, 707 (2004).

²⁰J. Li and S.-T. Wu, *J. Appl. Phys.* **95**, 896 (2004).

²¹M. Qiu, B. Jaskorzynska, M. Swillo, and H. Benisty, *Microwave Opt. Technol. Lett.* **34**, 387 (2002).