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Two-dimensional photonic crystals in antimony-based films fabricated by holography

M. Nalin,^{1,a)} J. W. Menezes,² L. Cescato,² E. S. Braga,³ H. Hernandez-Figueroa,³ S. J. L. Ribeiro,⁴ Y. Messaddeq,⁴ and M. Siu Li⁵

¹*Instituto de Física, MAV, São Paulo State University (UNESP), 17033-360 Bauru, São Paulo, Brazil*

²*Laboratório de Óptica, DFMC, Instituto de Física Gleb Wataghin, Universidade Estadual de Campinas (UNICAMP), 13083-970 Campinas, São Paulo, Brazil*

³*Faculdade de Engenharia Elétrica e Computação, Universidade Estadual de Campinas (UNICAMP), P.O. Box 6101, 13081-970 Campinas, São Paulo, Brazil*

⁴*Instituto de Química, LAMF, São Paulo State University (UNESP), 14801-970 Araraquara, São Paulo, Brazil*

⁵*Instituto de Física de São Carlos, USP P.O. Box 369, 13560-970 São Carlos, São Paulo, Brazil*

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In this work, we demonstrated the fabrication of two-dimensional (2D) photonic crystals layers (2D-PCLs) by combining holographic recording and the evaporation of antimony-based glasses. Such materials present high refractive indices that can be tuned from 1.8 to 2.4, depending on the film composition; thus, they are interesting dielectric materials for fabrication of 2D-PCLs. The good quality of the obtained samples allowed the measurement of their PC properties through the well-defined Fano resonances that appear in the transmittance spectrum measurements at different incidence angles. The experimental results are in good agreement with the calculated band diagram for the hexagonal asymmetric structure. © 2008 American Institute of Physics.

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Antimony contained glassy materials are promising for several photonic applications. Recently, interesting studies concerning the nonlinearity,¹⁻³ the optical,⁴ and the photochromic⁵ properties of this compound were reported in both glasses and films. Some of the interesting properties presented by these glasses are high refractive index (≈ 2), large nonlinear refractive index, n_2 large Kerr effect, low two photon absorption coefficients, large infrared transmittance, and high thermal stability. Such properties make antimony glasses useful for all-optical switching devices.

Photonic crystals are optical structures in which the light propagation is forbidden in determined region of the spectrum due to a strong periodic modulation of the refractive index.⁶ Two-dimensional photonic crystal layers (2D-PCLs) have been extensively fabricated using GaAs and Si semiconductors that can be considered high refractive index dielectric materials for light propagation in the near infrared region.^{7,8} 2D-PCLs have been studied for a large variety of applications such as control of spontaneous emission,^{9,10} plasmon assisted optics,^{11,12} and so on.

2D-PCLs have been achieved mainly by using an electron beam (EB) combined with dry etching^{13,14} and more recently by focused ion beam (FIB) lithography.^{15,16} Nevertheless, these techniques require expensive equipments. Holographic lithography (HL) is a less expensive method for fabrication of 2D-PCLs in large areas.^{17,18} Such method, however, generates generally many defects that carry difficulties in the measurement of the photonic properties of the sample.

In this letter, we fabricate 2D-PCLs using the combination of a high contrast HL, to generate a high aspect ratio photoresist template, the evaporation of antimony-based glasses, and the “lift-off” process of the template. The quality of the produced samples, is good enough to obtain the well-defined Fano resonances⁶ in the transmittance spectrum measurements that allow us to obtain the experimental band diagrams of the 2D PCLs.

The simplest way to obtain 2D-PCLs is a double exposure of two-beam interference patterns, with an appropriate rotation of the sample between the exposures. By rotating the sample of 90° or 60°, squared or hexagonal geometries can be obtained, respectively. Hexagonal lattices, however, are more appropriate for fabrication of 2D-PCL because they present photonic band gaps (PBGs) for a larger range of filling factors and refractive indices.⁶ The rotation of 60°, however, generates asymmetric (elliptical) column structures that reduce the range of filling factors for occurrence of PBGs.¹⁹ Dielectric layers with air holes are more convenient than dielectric columns for fabrication of 2D-PCL because they present higher effective refractive indices and can be self-sustained.

The experimental procedure concerning the 2D-PCL fabrication is represented in Fig. 1. In the first step, the positive photoresist (SC 1827 Rohn and Hass) film was deposited by spin coating technique. Using a rotation of 3000 rpm for 30 s, a film thickness of about 0.5 μm was achieved. The photoresist was then exposed twice (by rotating the sample 60° between each exposure) using the superimposition of two interference patterns generated by a holographic setup using the line $\lambda=458$ nm of a solid state laser. This setup warrants the repeatability and the high contrast of the fringes.²⁰ After exposure, the photoresist film was developed in AZ Developer diluted 1:4 during 40 s in order to obtain

^{a)}Author to whom correspondence should be addressed. Electronic mail: nalin@fc.unesp.br.

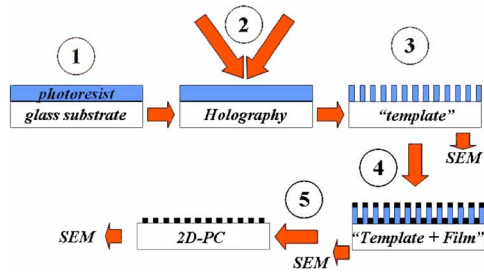


FIG. 1. (Color online) Schematic process of 2D-PCLs: (1) photoresist deposition by spin coating, (2) double exposure of two-beam interference patterns using $\lambda=457$ nm, (3) photoresist development, (4) antimony-based film deposition by EB-PVD, and (5) lift-off of the photoresist template.

the photoresist template. The resulting structures recorded in photoresist were analyzed by scanning electron microscopy (SEM) and are shown in Fig. 2(a). They are constituted of well-defined elliptical columns measuring around $0.5 \mu\text{m}$ in height and an average filling factor around 0.2.

Glasses with chemical composition $50\text{Sb}_2\text{O}_3-50\text{Sb}_2\text{S}_3$ (in mol %) were prepared following the process developed in Ref. 3. The next step consisted in the deposition of the antimony-based film on the template. The film was deposited on the hexagonal photoresist templates by EB physical vapor deposition (PVD) in vacuum (3×10^6 Torr) at room temperature using the glass sample as raw material. By using an EB gun operating at 5.75 kV and EB current at 6.0 mA, deposition rates of 5 nm s^{-1} were achieved. The template coated with the antimony film can be seen in the SEM photographs of Fig. 2(b). The antimony film deposition recovers more the top and the bottom of the template, keeping the vertical walls of the columns shadowed. This fact allows the further removal of the photoresist template by immersion of the sample in acetone (lift-off). The resulting antimony 2D-PCLs, with about 200 nm of thickness, can be seen in Fig. 2(c).

The refractive index value of the bulk glass target was measured ($n=2.1$). Due to the process of deposition, the refractive index of the homogeneous films, deposited at the

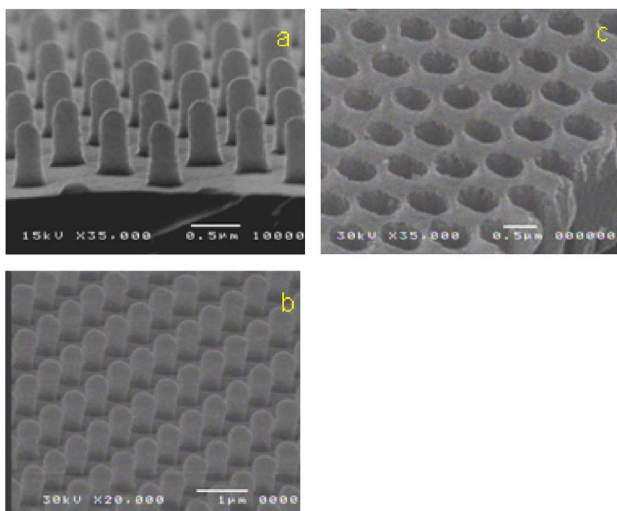


FIG. 2. (Color online) SEM photographs of the samples after each step of the process: (a) photoresist template on a glass substrate, (b) template deposited with antimony film, and (c) 2D-PCL antimony layer after lift-off of the photoresist template.

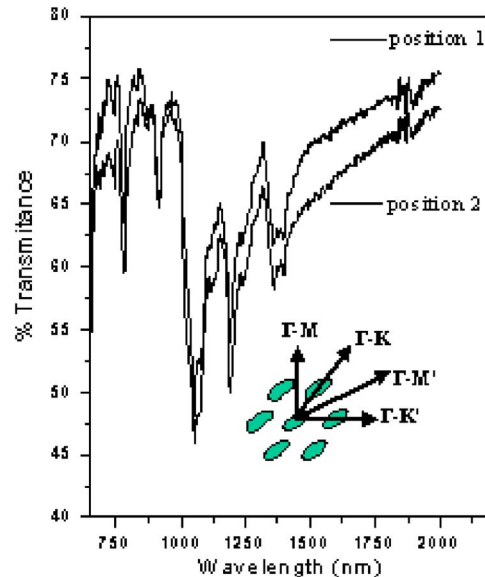


FIG. 3. (Color online) Transmission spectra for 2D-PCLs performed at two distinct regions of the sample.

same conditions on glass substrates, decreases to values between 1.9 and 2.0, for a film thickness around 350 nm.

The photonic layer was experimentally characterized by measuring the optical transmission^{21,22} spectrum as a function of incidence angle θ using a Cary 500 spectrometer, from Varian, in the range from 650 to 2000 nm and scan rate of 4 nm s^{-1} . A polarizer was placed before the sample to control the polarization of the incident light. The angle of incidence θ was defined with an angular resolution $\pm 1^\circ$ and it was changed from 0° to 60° . Measurements were done along the directions of the symmetry of the unit cell of the 2D-PCLs by selecting the correct angle in φ ,²³ for the TE and TM polarizations.

Figure 3 shows an example of the transmission spectra measurements of the 2D-PCLs along the direction $\Gamma-K$, for the TE polarization, for two different regions of the sample, separated of about 5 mm from each other. The transmittance curves display the resonant features (Fano resonances) when the frequency and the component of the wavevector parallel to the surface match a photonic mode in the layer. Note that the resonances peaks are as well defined as those obtained from samples fabricated by EB or FIB.¹⁵ This fact indicates that there is no substantial variation of the geometric parameters of the holes in the spot size area of the measurement ($\sim 3.5 \text{ mm}^2$).

Figure 4 shows the transmission spectra of the 2D-PCLs along the (a) $\Gamma-M$ and (b) $\Gamma-K$ directions. By changing the angle of incidence and the orientation of the sample, the dispersion of photonic modes in all directions of the Brillouin zone can be mapped.^{7,22} As it can be expected, the intensity of the resonant peaks decreases for higher angles. At low angles, the spectra were characterized by the presence of two main bands at around 818 and 1175 nm. For higher θ values, the spectra became more complex.

In order to verify the photonic properties of the fabricated 2D-PCLs, the dispersion of the photonic modes was theoretically calculated using the 2D finite element method.²³ In the calculus, we considered a hexagonal infinite array of

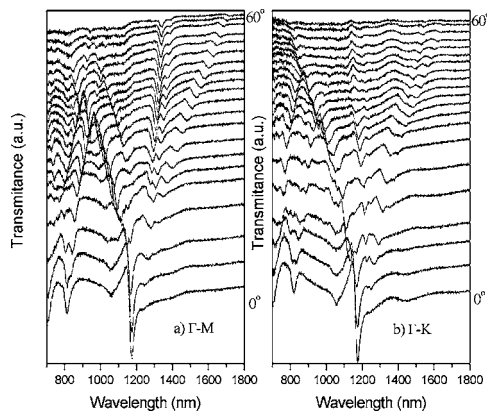


FIG. 4. Transmission spectra obtained for the 2D-PCL sample by changing the incidence angle. Measurements were done along the (a) Γ - M and (b) Γ - K orientations, both for TE polarization.

elliptical air columns engraved in a dielectric material with $R/a=0.217$ ($a=0.808$ nm) and $r/R=0.84$, where R is the semimajor axis and r is the semiminor axis of the elliptical section. These geometrical parameters were measured from SEM top view photographs of the sample used for the optical measurements. The refractive index value used in the simulations of the band diagram was $n=1.9$, which corresponds to the value that best fits the experimental points. This value is in good agreement with the expected range for refractive index of our samples (1.9–2.0). The calculated band diagrams of the structures are shown in Fig. 5, for the directions Γ - M and Γ - K . Such band diagrams were calculated for the TE polarization in which the electrical light field is parallel to the layer plane (perpendicular to the column axis). Note that to calculate the photonic band diagrams for the elliptical holes, it is necessary to add new directions to represent $\frac{1}{4}$ of the Brillouin zone¹⁹ (inset of Fig. 3).

From the energy positions of the Fano resonances observed in the transmittance measurements, the experimental points of the dispersion curves can be obtained.⁵ Such results are plotted in Fig. 5 together with the theoretical curves.

As it can be seen in Fig. 5, there is a good overall agreement between experimental and calculated spectra. The accordance is better for the lower bands. Theoretical and experimental dispersion curves obtained for the other

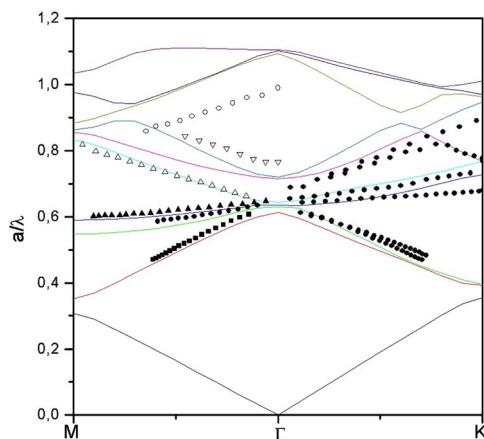


FIG. 5. (Color online) Comparison between the theoretical and experimental band diagrams obtained for the 2D-PCL sample.

symmetry directions, Γ - M' and Γ - K' , presented similar accordance, both for TE and TM polarizations.

Such results confirm the band structures presented by 2D-PCLs based on antimony fabricated by HL and lift-off process. Thus, from our knowledge, this is the first time that the Fano resonances are measured in 2D photonic crystals recorded by holography. Although other papers demonstrated the possibility to fabricate 2D-PCLs by using holography,²⁴ neither of them showed the Fano resonances.

Although the geometry of the holographically generated structures cannot be arbitrarily defined, the arising of a complete PBG can be achieved by choosing a proper refractive index of the film that can be obtained by changing the composition of the antimony glass. On the other hand, the tuning of the PBG can be easily performed, for any wavelength from the infrared to visible region, by changing the fringe period that is the best controllable parameter of a holographic setup.

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