Electrical tuning of three-dimensional photonic crystals using polymer dispersed liquid crystals

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Electrically tunable three-dimensional photonic crystals with a tunable wavelength range of over 70 nm of stop gaps between 3 and 4 μm have been generated in a liquid crystal-polymer composite. The photonic crystals were fabricated by femtosecond-laser direct writing of void channels in an inverse woodpile configuration with 20 layers providing an extinction of infrared light transmission of 70% in the stacking direction. Stable structures could be manufactured up to a liquid crystal concentration of 24%. Applying a direct voltage of several hundred volts in the stacking direction of the photonic crystal changes the alignment of the liquid crystal directors and hence the average refractive index of the structure. This mechanism permits the direct tuning of the photonic stop gap. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1861131]

For fast transmission and high-bandwidth information delivery, in the not too distant future optical systems may displace conventional electric transmission wires for computer networks, circuit tracks for chip to chip communications, and possibly electronic intrachip transmission and processing of data. As luminary analogues of semiconductors, photonic crystals[1] which are periodic dielectric structures with periodicity of the order of the wavelength of light promise the level of control over the propagation of light that is required for these new devices. Capable of inhibiting light propagation over a spectral region known as the photonic band gap, they can feature specific functional defects required for future electro-optic and all-optical devices.

One particularly interesting area is the development of tunable photonic crystals, which permit the properties of the band gap to be tuned either by changing the structural arrangement or the average refractive index. Structural changes resulting in a shift of the band gap position can be achieved, for example, by external compression of structures[2,3] and mechanochromic effects,[4] volume phase transition of hydrogels[5] or thermal expansion.[6] These structural changes can give rise to large changes in the position and/or characteristics of the stop band but are generally not suitable for real-world optical devices as their response time is exceedingly high. In contrast, refractive index changes have been accomplished by photochemical control,[5,7] the use of nonlinear materials,[8,9] thermal phase transitions[10] or on the use of electrically, optically, or magnetically sensitive materials.[11]

The introduction of liquid crystals into the system appears to offer the most promise as they exhibit large variations in refractive index depending upon the alignment state of the director (long molecular axis), which can be changed by the application of an electric field. Most easily, liquid crystal is infiltrated into a pregenerated structure such as the inverse opal.[12–14] However, this decreases the refractive index contrast and, therefore, reduces the effectiveness of the system.[15]

In contrast, we report in this Letter the use of polymer dispersions to introduce the liquid crystal into a host polymer. In this material, the liquid crystal forms into small droplets trapped inside the solidified polymer matrix. The droplet size can be varied in the manufacture process and can be generated from hundreds of microns to a few nanometers.[16–20] Polymer dispersed liquid crystals (PDLCs) have been applied, for example, to spatial light modulators,[21] variable focus lens systems,[22] and three-dimensional optical data storage.[23] Two- and three-dimensional photonic crystals have also been generated in PDLCs using holographic writing techniques.[19,24] However, the dielectric contrast between the lattice elements in this method is restricted to the difference between the polymer host and the liquid crystal. For the experiments described in this Letter the polymer material used consisted of urethane oligomers having C=C unsaturation, which were cross-linked by a thiol-ester oligomer (NOA63 optical adhesive, Norland Inc.; refractive index $n_{\mathrm{NOA}}=1.56$). The liquid crystal (E49, Merck) was a eutectic mixture containing 4-pentyl 4-cyano biphenyl. It has an extraordinary refractive index (n^{e}_{LC}) of 1.74 and an ordinary refractive index (n^{o}_{LC}) of 1.53.

Utilizing a multiphoton-induced void channel microfabrication technique,[25,26] which allows for the generation of arbitrary three-dimensional structures, we created photonic crystals inside a solidified block of the liquid crystal polymer. The ablated void channels create the required dielectric contrast against the polymer-liquid crystal background. The fabrication setup is similar to that we used before[25] except for a few modifications. A femtosecond-pulsed Ti:sapphire laser (Tsunami, Spectra Physics, repetition rate 82 MHz, pulse width 80 fs) operating at a wavelength of 700 nm was focused into the polymer mixture using an Olympus 60× numerical aperture (NA) 1.45 oil immersion objective. The samples were translated in the resting focus by a 200 ×200 μm piezoelectric nanopositioner combined with a 350 μm z-drive (both Physik Instrumente, 2 nm resolution, 10 nm repeatability). Exposure of the sample to the laser beam was controlled by a mechanical shutter. Scanners and shutter were fully software-controlled.

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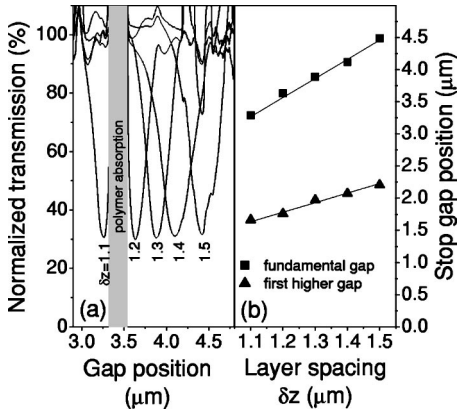

FIG. 1. (a) Infrared spectra showing the positions of the fundamental stop gap for a PDLC material with 6% E49 with the layer spacing δz incrementally increasing from 1.1 to 1.5 μm. (b) Stop gap positions of the fundamental and first higher-order gaps fitted to the Bragg condition.

The void channels were stacked into an inverse woodpile structure[25] with alternating layers of crisscrossed channels creating a face-centred tetragonal unit cell. Its stop gaps in the stacking direction satisfy the Bragg condition: $m\lambda_{\text{gap}} = 2\delta z n_{\text{avg}}$, where m is the order of the gap, δz is the layer spacing, and n_{avg} the average refractive index of the photonic crystal. This is demonstrated in Figs. 1(a) and 1(b) by five woodpile photonic crystal structures containing 6% E49, which were generated with in-plane spacings δx and δy of 1.57 μm and a variation of the layer spacing δz increasing from 1.1 μm to 1.5 μm in steps of 0.1 μm. The position of the fundamental and first higher-order stop gaps moves from 3.26 μm to 4.47μm and from 1.15 μm to 2.25 μm, respectively. Fitting these data by the Bragg condition yields $n_{\text{avg}} = 1.49$. The lower average refractive index compared to n_{NOA}, n^o_{LC}, and n^e_{LC} reflects both the material changes upon void generation[26] and the reduction of the refractive index values of the polymer[27] and the liquid crystal[28,29] in the mid-infrared. The band gaps were measured with a Nicolet Nexus Fourier transform infrared spectrometer with a Continuum infrared microscope to analyse the spectral throughput of the crystal structure in the stacking direction. The 32× NA 0.65 Reflechromat objective and condenser were corrected for glass slide and coverslip. Each spectrum consisted of 200 scans acquired with a resolution of 4 cm^{-1}.

To achieve electrical tuning, cells with transparent indium tin oxide (ITO) electrodes on either side were made [see Fig. 2(a)]. The ITO films grown to a thickness of approximately 35 nm by thermal vacuum deposition are effectively transparent to infrared and visible light and allow an electric potential to be applied to either side of the PDLC cell. No other insulating film was placed over the ITO coatings. The spacer used for the well structure was acrylic tape with a thickness of 140 μm. Wires were connected to the conductive coatings where the electrical potential could be applied.

The alignment of the liquid crystal directors by a voltage applied to the ITO electrodes is shown in Fig. 2(b). The prepolled state (no voltage applied) is approximated as a random liquid crystal director alignment with its long molecular axis freely positioned at any angle θ relative to the electric field of the probing light. Using the ellipsoidal relation

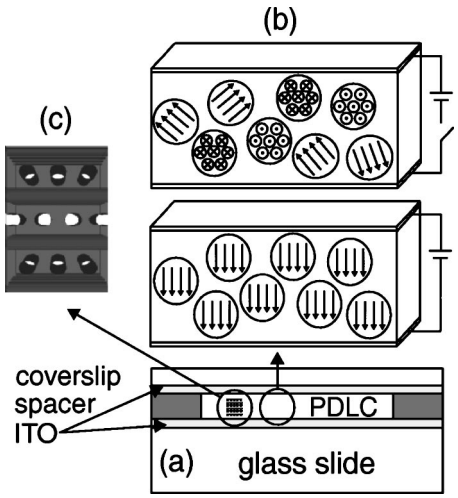

FIG. 2. (a) Schematic diagram of the cell structure. ITO coatings of glass slide and coverslip provide transparent electrodes for the application of a uniform electric field to the cell. (b) The liquid crystal alignment states within the cell. With no field applied (prepolled state, upper image) the liquid crystal directors are randomly aligned. Otherwise, they align parallel with the electric field (lower image). (c) Sketch of the PDLC void channel woodpile photonic crystal.

$$\left(\frac{1}{n_{\text{LC}}}\right)^2 = \left(\frac{\cos\theta}{n_e}\right)^2 + \left(\frac{\sin\theta}{n_o}\right)^2$$

results in an average refractive index of $n^{\text{prepolled}}_{\text{LC}} = 1.60$. As E49 has a positive dielectric anisotropy, the directors align parallel to the applied electric field reducing the refractive index of the liquid crystals to $n^{\text{polled}}_{\text{LC}} = n^o_{\text{LC}}$ for light propagation in the stacking direction. This controlled refractive index variation tunes the position of the band gap to shorter wavelengths by operating on the average refractive index n_{avg} of the composite material. Various woodpile void-channel photonic crystals were fabricated within the sandwiched structure [Fig. 2(c)].

To assess the maximum quantity of liquid crystal allowable for efficient void channel generation, four cells were manufactured with concentrations of liquid crystal of 0%, 10%, 24%, and 30%. Cells with higher concentrations were increasingly unstable with consistent void channel formation impossible at 30% and higher. The degree of gap suppression as a function of number of layers is shown in Fig. 3. As the amount of liquid crystal increases, the effects of scattering of

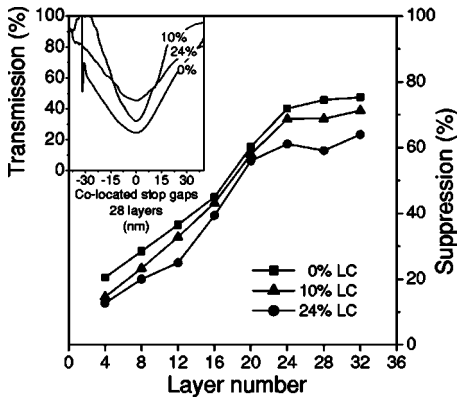

FIG. 3. Degree of stop gap suppression as a function of the number of layers for three liquid crystal concentrations. Inset: Corresponding infrared spectra of the 28-layer structures.

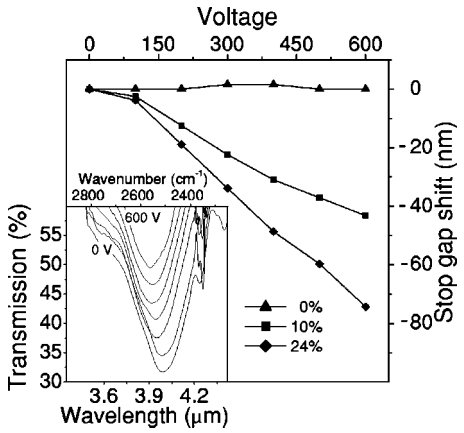

FIG. 4. Shifts in the stop gap position with the applied voltage for PDLC cells with 0%, 10%, and 24% liquid crystal. The reference cell without liquid crystal shows no stop gap movement. Note also the Freedericksz threshold of 100 V. Inset: FTIR spectra demonstrate the gap shift to shorter wavelengths as a function of the direct voltage (0–600 V in steps of 100 V) across a cell with 24% liquid crystals.

the writing beam (inhibits delivery of writing power) and inhomogeneities of the material (microchannels start to fill with liquid crystal) result in a plateauing of the suppression values.

To characterize these active photonic crystals, an increasing potential up to 600 V was applied to each cell and the infrared transmission spectrum of the crystal was recorded. As seen in Fig. 4, there is little change in the spectral position of the band gap until a threshold voltage of approximately 100 V is reached where there is a distinct variation as more liquid crystals align with the applied voltage.[18] This initial lag is attributed to the Freedericksz transition[30] where the torque from the applied electric field has to deform the current director configuration and commence an abrupt orientational onset. After the onset, a roughly linear increase in the band gap position is observed with the increasing electrical potential. We expect that this rise will plateau (sometime after 600 V) as the maximum number of liquid crystal molecules participate in the alignment process. At a potential of 600 V, a maximum deflection of the band gap of 74 nm is observed for the 24% sample. The 74 nm spectral shift is larger than that one may expect from an initial randomly oriented liquid crystal state. However, this result may be expected when initial partial in-plane alignment of the liquid crystals by the E-field of the writing laser beam may occur during the manufacture process.[23] The first higher-order stop gaps also exhibits a controlled variation with the applied potential.

In conclusion, three-dimensional photonic crystals have been generated in polymer dispersed liquid crystals which permit tuning of the spectral position of the band gaps between 3 and 4 μm via the application of an electrical field. A midgap wavelength shift of 74 nm has been demonstrated with the application of 600 V to the cell. The band gap shift reflects the refractive index variation of the liquid crystals as a result of the realignment of its directors in the external electric field. The multiphoton manufacture process allows the generation of arbitrary structures in a controlled fashion with band gaps being observed with as little as 4 layers. This type of electrically tunable photonic crystal may serve as a switch in an integrated optical circuit.

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