

Post-Process Tunability of Folded One-Dimensional All-Polymer Photonic Crystal Microcavity Lasers

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We report on wavelength tunability of all-polymer one-dimensional photonic crystal lasers after fabrication via a melt-process co-extrusion technique. Wavelength tuning of two types of lasers is discussed; (1) an elastomeric Distributed Bragg Reflector (DBR) laser with an active layer between multilayers of alternating elastomeric polymer layers that is mechanically tuned via an induced strain, and (2) a Distributed Feedback (DFB) laser with alternating dye-doped and undoped layers that is tuned by terracing the surface layer that becomes the center layer upon folding to create a defect in regularity of the DFB laser structure. Both of the lasers were doped with rhodamine 6g in the active volume and we report on both the range of tunability and lasing characteristics as well as the effects of random thickness variations for folded multilayer structures.

1. INTRODUCTION

Multilayer all-polymer Distributed Bragg Reflector (DBR) lasers and Distributed Feedback (DFB) lasers were made by a co-extrusion melt process technique. The co-extrusion melt process allows rapid, large-volume low-cost, solvent-free production of reflecting multilayer structures, which allows for roll-to-roll mass production. The resulting polymer microcavity lasers are

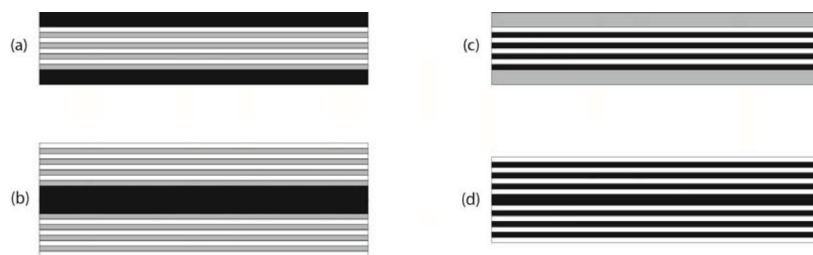


FIGURE 1

Structures of (a) the DBR directly after the extrusion process, (b) the DBR laser after folding and removing the outer skin layers, (c) the DFB laser after the extrusion process, and (d) one version of the defect DFB laser after removing the skin layers and then folding. The gain layers are shown in the darkest color although the defect DFB shown in (d) can be folded on either the doped or undoped side.

low-cost and can be laminated onto diode pump lasers or optical circuits. This makes for the possibility of a variety of applications including sensing and optical data processing. The co-extrusion melt process method used to create these lasers is described in earlier reports of DBR and DFB multilayer laser structures.[1,2,3,4,5]

After the films have been extruded, they are manipulated to create the tunable DBR elastomer and folded defect DFB lasers, where the fold is held together by static cling. For the elastomeric DBR laser, the active dye-doped layer is extruded as one of the skin layers of the alternating polymer Bragg reflecting quarter-wavelength stack. The laser is then made by folding the multilayer film onto the dye-doped skin layer. The defect DFB laser, on the other hand, is made by including the laser dye in alternate layers throughout the quarter-wavelength stack and folding the film to create a central half-wavelength defect layer. Note that folding on either side of the DFB laser will create a defect in the reflection band. The DBR and defect DFB lasers are shown in Fig. 1 before and after folding.

2. POST-PROCESS TUNABILITY OF THE ELASTOMER DBR LASERS

The elastomer DBR mirrors are composed of a multilayer system of alternating 128 layers of a fluoroelastomer terpolymer of vinylidene fluoride, hexafluoropropylene, and tetrafluoroethylene (THV, refractive index $n \approx 1.36$), and ethylene-octene (EO, $n \approx 1.48$). The elastomeric multilayers are extruded with a rhodamine 6g (R6G) dye-doped Lotader elastomeric skin layer, which acts as the thick active central gain medium when the films are folded to create the microcavity Bragg laser. The Lotader skin layer is a blend of ethylene terpolymer with 40% by weight acrylic ester and glycidyl methacrylate [Arkema,

www.lotader.com]. The average layer thickness of the one-dimensional PhC is approximately 110nm and the center layer thickness is approximately 30μm after folding. A complete description of the elastomer lasers can be found in Ref. [5].

Tunability was achieved in the elastomer laser by mechanically stretching the laser in the direction perpendicular to the stacking dimension. Initially, the films were stretched to 150% of their original length to study the relationship between the photonic bandgap and the strain. At a stretching ratio of 150% (strain of 50%), the center of the reflection band shifted to ~400nm from its original spectral location of ~625nm. Not only is the thickness reduced by stretching the PhC, but induced-birefringence also occurs in both the PhC elastomeric multilayers and the highly polar R6G dye-doped skin layers. A pseudo-affine model was used to model the shift of the reflection band center wavelength, λ , as a function of the lateral strain, ϵ . [6] The photonic band peak wavelength due to birefringence can be written in terms of the initial peak wavelength, λ_0 , such that

$$\frac{\lambda}{\lambda_0} = \frac{n_1 + n_2}{(n_1^{(0)} + n_2^{(0)})\sqrt{1 + \epsilon}}$$

where the stretching strain perpendicular to the direction of the PhC dimension is simply, $\epsilon = \Delta l / l_0$, wherein $\Delta l = l - l_0$ with l and l_0 denoting the length and initial length, respectively. Here, $n_1^{(0)}$ and $n_2^{(0)}$ denote the initial refractive indices of the direction of stretching and the direction perpendicular to stretching, where n_1 and n_2 are the final indices of refraction. At small strains, $\leq 20\%$, the birefringence is negligible, where the difference between the refractive indices is $\Delta n \leq 0.004$. In this case, linearly polarized light in any direction, regardless of the relative direction of induced strain, should give us the same result within the experimental uncertainty. With this knowledge, we only treat the change in photonic band gap as a function of the induced lateral strain in a single direction perpendicular to the PhC dimension.

We find that even at small strains, $\epsilon \leq 20\%$, the wavelength tunability is large due to the shift in the photonic band gap. The relative height of the peak wavelength emission spectra as a function of the wavelength is shown in Fig. 2 for a typical tuning experiment of the elastomer lasers. Note that the wavelength with maximum power output is a function of the gain profile and the macroscopic geometry. Pumped by a 7ns pulsed 532nm source, the lasing efficiencies of these DBR lasers were approximately 0.8% with thresholds of 0.9mJ/cm² at an output wavelength of 600nm. The tunable emission spectrum for these samples under small strains spanned the region of ~570nm to ~625nm.[5]

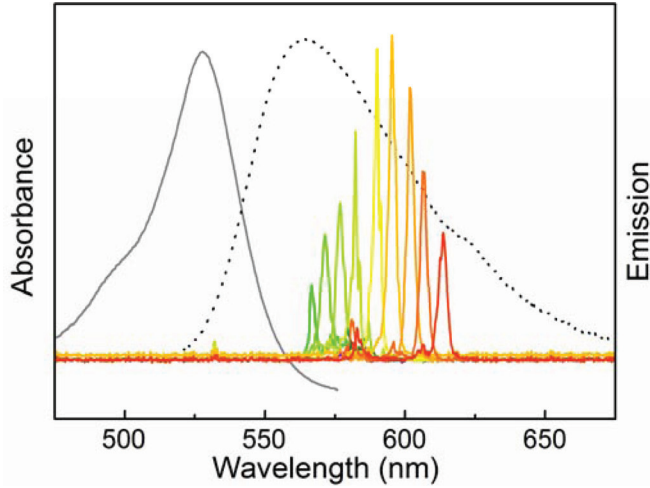


FIGURE 2

The series of sharp curves are the emission spectra at different stretching ratios for the DBR laser film. The solid gray curve to the right is the absorption spectrum and the dotted line is the fluorescence spectrum.

3. POST-PROCESS TUNABILITY OF THE DFB LASERS

The second multilayer tunable laser is constructed using a defect DFB structure. Unlike in the elastomer lasers for which the active layer is the center-folded skin layer, the active layers in the DFB lasers are co-extruded as every other layer of the alternating multilayer PhC. The multilayer DFB lasers consist of alternating layers of THV and a 25% by weight acrylonitrile in polystyrene blend (SAN25, $n \approx 1.57$) with R6G dissolved into the SAN25 layer at a nominal concentration of 1% by weight. The DFB lasers are initially extruded with a low density polyethylene (LDPE) skin layer on each side, and additional information regarding the co-extruded multilayer DFB laser can be found in Ref. [7]. The skin layers are removed before the post-processing steps in which just the multilayers are either folded to create a defect layer or cut and stacked to create a DFB laser with twice the number of initially co-extruded layers.

Folding onto the same refractive index material creates a $\lambda_{center}/2n_{fold}$ thick layer in the center of a multilayer DFB with approximately $\lambda_{center}/2n_{avg}$ periodic layer thicknesses, where λ_{center} defines the nominal center of the band gap, where n_{fold} is the index of refraction of the center layer and n_{avg} is the average index of refraction of the stack. The width of the reflection band is a function of the number of layers and the refractive index difference Δn . An increase in transmission at the center of the photonic band gap occurs as a result of folding and binding together two layers of the same refractive

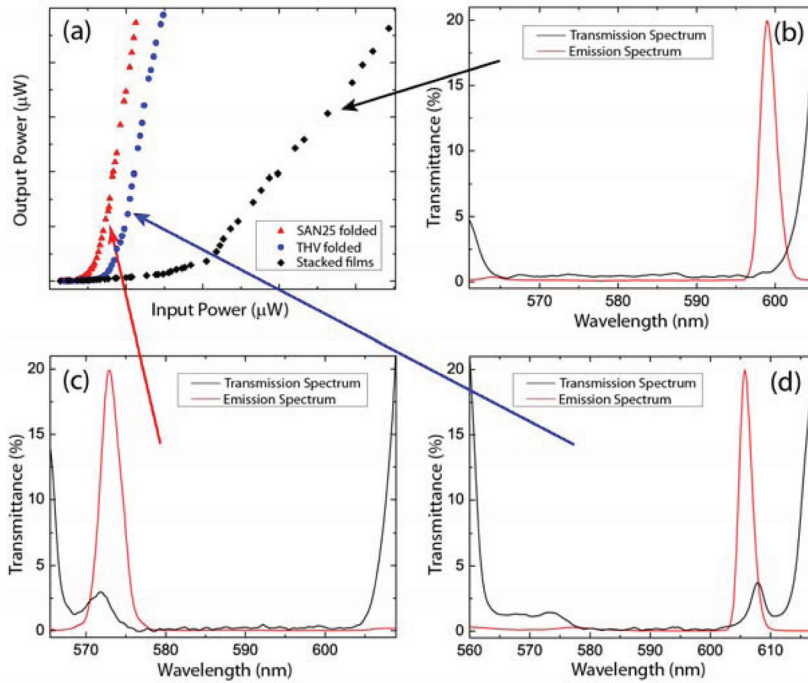


FIGURE 3

(a) The conversion efficiency curves of defect DFB films created by folding onto the THV side and SAN25 side to create a defect DFB, and two 64-layer films stacked to create a 128-layer DFB. The transmission and emission spectra of the (b) stacked, (c) SAN25 folded, and (d) THV folded films with arrows that point to their respective conversion efficiency curve.

index. This “impurity band” is caused by explicitly breaking the discrete translation symmetry,[8] where the position is a function of the phase accumulation.[9] The binding at the folded halves was achieved by adhering static charged interfaces together, where the adhering force is greatest immediately after removing the LDPE skin. To assess the efficiency of the folded “defect” structure, we also created stacked DFB structures of the same total number of layers, by adhering two films in the same orientation. A typical set of efficiency curves is shown in Fig. 3(a) with the corresponding transmission and emission curves shown in Figs. 3(b)–(d) for the respective stacked 64-layer film to create (b) a 128-layer DFB laser without a defect, (c) a 64-layer film folded on SAN25 to make a 128-layer high index defect DFB laser, and (d) a 64-layer film folded on THV to make a 128-layer low index defect DFB laser. Lasing efficiencies up to $10 \pm 1\%$ were recorded for defect DFB lasers and up to $3.0 \pm 0.3\%$ for the stacked films. On average, the lasing efficiencies of the defect films were between 3 and 6 times higher than the corresponding stacked films. Furthermore, the threshold values were as low as 110 kW/cm^2 for the SAN25 and THV defect lasers and 240 kW/cm^2 for the stacked lasers.

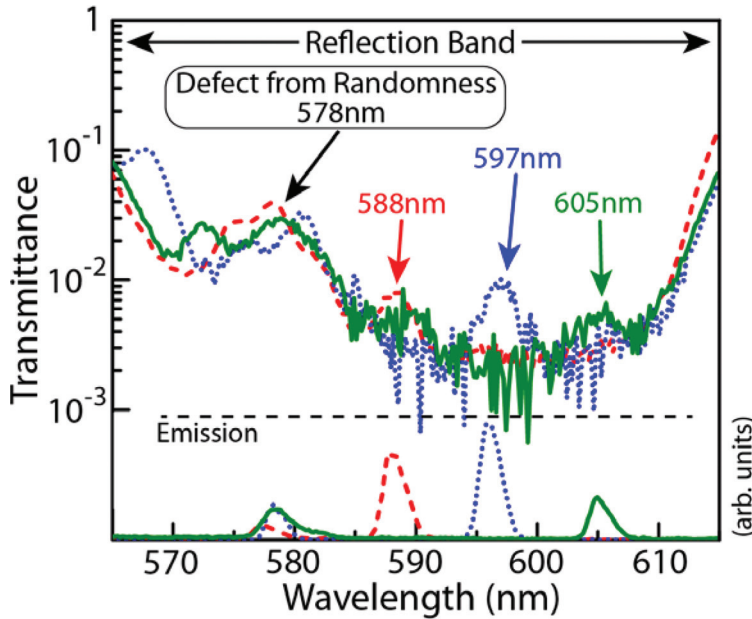


FIGURE 4

Transmission curve (log scale) of 128-layer terraced-defect DFB laser film at three different center thicknesses and the corresponding lasing spectra at the bottom.

In addition to the benefits of increased lasing efficiency and reduced thresholds from center layer defects, the wavelength of the band defect can be moved by terracing the surface layer that becomes the center defect layer upon folding. The lasing wavelength can then be tuned through terracing because the stimulated output wavelength follows the defect position in the photonic band gap due to the low group velocity at the defect wavelength. [10] The defect position within the reflection band shown in Fig. 4 is moved from the long wavelength edge to the short wavelength edge of the band by terracing the center layer to three different thicknesses. Additional details on this process can be found in Ref. [11].

4. SENSITIVITY OF TUNABLE DFB LASERS TO LAYER THICKNESS VARIATIONS

A broad range of available lasing wavelengths appear in these R6G defect DFB lasers, limited only by the gain spectrum and movement of the defect position within the reflection band. Variations in the average layer thickness of a multilayer PhC during the extrusion process affects the locations of the photonic band gap and the band defect. Thus, in addition to folding a DFB

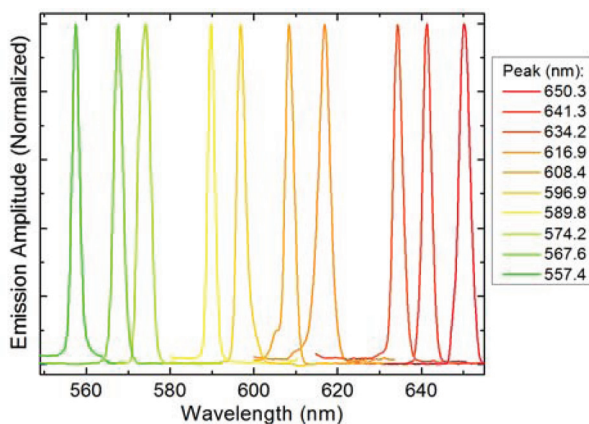


FIGURE 5

The normalized emission spectra at different locations of a defect DFB film that displays lasing wavelengths over a large spectral range.

laser to create a defect DFB laser, one can adjust the lasing wavelength by changing the pump location over the film surface. Here, we take advantage of the change in the reflection band spectral location due to the changes in the average layer thickness, which, for small layer-to-layer thickness deviations, corresponds to a change in the defect lasing location. By using this method, we have found some R6G-doped SAN25 DFB lasers with bulk layer thickness gradients that show tunable lasing from ~ 550 nm to ~ 650 nm by scanning across a 3 cm region with varying defect locations. Fig. 5 shows the normalized emission spectra at a few locations over a 128-layer defect DFB made from folding a 64-layer DFB on the R6G dye-doped SAN25 layer.

Due to the extrusion process, individual layer thicknesses vary during the extrusion and multiplying process. For the remainder of this section we study the effects of layer-to-layer thickness variations while holding the average layer thickness constant. Here, we study (1) the uncertainty in defect location as a function of the degree of layer-to-layer thickness variations and (2) the ability to readjust a defect's spectral location that is not in the predicted location due to random deviations of individual layer thicknesses throughout the multilayer.

The predictability of the lasing wavelength in a folded-defect DFB laser depends on the precision of the layer-to-layer thicknesses. We assume a Gaussian distribution of layer thicknesses. The Gaussian distribution is assumed purely for computational simplicity and introduces an uncertainty in the layer-to-layer thickness distribution that can be altered by a single parameter, though past studies have shown that the layer-to-layer thickness variations are not necessarily Gaussian.[1,5] Because the transmission of an electric field propagating through the multilayer can be written as the product

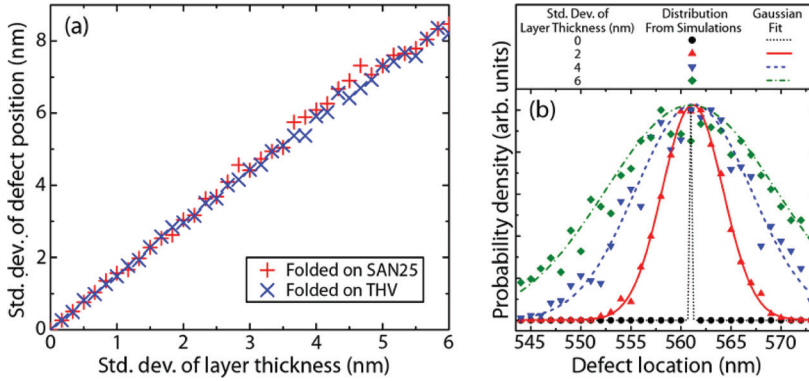


FIGURE 6

(a) The standard deviation of the defect location as a function of the standard deviation in layer thickness for a 128-layer defect DFB laser. (b) The form of the probability density as a function of defect location for a defect DFB created by folding a 64-layer film on the SAN25 layer.

of the linear transfer matrices of the individual layers,[12,13] we expect the deviation of the defect location to be linearly dependent on the standard deviation of layer thicknesses for small uncertainties that can retain a resultant Gaussian distribution. To study this effect, the standard deviation of the fold-defect location as a function of the standard deviation of layer thicknesses is shown in Fig. 6(a).

As previously mentioned, the standard deviation in layer thicknesses was studied for layered systems with an average layer thickness of 95nm. The uncertainty was studied from 0nm (perfect system) up to a 6nm standard deviation in increments of 1/6 of a nanometer standard deviation. Each increment was simulated for 1000 iterations to find the mean and standard deviation of the reflection band defect location. These simulations were carried out on a numerically constructed 64-layer THV/SAN25 PhC to create a 128-layer defect PhC. Each iteration was assigned a new set of layer thicknesses by a random number generator whose output was a normal distribution with a mean of 95nm. Both THV folded and SAN25 folded films were studied and showed the same fundamental results (see Fig. 6(a)). The standard deviations of defect location are linearly dependent on the layer thickness deviation for these small layer-to-layer thickness deviations and show an approximate $145\% \pm 5\%$ slope dependence. The average optical depth per layer is simply the average refractive index, $n_{ave} \approx 1.465$, multiplied by the layer thickness. If we treat the deviation in average optical depth per layer as opposed to the deviation in the average thickness per layer, then we find that the slope dependence is $99\% \pm 3\%$. In other words, we have an approximate one-to-one correlation, for relatively small uncertainties, between the deviation in optical depth and the deviation in the spectral location of the defect.

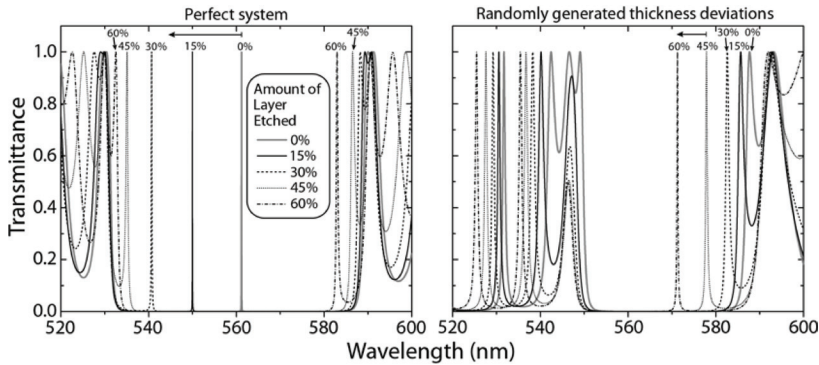


FIGURE 7

Simulations that show a terraced defect layer created by folding a 64-layer film onto the SAN25 side and terracing the center layer. The defect moves toward the short wavelength band edge as the amount of etching is increased. The defect in a perfect system starts near the center of the band while the defect due to randomness in layer-to-layer thickness was initially shifted to the long wavelength edge. After a significant amount of terracing, we see the defect reappear at the long wavelength band edge of the perfect system.

Fig. 6(b) shows the probability distribution of the calculated defect position in the reflection band normalized to unit amplitude for selected widths of the Gaussian distributions of layer thicknesses. We see that the edges of the probability distribution of the spectral defect location are already nearing the edges of the reflection band for 6nm layer thickness deviations. The band edges in a perfect system are shown in the left graph of Fig. 7, where the left and right edges are approximately 531nm and 589nm, respectively. The main reason for keeping the deviations small is due to the occurrence of band edge deformations in the presence of layer-to-layer thickness variations. The right graph in Fig. 7 shows large band edge deformations for a 10nm standard deviation from a 95nm mean in the individual layer thicknesses. Furthermore, these large deviations can cause multiple defects in the system in addition to multiple reflection bands. Thus, by keeping the analysis illustrated in Fig. 6 confined to small deviations in layer thickness (≤ 6 nm) we can neglect the possibility of interference from band edge deformations and multiple defect locations.

The band defect location may not appear at the desired location within the reflection band when layer thickness deviations are large. For a system in which that happens, one can still move the defect location to the desired location within the reflection band by terracing the center layer. Fig. 7 graphs the transmission spectra of simulated 64-layer film folded onto the SAN25 side to make a 128-layer defect DFB laser for a perfect system and a system with a 10nm standard deviation in layer thickness with a mean layer thickness of 95nm. Similar to the experiment shown in Fig. 4, the defect (found on the long wavelength edge of the reflection band in this case) is moved in the short

wavelength direction by terracing the center layer as shown by the arrows. The distance traveled by the defect depends on the amount of material removed, where both THV and SAN25 defect locations are similarly displaced. Note that if the amount of material removed displaces the defect beyond the short wavelength band edge, then a new defect appears at the long wavelength band edge. This is illustrated in Fig. 7(a), where an additional defect appears at the right edge of the reflection band at a 45% etch in the SAN25 layer, which is a doubled defect system. Also, note that the model shown in Fig. 7(a) does not reflect a perfect quarter wavelength system at a 50% etch because all layers are not a quarter wavelength in optical thickness due to every layer being 95nm thick regardless of the material (high or low relative index).

5. CONCLUSIONS

A co-extrusion melt process has been used to fabricate two types of polymeric multilayer dye lasers, which are each folded to create DBR and defect DFB lasers that have been observed to possess enhanced lasing efficiencies and lower thresholds. Furthermore, post-process tunability of the wavelength has been shown by mechanical deformations in the elastomer DFB lasers and terracing of the center fold layer of the defect lasers. Surface scans of laser films with high variability showed a large range in lasing wavelengths of the defect DFB lasers. Moreover, we studied how the layer thickness variability in the melt process of these films changes the location but not the fundamental dispersive character of the in-band defects. Thus, we have shown in practice that the post-processing steps of folding and terracing the center layer width can counteract the shift due to uncertainty in the spectral location of these in-band defects, effectively stabilizing the lasing behavior as compared with the unfolded films.

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