Appl. Phys. B 82, 539–541 (2006)
DOI: 10.1007/s00340-005-2120-y

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Lasing action in two-dimensional organic photonic crystal lasers with hexagonal symmetry

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Received: 15 November 2005
Published online: 25 January 2006 •

ABSTRACT We investigate fluorescence and lasing action from strongly modulated two-dimensional surface relief structures with hexagonal symmetry onto which thin films of optically active organic material have been deposited. As compared with second-order laser structures with square symmetry, these organic photonic crystal lasers exhibit unusual feedback mechanisms. As a result, we observe surface-emitting lasing action with a central beam normal to the surface and a hexagonal emission pattern of side-beams whose direction slightly deviates from the normal. A corresponding theoretical analysis allows us to determine the photonic bandstructure and the low-threshold laser modes in this system. These results agree very well with fluorescence data and confirm the hexagonal lasing pattern and the corresponding emission angles.

PACS 42.55.Tv; 42.70.Jk; 42.70.Qs

Photonic crystal structures provide the basis for novel laser sources with distinct properties. A single cavity embedded in a photonic crystal and equipped with an active medium represents the laser configuration with the smallest possible modal volume [1]. Equally fascinating is the realization of photonic crystal lasers through higher-dimensional distributed feedback. The strong feedback mechanisms in these systems may lead to low-threshold behavior [3–6], and the corresponding non-Markovian radiation dynamics near a photonic band edge may lead to unusual emission spectra and photon statistics [2].

Owing to their broad gain spectrum and easy processability as well as associated low-cost and large-area production technologies, organic thin films deposited on replicated plastic substrates represent promising candidates for the latter class of photonic crystal lasers. Square [5, 6] and hexagonal [3] lattice geometries have been investigated. In this letter we report on the fabrication and characterization of surface-emitting, i.e., second-order distributed feedback, two-dimensional organic photonic crystal lasers based on lattices with hexagonal symmetry. We fabricate micro-structured surface relief structures that serve as substrates for thin films of organic gain media through interference lithography. The beam of an Ar^+-laser operating at 363.8 nm is split into two separate beams. After being spatially filtered and expanded, these beams are superimposed onto a film of UV-sensitive photoresist so that in the subsequent development step, the resulting sinusoidal intensity profile is transferred into a surface relief structure. Through the angle of incidence θ of the laser beams, we can vary the periodicity $\Lambda = \lambda/2 \sin\theta$ from about 100 μm down to 200 nm. At Fraunhofer ISE, micro structures on areas as large as one square meter can be fabricated.

Substrates with hexagonal symmetry can be generated by three consecutive exposures, where the sample is rotated by 60° after the first and second exposure step. As a result, the form factor (shape) of the unit cell depends on the phase relation of the third exposure relative to the first and second exposure which is outside experimental control in our current setup. Figure 1 shows SEM micrographs of the resulting structures in photoresist for two extreme cases: the maxima lines (a) or minima lines (b) of all three exposures intersect. Due to nonlinearities of the development process and the photoresist, the sinusoidal interference pattern is distorted during transfer into the photoresist. The actual profile can, in principle, be determined from SEM and AFM measurements of the actual structure. For the structures shown in Fig. 1, we approximate the corresponding surface relief profile through

$$d_{\mathrm{hex},i} = d_{\mathrm{mod}} \left(\sin\left(\left(2\pi x + \frac{2\pi y}{\sqrt{3}} + a_i\pi \right) / \Lambda \right) + \sin\left(\left(\frac{4\pi y}{\sqrt{3}} + a_i\pi \right) / \Lambda \right) + \sin\left(\left(-2\pi x + \frac{2\pi y}{\sqrt{3}} + a_i\pi \right) / \Lambda \right) + b_i \right) . \quad (1)$$

where $a_1 = 3/2$, $b_1 = 3$, $a_2 = 1/2$, $b_2 = 3/2$ and the indices 1 and 2, respectively, refer to structures a and b in Fig. 1. In what follows, we restrict ourselves to structures of type (a). Onto this substrate (index of refraction $n_{\mathrm{sub}} \approx 1.5$), we deposit through spin-coating a thin film of MeLPPP (methyl-substituted ladder-type poly(para-

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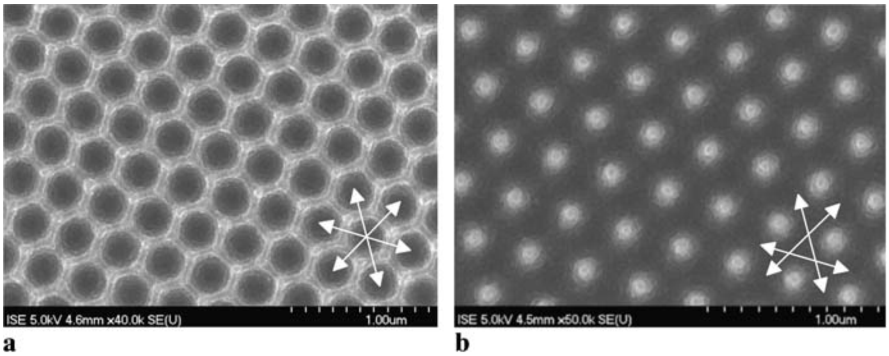

FIGURE 1 SEM-micrographs of different hexagonal relief structures in photoresist, resulting from different phase relations of the exposures

phenylene)), a π-conjugated polymer with attractive features such as high luminescence quantum yield in the solid state (about 30% [7]) which is employed in blue light emitting diodes and lasers. The resulting organic photonic crystal laser structures exhibit photonic bandstructures for the quasi-guided modes of the waveguiding MeLPPP layer. This layer is characterized through a spatially varying thickness $d_{\mathrm{MeLPPP}}(x, y) = d_0 + d_{\mathrm{hex},1}(x, y)$ ($d_0 = 150$ nm and $d_{\mathrm{mod}} = 300$ nm; see (1)) and an index of refraction $n_{\mathrm{MeLPPP}} \approx 1.7$. The corresponding bandstructure within the frequency interval where MeLPPP exhibits appreciable luminescence may be calculated from fully vectorial reflection calculations [8, 9]. For a given angle of incidence and polarization, we determine the quasi-guided modes (label α) as peaks in the reflection spectrum and the spectral width of these peaks allows us to assign an out-of-plane loss rate, $\kappa_\alpha^\perp$ to these modes. The TM-like polarized modes exhibit a higher damping due to weaker confinement and a larger overlap with the substrate. Therefore, we display in Fig. 2 only the bandstructure for TE-like quasi-guided modes and their associated out-of-plane losses. As demonstrated in Fig. 3, the results of these bandstructure calculations agree qualitatively very well with angle-resolved photo-luminescence measurements from our sample.

In order to understand lasing behavior in these systems, we employ a semiclassical theory of lasing action in photonic crystals [10]. Within this theory, the lasing threshold for a given mode α (with frequency ω_α) is determined by requiring that the effective unsaturated gain coefficient

$$\mathcal{g}_\alpha = g_\alpha \frac{4\pi |\boldsymbol{d}_{12}|^2 \omega_\alpha R}{\hbar \gamma_\perp \gamma_\parallel} \tag{2}$$

equals the total losses

$$\kappa_\alpha = \kappa_\alpha^{\mathrm{mat}} + 2\left(\kappa_\alpha^\perp + \kappa_\alpha^\parallel\right) . \tag{3}$$

The effective gain enhancement $g_\alpha = \int d^2r |\boldsymbol{E}_\alpha|^2 v(\boldsymbol{r})$ describes how the eigenmode $\boldsymbol{E}_\alpha(\boldsymbol{r})$ samples the distribution $v(\boldsymbol{r})$ of active material within the unit cell. The dipole dephasing and decay rate of the electric dipole moment $\boldsymbol{d}_{12}$ of

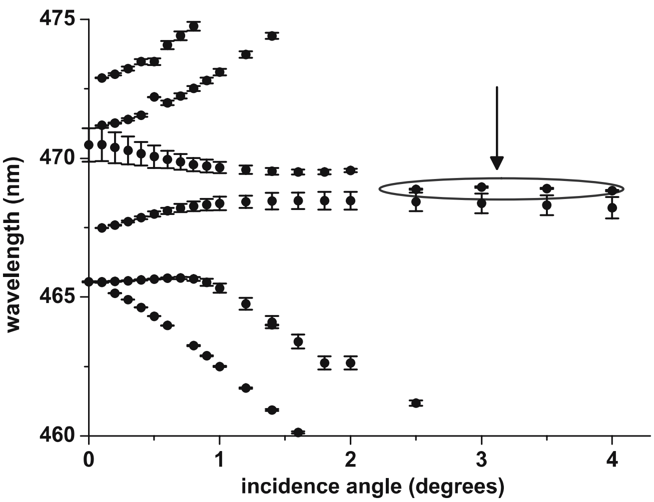

FIGURE 2 Bandstructure along the ΓK-direction for for an organic PC laser structure with hexagonal symmetry determined through reflectivity calculations ($d_0 = 150$ nm, $d_{\mathrm{mod}} = 300$ nm). The FWHM-width of the observed resonances is indicated by error bars that have been enlarged by a factor of four for better visibility. There are several modes with low in-plane losses (i.e., low group velocities) as well as low out-of-plane losses (i.e., low FWHM-widths of the resonances) at the Γ-point. In addition, we observe one mode also meeting these criteria at an angle between 2.5° and 4°, as indicated by the *arrow*. This mode can be identified as the laser mode responsible for the emission at a small angle (cf. Fig. 4)

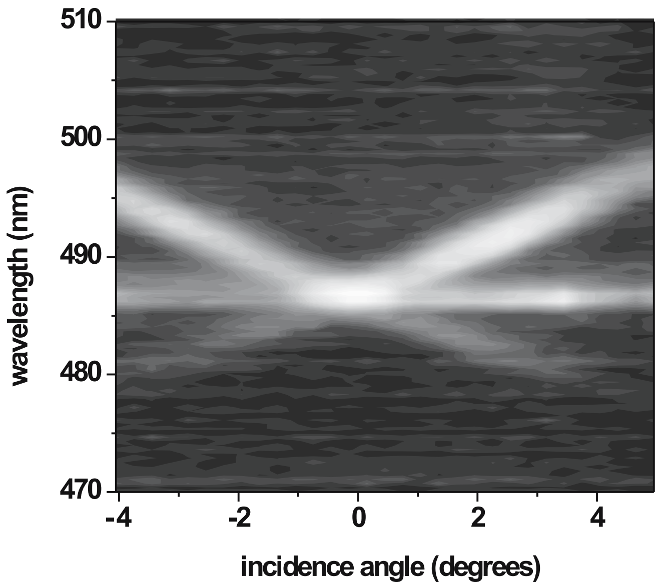

FIGURE 3 Angle-resolved PL-measurement along the ΓK-direction for a laser based on a substrate with hexagonal symmetry (see Fig. 1). These data agree well with the corresponding calculated bandstructure (cf. Fig. 2). The deviation in wavelength of approx. 15–20 nm could be reduced by a more sophisticated modelling of the surface relief structure, albeit without providing further physical insight

the corresponding lasing transition are denoted by $\gamma_\perp$ and $\gamma_\|$, respectively. In addition, R represents the incoherent pump rate at which molecules are pumped from the lower to the upper state of this transition. Finally, $\kappa_\alpha^{\text{mat}}$ and $\kappa_\alpha^\|$, respectively, represent effective material and in-plane losses of the mode α.

Our numerical evaluations show that the effective unsaturated gain coefficient for the modes displayed in Fig. 2 differ by at most 10%. In view of the substantial variation of the out-of-plane losses (see Fig. 2), we conclude that the laser modes with the lowest threshold are determined through minimal total losses. If we neglect material losses (or assume them to vary in a fashion similar to the gain coefficients), the total losses κ_α consist of both, in-plane losses $\kappa_\alpha^\|$ and the out-of-plane losses $\kappa_\alpha^\perp$. For a finite pump region of the active material, the in-plane losses are determined by the rate at which energy leaves the pump region, i.e., via the flux of the Poynting-vector associated with mode α through the surface of the pump region. Therefore the in-plane losses $\kappa_\alpha^\|$ of mode α are proportional to the group velocity of this mode.

The above analysis of the photonic bandstructure and associated gain coefficients and total loss rates suggest that for our hexagonal photonic crystal laser structures, we obtain several modes with low-lying laser thresholds (see Fig. 2). Zero group-velocity modes with low out-of-plane losses occur for 0°-incidence angle (Γ-point) as well as for $\approx 3°$-incidence angle. While the former mode occurs in square [5, 6] and hexagonal lattices, we find through extensive numerical studies [11] that the latter mode occurs only in hexagonal lattices.

This prediction is confirmed by the emission pattern we obtain when pumping our structure above the lasing threshold, using a frequency doubled regeneratively amplified mode-locked Ti:sapphire laser. Figure 4 shows the laser emission from the described organic photonic crystal laser with hexagonal symmetry. Besides a central beam normal to the surface, we observe a six-fold symmetric emission pattern whose individual beams appear at an angle of $\approx 4.5°$ away from the normal which is in reasonable agreement with the predicted results.

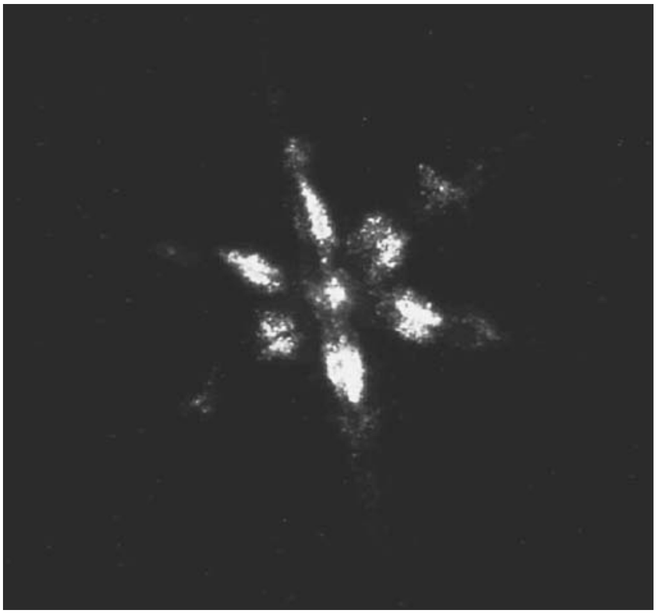

FIGURE 4 Far-field emission pattern of the laser emission from an organic PC laser with hexagonal symmetry. The individual beams of the sixfold symmetric emission pattern emerge from the sample at an angle of $\approx 4.5°$ away from the normal to the surface. The direction of the central beam is along the normal

In summary, we have carried out a comprehensive theoretical and experimental study of two-dimensional organic Photonic Crystal lasers. We have calculated the quasi-guided modes and their associated loss rates through a rigorous electromagnetic analysis. Using semiclassical laser theory for photonic crystals, we have been able to determine the low-threshold lasing modes. In hexagonal lattices, this theory predicts that simultaneous lasing of two distinct types of modes should occur. While lasing modes emitting normal to the surface have already been observed in square lattice geometries, hexagonal lattice feature a second, unique type of lasing mode along the ΓK-direction in reciprocal space, leading to off-normal emission with six-fold rotational symmetry. The theoretical predictions of bandstructure and low-threshold lasing modes are qualitatively in excellent agreement with corresponding measurement of the photo-luminescence and lasing action.

ACKNOWLEDGEMENTS We acknowledge the support of the Deutsche Forschungsgemeinschaft (DFG) within the Priority Programme SP 1113 "Photonische Kristalle" (projects Bu 1107/3-1, Bu 1107/3-2, Lu 204/15-1, Lu 204/15-2, Le 878/6-1 and Le 878/6-2). The research of M.D. and K.B. was further supported by the DFG-Center for Functional Nanostructures (CFN) at the University of Karlsruhe within project A 1.2.

REFERENCES

1 O. Painter, R.K. Lee, A. Scherer, A. Yariv, J. D. O'Brien, P.D. Dapkus, I. Kim, Science **284**, 1819 (1999)
2 N. Vats, S. John, Phys. Rev. A **58**, 4168 (1998)
3 M. Meier, A. Mekis, A. Dodabalapur, A.A. Timko, R.E. Slusher, J.D. Joannopoulos, O. Nalamasu, Appl. Phys. Lett. **74**, 7 (1999)
4 S. Imada, S. Noda, A. Chutinan, T. Tokuda, M. Murata, G. Sasaki, Appl. Phys. Lett. **75**, 316 (1999)
5 S. Riechel, C. Kallinger, U. Lemmer, J. Feldmann, A. Gombert, V. Wittwer, U. Scherf, Appl. Phys. Lett. **77**, 2310 (2000)
6 G.A. Turnbull, P. Andrew, W.L. Barnes, I.D.W. Samuel, Phys. Rev. B **67**, 165 107 (2003)
7 A. Haugeneder, M. Neges, C. Kallinger, W. Spirkl, U. Lemmer, J. Feldmann, M.C. Amann, U. Scherf, J. Appl. Phys. **85**, 1124 (1999)
8 D.M. Whittaker, I.S. Culshaw, Phys. Rev. B **60**, 2610 (1999)
9 V.N. Astratov, D.M. Whittaker, I.S. Culshaw, R.M. Stevenson, M.S. Skolnick, T.F. Krauss, R.M. De La Rue, Phys. Rev. B **60**, R16 255 (1999)
10 L. Florescu, K. Busch, S. John, J. Opt. Soc. Am. B **19**, 2215 (2002)
11 K. Forberich, Ph.D. thesis, Universität Freiburg (2005)