Low threshold edge emitting polymer distributed feedback laser based on a square lattice

A. E. Vasdekis, G. A. Turnbull, and I. D. W. Samuel[a)]
Organic Semiconductor Centre and Ultrafast Photonics Collaboration, School of Physics and Astronomy, University of St. Andrews, St. Andrews, Fife KY16 9SS, United Kingdom

P. Andrew and W. L. Barnes
Thin Film Photonics Group, School of Physics, University of Exeter, Stocker Road, Exeter EX4 4QL, United Kingdom

(Received 9 November 2004; accepted 21 February 2005; published online 11 April 2005)

We report the demonstration of a low-threshold, edge-emitting polymer distributed feedback laser based on a square lattice. The lattice constant was 268 nm, which corresponds to a lattice line spacing in the ΓM symmetry direction of the Brillouin zone of 189 nm. The latter was employed to provide feedback at 630 nm via a first order diffraction process. The device operated on two longitudinal modes, which were situated on the band-edge near the M symmetry point. The two modes had thresholds of 0.66 nJ and 1.2 nJ—significantly lower than comparable surface-emitting DFB lasers. Angle dependent photoluminescence experiments were performed to investigate the effect of the square lattice on the laser operation and the origin of the low threshold. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1898430]

Conjugated polymers have attracted considerable interest during the past decade because they possess both functional photophysical properties and simple device fabrication. Their strong absorption and broadband emission are two characteristics that make them ideal candidates for solid-state lasers in the visible spectral range.[1,2] To this end, multiple resonator configurations have been suggested, such as microcavities,[3] microdisk lasers,[4] and distributed feedback (DFB) structures.[5–10] The latter are particularly attractive as they combine low threshold operation and well defined output beam, with the possibility of simple fabrication.[5]

A generic schematic of a DFB polymer laser is shown in Fig. 1(a). A corrugated conjugated polymer acts as both gain medium and resonator, and is optically pumped. The operation of such a structure is based on the periodic modulation of the refractive index, which is responsible for the formation of a standing wave (Bloch mode[11]) through interference effects in the plane of the polymer film. The mode is additionally confined in the plane of the propagation direction via total internal reflection. The refractive index periodicity can either be one or two-dimensional, resulting in single or multiple resonance axes, respectively. Doubly periodic organic lasers have been investigated, employing either a hexagonal[6] or square lattice.[7] The advantages of the two-dimensional periodicity are higher gain and more coherent output modes.

To date, polymer lasers based on square feedback lattices have employed an optical periodicity (lattice constant multiplied by the effective refractive index of the optical mode) equal to the lasing wavelength, so that feedback is provided by second order diffraction from the grating. These structures operate at the Γ point of the Brillouin zone and have the advantage of being surface emitting, though this simultaneously imposes extensive scattering losses stemming from coupling to free space radiation. In this letter, we show that a different symmetry of the Brillouin zone of a square lattice can be used, producing an edge-emitting laser with lower threshold.

For the case of a square lattice, there are two mirror lines defined by the Miller indices (10) and (11), oriented at an angle $\Phi = 45°$ with respect to each other [Fig. 1(b)]. In the corresponding Fourier space representation there are two reciprocal lattice vectors, each of which could satisfy the Laue condition, and thus provide feedback for a particular incident wavevector.[12] In Fig. 1(b) the two vectors are denoted as $\mathbf{G_1}$ and $\mathbf{G_2}$ and point along the two symmetries of the Brillouin

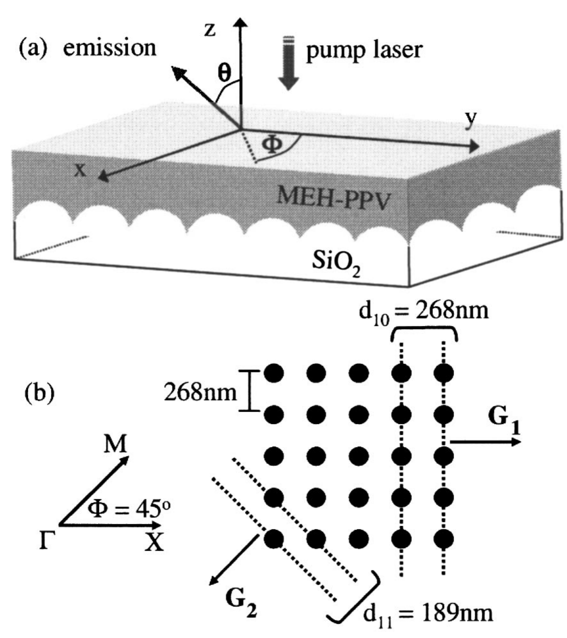

FIG. 1. (a) Cross-sectional view of a 2D DFB polymer laser; the angles θ (polar angle to the z axis) and Φ (azimuthal angle in the x-y plane) correspond to the direction of emission and the symmetries of the Brillouin zone (the ΓX for $\Phi=0°$ and the ΓM for $\Phi=45°$) respectively; (b) the real space lattice of the square diffraction grating. The mirror planes (10) and (11) are illustrated, along with the grating vectors ($\mathbf{G_1}, \mathbf{G_2}$) for the two symmetries of the Brillouin zone.

[a)]Electronic mail: idws@st-and.ac.uk

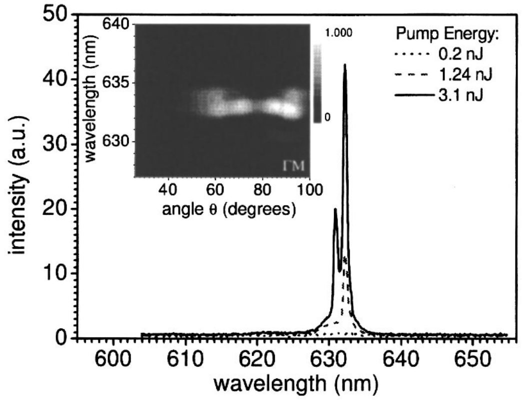

FIG. 2. Output spectra of the polymer laser above and below threshold. Inset shows a contour plot of the emission's wavelength vs the angle θ measured in the plane $\Phi=45°$.

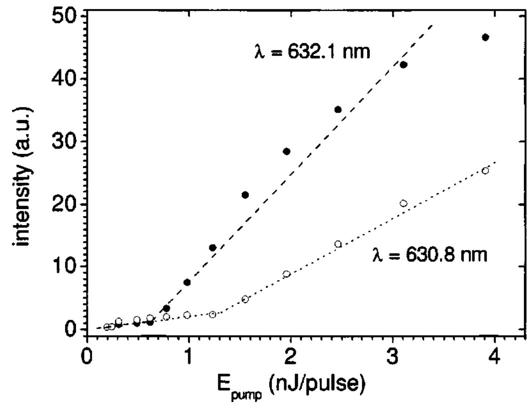

FIG. 3. Laser output as a function of pump energy for the two longitudinal modes

zone ΓX and ΓM. In addition, the modulus of the reciprocal lattice vectors is inversely proportional to the interplane spacings and hence the wavelength of the supported mode will be different for the two resonance axes.

In the current work, the substrate grating was defined holographically in a photoresist and then dry etched into the silica.[8] It had a depth of ~60 nm and periodicities of 268 nm in the ΓX direction (d_{10}) and hence 189 nm in the ΓM (d_{11}). The latter was employed to provide distributed feedback for a lasing mode in the red part of the spectrum, via a first order diffraction process. The optical gain medium was a thin film (200 nm thick) of the conjugated polymer poly[2-methoxy-5-(2′-ethylhexyloxy)-1,4-phenylene vinylene] (MEH-PPV) that was spin coated on the bigrating. This structure supported only the fundamental transverse electric mode in the spectral region of the polymer gain. The polymer laser was pumped optically using a frequency-doubled, passively Q-switched $Nd:YVO_4$ microchip laser that produced 1 ns pulses. The beam was focused to a spot of ~25 μm radius and the output of the polymer laser was measured using a fiber-coupled CCD spectrometer with a resolution of 0.7 nm. In order to avoid photo-oxidation of the polymer film, the sample was held under a vacuum of 10^{-4} mbar during operation.

In Fig. 2, the output spectra as a function of the pump energy are shown. Two modes reached threshold in this device, at 630.8 and 632.1 nm. The threshold and full width at half-maximum for the longer wavelength mode were 0.66 nJ and 0.74 nm respectively, while for the shorter wavelength mode these values were 1.2 nJ and 0.83 nm (Fig. 3). The laser emission was measured parallel to the ΓM symmetry direction ($\Phi=45°$) at $E_{pump}=1$ nJ and, as shown in the inset of Fig. 2, was found to be most intense at $\theta=90°$. The beam was highly divergent, which is typical for edge emission from thin films. We attribute the non uniform intensity variation with the angle θ to the poor edge quality of the device.

The threshold of this device was lower than the value of 4 nJ reported for a surface emitting polymer laser based on MEH-PPV, where a 65% deeper square lattice grating was used with a period of 409 nm.[9] Two factors lead to the lower threshold operation of the current device. The first is the increased Q-factor of the resonant modes that is associated with the fulfillment of the Bragg condition below the light line[13] and the second is the smaller spot size of the pump light used in this experiment.

The two lasing modes correspond to different Bloch modes that satisfy the Bragg condition for a lattice constant of 189 nm (ΓM). They differ in spatial localization with respect to the grating planes and hence are characterized by different effective refractive indices.[14] The asymmetry in gain and loss, associated with the different spatial distributions of the optical field in each mode, is responsible for their different thresholds. The lower wavelength mode has a lower effective refractive index as it is localized on a lower refractive index region, suffering both from intrinsically poorer waveguiding and less amplification (see Fig. 4 for the experimental dispersion diagram). The observation of two-wavelength operation is different from other two-dimensional, vertical-emitting polymer DFB lasers, in which all the spatial harmonics of the lasing mode are coupled inside the air cone. In such lasers, there is strong discrimination between the two band-edge modes arising from different radiation losses and coupling strengths to free space radiation.[15] Our observation of two-mode oscillation shows that the mechanism of strong mode discrimination is not present when operating below the light line (feedback occurring for in-plane wave vectors larger than ω_{laser}/c).[8]

In order to probe further the optical properties of the device, the spontaneous emission spectra were measured as a function of the angle of diffraction [θ in Fig. 1(a)], an experiment that has been shown to reveal information about the propagation conditions and scattering losses of a mode that propagates in the periodic structure.[8] In essence, due to momentum conservation in the plane of the film, the angle of

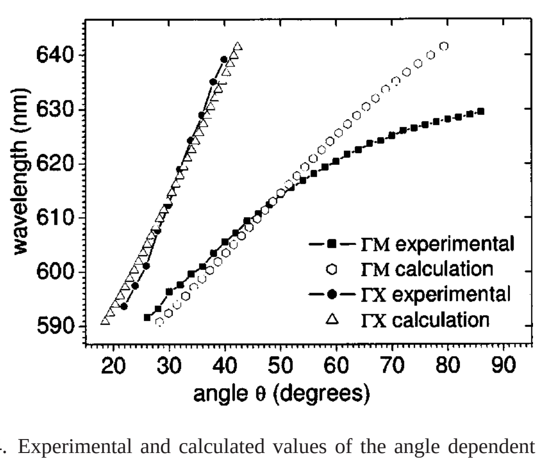

FIG. 4. Experimental and calculated values of the angle dependent photoluminescence measurements.

diffraction corresponds to the wave vector of a guided mode scattered above the light line. The spontaneous emission was generated by pumping the MEH-PPV film with the second harmonic of a continuous wave Nd:YAG laser and the symmetries of the Brillouin zone were determined by means of the polarization state of the emitted light.

The experimental results (Fig. 4) across the ΓM symmetry were collected at an angle $\Phi = 45°$ and showed that the lasing wavelengths are situated at the edge of a stop-band close to the M point. The stop-band manifests itself via the decrease of the rate of change of the energy of the mode (λ) with its wave vector (θ), which is essentially the group velocity of the propagating mode. Furthermore, the laser resonance, which is supported by the grating vector $\mathbf{G_2}$, is in a spectral range that corresponds to an angle of $\theta = 90°$ and hence is edge coupled as was the case for the stimulated emission. For comparison, the dispersion relationship across the ΓX direction ($\Phi = 0°$) was measured to find that the group velocity is constant within this frequency region. We also observed a weak output at $\theta = 35°$ in the spectral region of the laser resonances due to a mode propagating at $\Phi = 45°$ being scattered by $\mathbf{G_1}$. The measurements were found to agree with the calculated scattering angles where we assumed that the corrugation does not modify the effective refractive indices of the propagating modes.[10] For this calculation, the refractive indices of the MEH-PPV film and the substrate were measured by variable angle spectroscopic ellipsometry.

In conclusion, we have demonstrated an edge emitting polymer laser with a low threshold that is attributed to a high Q-factor of the resonant modes. The decreased losses were associated with the operation below the light line. For the same reason, two-mode oscillation was observed. The angle dependent laser emission and photoluminescence experiments confirmed this, as in both measurements light emission at the lasing wavelengths was edge coupled. Finally, the photoluminescence experiment revealed that the grating planes normal to the ΓX symmetry direction contributed only to weak scattering losses, while the respective ones normal to the ΓM direction that provided the feedback were responsible for a stop band and a reduced group velocity in the spectral region of the laser wavelength.

Fruitful discussions with Professor T. F. Krauss are respectfully acknowledged. We are grateful to EPSRC for financial support and to Covion for the supply of the polymer MEH-PPV. The assistance of Dr. C. Yates with the ellipsometry measurements is also gratefully acknowledged.

[1] M. D. McGehee and A. J. Heeger, Adv. Mater. (Weinheim, Ger.) **12**, 1655 (2000).
[2] N. Tessler, Adv. Mater. (Weinheim, Ger.) **11**, 363 (1999).
[3] N. Tessler, G. J. Denton, and R. H. Friend, Nature (London) **382**, 695 (1996).
[4] M. Kuwata-Gonokami, R. H. Jordan, A. Dodabalapur, H. E. Katz, M. L. Schilling, and R. E. Shusher, Opt. Lett. **20**, 2093 (1995).
[5] J. R. Lawrence, P. Andrew, M. Buck, W. L. Barnes, G. A. Turnbull, and I. D. W. Samuel, Appl. Phys. Lett. **81**, 1955 (2002).
[6] M. Meier, A. Mekis, A. Dodabalapur, A. Timko, R. E. Slusher, J. D. Johannopoulos, and O. Nalamasu, Appl. Phys. Lett. **74**, 7 (1999).
[7] S. Riechel, U. Lemmer, J. Feldmann, S. Barley, A. G. Muck, W. Butting, A. Gombert, and V. Wittwer, Opt. Lett. **26**, 593 (2001).
[8] G. A. Turnbull, P. Andrew, W. L. Barnes, and I. D. W. Samuel, Phys. Rev. B **67**, 165107 (2003).
[9] G. A. Turnbull, P. Andrew, W. L. Barnes, and I. D. W. Samuel, Appl. Phys. Lett. **82**, 313 (2003).
[10] W. Holzer, A. Penzkofer, T. Pertsch, N. Danz, A. Brauer, E. B. Kley, H. Tillmann, C. Bader, and H.-H. Horhold, Appl. Phys. B: Lasers Opt. **74**, 333 (2002).
[11] For a review of photonic Bloch waves: P. St. J. Russell, T. A. Birks, F. D. Lloyd-Lucas, in *Confined Electrons and Photons*, edited by E. Burstein and C. Weisbuch (Plenum, New York, 1995).
[12] See, e.g., N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Thomas Learning, London, 1985).
[13] Y. Akahane, T. Asano, B-S. Song, and S. Noda, Nature (London) **425**, 944 (2003).
[14] H. Benisty, D. Labilloy, C. Weisbuch, C. J. M. Smith, T. F. Krauss, D. Cassagne, A. Beraud, and C. Jouanin, Appl. Phys. Lett. **76**, 532 (2000).
[15] R. F. Kazarinov and C. H. Henry, IEEE J. Quantum Electron. **21**, 144 (1984).