Monolithic polymer microcavity lasers with on-top evaporated dielectric mirrors

Luana Persano, Pompilio Del Carro, Elisa Mele, Roberto Cingolani, Dario Pisignano et al.

Citation: [Appl. Phys. Lett.](#) **88**, 121110 (2006); doi: 10.1063/1.2179611

View online: <http://dx.doi.org/10.1063/1.2179611>

View Table of Contents: <http://apl.aip.org/resource/1/APPLAB/v88/i12>

Published by the [American Institute of Physics](#).

Additional information on Appl. Phys. Lett.

Journal Homepage: <http://apl.aip.org/>

Journal Information: http://apl.aip.org/about/about_the_journal

Top downloads: http://apl.aip.org/features/most_downloaded

Information for Authors: <http://apl.aip.org/authors>

ADVERTISEMENT

The advertisement banner features a background of orange and yellow diagonal stripes. At the top, the "AIP Applied Physics Letters" logo is displayed in white. Below the logo, on the left, is a white envelope icon. To its right, the text "Accepting Submissions in Biophysics and Bio-Inspired Systems" is written in black. Further right, a white button with the text "Submit Today" in orange is shown. On the far right, the "AIP Publishing" logo is displayed in blue and yellow.

Monolithic polymer microcavity lasers with on-top evaporated dielectric mirrors

Luana Persano,^{a)} Pompilio Del Carro, Elisa Mele, Roberto Cingolani, and Dario Pisignano
 NNL, National Nanotechnology Laboratory of INFM-CNR, c/o Palazzine Garrisi,
 Università degli Studi di Lecce, via Arnesano, I-73100 Lecce, Italy

Margherita Zavelani-Rossi, Stefano Longhi, and Guglielmo Lanzani
 National Laboratory for Ultrafast and Utraintense Optical Science ULTRAS-INFM-CNR, Dipartimento di
 Fisica, Politecnico di Milano, P.zza L. Da Vinci 32, I-20133 Milano, Italy

(Received 31 August 2005; accepted 17 January 2006; published online 22 March 2006)

We report on a *monolithic* polymeric microcavity laser with *all dielectric mirrors* realized by low-temperature electron-beam evaporation. The vertical heterostructure was realized by 9.5 TiO_x/SiO_x pairs evaporated onto an active conjugated polymer, that was previously spincast onto the bottom distributed Bragg reflector (DBR). The cavity supports single-mode lasing at 509 nm, with a linewidth of 1.8 nm, and a lasing threshold of 84 μJ/cm². We also report on the emission properties of the polymer we used, investigated by a pump-probe technique. These results show that low-temperature electron-beam evaporation is a powerful and straightforward fabrication technique for molecular-based fully integrable microcavity resonators. © 2006 American Institute of Physics. [DOI: 10.1063/1.2179611]

Since the discovery of optical gain from conjugated molecular systems,^{1–7} a lot of effort has been directed toward the realization of competitive organic-based solid-state lasers,⁸ enabling a high degree of integration with plastic optoelectronics, and low-cost fabrication and processing. In particular, the interest in microcavity structures is motivated by their excellent lasing performances and by their highly scalable integration into optoelectronic circuits for the realization of multidimensional arrays,⁹ memories,¹⁰ photodiodes,¹¹ and sensors.¹² The development of monolithic architectures onto organic semiconductors would allow one to explore the use of matrix-addressable arrays of organic vertical cavity emitting lasers (VCSELs) for optical processing, imaging and parallel interconnection. The use of VCSEL arrays is expected to be a feasible solution to support the continuous growth of data rates, thanks to their double-side emission, low-cost scale manufacturing, and testing.¹³ The combination of the integration capability, exclusive of vertical surface-emitting devices, and of the monolithic manufacturing onto soft matter can be a keystone of the future photonic interconnects for board-to-board and chip-to-chip applications. In particular, distributed Bragg reflector (DBR) heterostructures provide high values of the reflectivity by alternating only a few of two different films with a high refractive index contrast.

Unfortunately, the direct deposition of dielectric mirrors onto organic materials, needed for DBR-based monolithic VCSELs, is not straightforward because the high temperature (*T*), usually >200 °C, needed for this processes causes: (i) Loss of adhesion of the organic compounds, driven above their softness transition, and (ii) strong deterioration of their optical properties, especially in the oxygen atmosphere normally employed for reactive oxide growths. For these reasons, to date, organic microcavities have been generally realized by evaporating a metallic mirror^{1–4} or by sandwiching

an organic film between two prerealized DBRs.^{14,15} Very few studies have been performed on DBR fabrication by alternative low-*T* approaches.^{16,17} In this work, we employed low-*T* reactive electron-beam deposition for realizing a monolithic organic laser, whose cavity is made by two high-reflective dielectric mirrors. The cavity supports single-mode lasing at 509 nm, with a linewidth [full width at half maximum (FWHM)] of 1.8 nm and a threshold of 84 μJ/cm². This indicates that low-*T* electron-beam evaporation is a powerful and straightforward fabrication technique for molecular-based fully integrable microcavity resonators.

The bottom DBR of our microcavity was evaporated by a Temescal Supersource 2 electron-beam gun system onto Corning quartz. The mirror (7.5 λ/4 pairs of TiO₂/SiO₂) exhibits a 158 nm wide stop band centered at 500 nm. TiO₂ and SiO₂ films were deposited in oxygen atmosphere at 260 °C from 99.9% purity sources (Leybold, Germany). The conjugated copolymer, poly[(9,9-dioctylfluorenyl-2,7-diyl)-co-(1,4-diphenylene-vinylene-2-methoxy-5-{2-ethylhexyloxy}-benzene)] (American Dye Source, Canada) with a thickness (*d*) of about 155 nm, was then spincast onto the bottom mirror, and the top DBR was finally evaporated directly on the polymer film. The top DBR was composed of 9.5 λ/4 pairs of TiO_x/SiO_x deposited by low-*T* (*T* < 70 °C) electron-beam process in oxygen pressure. The top mirror exhibits a stop band centered at 480 nm with a FWHM of 143 nm. An out-of-cavity reference film, having the same thickness as the cavity layer, was fabricated by spin-coating from the same polymer solution. The reference film and the microcavity were excited by the second harmonic of a Ti:Sapphire amplified laser which delivers 180 fs pulses centred at 785 nm. The pump beam was incident normally through the quartz substrate or the bottom mirror and was focused on the polymer to a circular 80 μm diameter spot. The light emission from the film and from the lasing cavity was collected by an optical multichannel analyzer with a resolution of ~1 nm.

^{a)}Also at: ISUFI, Department of Nanoscience, Università di Lecce, via Arnesano, I-73100 Lecce, Italy; author to whom correspondence should be addressed; electronic mail: luana.persano@unile.it

FIG. 1. Transmittance spectra of the bottom (continuous line) and the top (dashed line) DBR, and scheme of the microcavity structure.

The chamber temperature during the electron beam evaporation crucially affects the resulting oxide films.¹⁸ According to the Cauchy dispersion relation for the wavelength-dependent refractive index,¹⁹ we estimated for the evaporated transparent films of titanium oxide values of $n=2.22$ and 2.01 (at about 520 nm) upon high- T and low- T evaporation, respectively. The achieved values are consistent with a bioxygen content only for the high- T deposited film.²⁰ The transmittance spectra of the bottom and top DBRs are displayed in Fig. 1. Both mirrors reflect more than 99% and the stop band of the $\text{TiO}_2/\text{SiO}_2$ mirror is about 15 nm larger than that of the $\text{TiO}_x/\text{SiO}_x$ DBR. This difference is attributed to the lower n contrast of the dielectric materials grown under low- T conditions.²⁰

Figure 2(a) shows the normalized photoluminescence

FIG. 2. Normalized emission spectra of the reference film (a) and of the microcavity (b) for low pump fluences [$96 \mu\text{J}/\text{cm}^2$ in (a) and $52 \mu\text{J}/\text{cm}^2$ in (b)] (dashed lines) and for high pump fluences [$264 \mu\text{J}/\text{cm}^2$ in (a) and $204 \mu\text{J}/\text{cm}^2$ in (b)] (continuous lines). Inset: Normalized laser emission spectra obtained by two cavities with different cavity lengths.

(PL) emission spectra for the out-of-cavity reference film at low ($96 \mu\text{J}/\text{cm}^2$, dashed line) and at high ($264 \mu\text{J}/\text{cm}^2$, continuous line) pump fluence. At low excitation, the spectrum is broad with overlapping vibronic peaks [I, II, III in Fig. 2(a)], at excitation fluences higher than $148 \mu\text{J}/\text{cm}^2$ a narrowing due to amplified spontaneous emission (ASE) is observed. ASE peaks at 517 nm with a FWHM of ~ 16 nm. Figure 2(b) shows, in dashed line, the PL emission of the copolymer inside the cavity for a low pump fluence ($52 \mu\text{J}/\text{cm}^2$ incident on the polymer). The PL spectrum in this case is modulated by the cavity and shows a quite broad peak, centred at 509 nm, which does not correspond to the cavity mode because light was collected in very large cone angle (50°). Note that this peak is not due to ASE as it is obtained at a very low pump (lower than threshold for ASE), and its central wavelength is different from that obtained in the previous case. The second peak (at 552 nm) is out of the cavity stop band and it is simply due to the high transmission of the top mirror at this wavelength. At excitation fluences higher than $84 \mu\text{J}/\text{cm}^2$, the emission collapses in a highly resolved peak, centred at $\lambda_0=509$ nm, with a FWHM of 1.6 – 2 nm, corresponding to laser action [continuous line in Fig. 2(b)]. It is worth pointing out that the cavity provides a very good feedback and above threshold the emission is directional, showing a low divergence and a narrow spectrum. In addition, by changing the thickness of the active organic layers from 156 to 176 nm ($\Delta d=20$ nm), we could vary the laser emission from $\lambda=509$ nm to 518 nm ($\Delta\lambda=9$ nm) [inset of Fig. 2(b)]. This result also shows that we can easily exploit the whole region of gain of the copolymer.

The dependence of the FWHM linewidth and of the intensity of the emission on the pump fluence incident on the polymer are shown, respectively, in Figs. 3(a) and 3(b), for the reference film (empty circles) and for the microcavity (full circles). The film shows an emission spectrum with an ASE peak, for excitation fluences higher than $148 \mu\text{J}/\text{cm}^2$, with a FWHM of ~ 16 nm [Fig. 3(a)] and an input-output characteristic with a sublinear evolution [Fig. 3(b)] due to saturation. Differently, the input-output characteristic of the microcavity shows a clear threshold at a fluence of $84 \mu\text{J}/\text{cm}^2$, followed by a linear increase, as expected for laser action. Lasing is also confirmed by the behavior of the mode linewidth which shows a steep decrease by a factor of 10 at threshold. Note that the lasing threshold is 1.8 times lower than the pump fluence needed for the occurrence of ASE from the bare conjugated polymer film, thus confirming, once more, the effectiveness of the feedback mechanism induced by the monolithic microcavity. Above threshold, the laser emission has a defined polarization, with a ratio of ~ 3.5 between the component parallel to the pump laser and the perpendicular one.

At high excitation fluences, the FWHM of the laser emission increases as expected for pulsed pumped lasers in gain switching regime.²¹ In fact, as the pump fluence increases, the upper state population available for lasing action increases and the time required to deplete this state shortens, thus the output pulse duration decreases and, correspondingly, the spectral mode width increases. The temporal evolution and the mode width of the output pulse can be theoretically obtained by solving the laser rate equations, in the four-level space-independent approximation.²¹ In the calculations, we neglected the spontaneous decay from the upper laser level which takes place on a time scale much longer

FIG. 3. Reference film (empty circles) and microcavity (full circles) emission characteristics as a function of the pump fluence incident on the polymer: (a) FWHM mode width and (b) output intensity. The solid line in (a) is obtained by numerical simulation and the straight line in (b) is a linear fit to the input-output laser characteristic. Inset: Calculated time behavior of the laser pulses for different pump fluences ($160 \mu\text{J}/\text{cm}^2$ solid line and $660 \mu\text{J}/\text{cm}^2$ dashed line).

(tens of ps) than the output pulse duration (tens to hundreds of femtoseconds). The numerical results are in agreement with experimental data [Fig. 3(a)], except for the region of low pump powers where the measured FWHM is limited by the resolution of the spectrometer. The fitting has been obtained by assuming a cavity photon lifetime τ_c of 31 fs, yielding a cavity quality factor $Q=2\pi\tau_c c/\lambda_0 \approx 115$. The inset of Fig. 3(a) shows, as an example, the calculated time behavior of the laser output pulses at different excitation fluences, for a pump pulse centred at $t=0$.

The hypothesis of a long living upper laser level and the emission properties of the copolymer were investigated by pump-probe experiments on the reference film. Using the same excitation source as before and setting the probe at 510 nm, coincident with the laser emission, we measured the transient transmission changes $\Delta T/T$. The time evolution for low excitation fluences (where ASE does not take place) can be fitted by a monoexponential response with a time constant of ~ 16 ps (Fig. 4), which is much longer than the duration of the microcavity laser pulse as assumed in the rate equations. At long delays (greater than ~ 100 ps), a process of photoinduced absorption overcomes the gain (inset of Fig. 4). The pump-probe experiments allowed us also to estimate the gain cross section, at 510 nm, to be $3 \times 10^{-16} \text{ cm}^2$, even if this result probably underestimates the real value as a unit quantum efficiency for the emitting species was assumed. This value is among the best so far reported for organic active materials.²²

In conclusion, we demonstrated monolithic microcavity polymer lasers by direct electron-beam evaporation at low- T of high reflective DBR mirrors onto the active layer. The devices support laser emission around 509 nm with a threshold excitation fluence as low as $84 \mu\text{J}/\text{cm}^2$, and a peak line-width of ~ 1.8 nm. These results are of great importance to

FIG. 4. Time evolution of the normalized transient transmittance changes $\Delta T/T$ at a probe wavelength of 510 nm (circles) and best fit (solid line). Inset: $\Delta T/T$ on a longer time scale.

actually exploit both the potentialities of vertical surface-emitting devices for large-scale integration of optical circuits, and the flexibility and processing advantages of organic semiconductors.

- ¹N. Tessler, G. J. Denton, and R. H. Friend, *Nature (London)* **382**, 695 (1996); V. Bulović, V. G. Kozlov, V. B. Khalfin, and S. R. Forrest, *Science* **279**, 553 (1998).
- ²M. A. Díaz-García, F. Hide, B. J. Schwartz, M. D. McGehee, M. R. Andersson, and A. J. Heeger, *Appl. Phys. Lett.* **70**, 3191 (1997).
- ³T. Virgili, D. G. Lidzey, M. Grell, D. D. C. Bradley, S. Stagira, M. Zavelani-Rossi, and S. De Silvestri, *Appl. Phys. Lett.* **80**, 4088 (2002).
- ⁴X. Liu, D. Poitras, Y. Tao, and C. Py, *Can. J. Phys.* **82**, 481 (2004).
- ⁵S.-J. Park, I. H. Cho, and K.-W. Lee, *Thin Solid Films* **363**, 221 (2000).
- ⁶S. E. Burns, G. Denton, N. Tessler, M. A. Stevens, F. Cacialli, and R. H. Friend, *Opt. Mater. (Amsterdam, Neth.)* **9**, 18 (1998).
- ⁷D. Moses, *Appl. Phys. Lett.* **60**, 3215 (1992).
- ⁸V. G. Kozlov, V. Bulović, P. E. Burrows, and S. R. Forrest, *Nature (London)* **389**, 362 (1997); D. Pisignano, L. Persano, P. Visconti, R. Cingolani, G. Gigli, G. Barbarella, and L. Favaretto, *Appl. Phys. Lett.* **83**, 2545 (2001); D. Pisignano, M. Anni, G. Gigli, R. Cingolani, M. Zavelani-Rossi, G. Lanzani, G. Barbarella, and L. Favaretto, *ibid.* **81**, 3534 (2002); M. Berggren, A. Dodabalapur, R. E. Slusher, and Z. Bao, *Nature (London)* **389**, 466 (1997); N. Tessler, *Adv. Mater. (Weinheim, Ger.)* **11**, 363 (1999).
- ⁹T. Yoshikawa, H. Kosaka, K. Kurihara, M. Kajita, Y. Sugimoto, and K. Kasahara, *Appl. Phys. Lett.* **66**, 908 (1995).
- ¹⁰I. D. Nikolov, K. Kurihara, and K. Goto, *Nanotechnology* **14**, 946 (2003).
- ¹¹J. M. Lupton, R. Koeppe, J. G. Müller, J. Feldmann, U. Scherf, and U. Lemmer, *Adv. Mater. (Weinheim, Ger.)* **15**, 1471 (2003).
- ¹²J. Wang, S. T. Sanders, J. B. Jeffries, and R. K. Hanson, *Appl. Phys. B: Lasers Opt.* **72**, 865 (2001).
- ¹³Y. Liu, *IEEE Trans. Adv. Packag.* **25**, 43 (2002).
- ¹⁴T. Granlund, M. Theander, M. Berggren, M. Andersson, A. Ruzickas, V. Sundström, G. Björk, M. Granström, and O. Inganäs, *Chem. Phys. Lett.* **288**, 879 (1998).
- ¹⁵M. Theander, T. Granlund, D. M. Johanson, A. Ruzickas, V. Sundström, M. R. Andersson, and O. Inganäs, *Adv. Mater. (Weinheim, Ger.)* **13**, 323 (2001).
- ¹⁶B. Masanelli, A. Gagnaire, L. Berthelot, J. Tardy, and J. Joseph, *J. Appl. Phys.* **85**, 3032 (1999); Q. Song, L. Liu, T. Ling, L. Xu, and W. Wang, *Appl. Phys. Lett.* **82**, 2939 (2003); C. B. Poitras, M. Lipson, H. Du, Hui Du, M. A. Hahn, and T. D. Krauss, *ibid.* **82**, 4032 (2003).
- ¹⁷L. Persano, E. Mele, R. Cingolani, and D. Pisignano, *Appl. Phys. Lett.* **87**, 031103 (2005).
- ¹⁸S. Y. Kim, *Appl. Opt.* **35**, 6703 (1996).
- ¹⁹D. Poelman and P. F. Smet, *J. Phys. D* **36**, 1850 (2003).
- ²⁰L. Persano, R. Cingolani, and D. Pisignano, *J. Vac. Sci. Technol. B* **23**, 1654 (2005).
- ²¹O. Svelto, *Principles of Lasers* (Plenum, New York, 1998), Sec. 8.4.4.
- ²²M. Zavelani-Rossi, G. Lanzani, M. Anni, G. Gigli, R. Cingolani, G. Barbarella, and L. Favaretto, *Synth. Met.* **139**, 901 (2003).