Whispering-gallery-mode microring laser using a conjugated polymer

Y. Kawabe,[a] Ch. Spiegelberg, A. Schülzgen, M. F. Nabor, B. Kippelen, E. A. Mash,[b] P. M. Allemand,[c] M. Kuwata-Gonokami,[d] K. Takeda,[e] and N. Peyghambarian
Optical Sciences Center, The University of Arizona, Tucson, Arizona 85721

(Received 3 September 1997; accepted for publication 7 November 1997)

We observed laser emission in whispering gallery modes using a microring composed of a semiconducting polymer poly[2,5-bis-(2′-ethylhexyloxy)-*p*-phenylenevinylene coated on an etched fiber under transient and quasisteady-state pumping conditions. The threshold for laser oscillation was 1 mJ/cm^2 (0.1 MW/cm^2) and 30 μJ/cm^2 (300 MW/cm^2) for nanosecond and femtosecond excitation, respectively. The laser output showed superlinear dependence on the excitation energy above the threshold. The demonstration of lasing under quasisteady-state pumping shows the possibility to develop electrically pumped polymer lasers. © *1998 American Institute of Physics.* [S0003-6951(98)01802-6]

Microcavity lasers are of interest for both fundamental studies of cavity quantum electrodynamics and for applications as integrated optical elements because of their unconventional lasing characteristics caused by storing coupling between the optical field and the active medium. Several types of microcavities such as spheres, rings, disks, and Fabry–Perot cavities made by semiconductors, organic dye solutions, and dye-doped polymers have been reported.[1–7] In order to realize low threshold lasers, it is necessary to utilize a high Q cavity to confine the light in a gain region, and to also have a strong coupling ratio between field and matter. Whispering gallery modes (WGM) oscillations from microring cavities have the advantage that high Q values are easily obtained even in a very small mode volume and that the number of modes contributing to laser oscillations can be reduced.

Light-emitting semiconducting polymers have been extensively studied for their application in displays.[8,9] Large optical gain has been obtained by fs spectroscopy on films of poly-*p*-phenylenevinylene (PPV) derivatives.[10,11] These results suggest the possibility of developing microcavity lasers using conjugated polymers as active materials. Recently, line narrowing of the emission has been reported in optically pumped PPV derivatives.[12–16]

The combination of light-emitting polymers with optical fibers can lead to integrated laser cavities with a high Q value. Such structures are expected to play an important role in electrically injected devices. For the future realization of polymer lasers, it is important to optimize the cavity configuration to comply simultaneously with optical and electronic requirements.

In this letter, we show lasing action in WGM in a microring cavity made by a PPV derivative pumped by ns or fs laser pulses. We used poly[2,5-bis-2′-ethylhexyloxy)-*p*-phenylenevinylene] (BEH-PPV) as the laser medium.[17] The molecular weight was about 10^6. The fiber was etched by hydrofluoric acid to a typical diameter of 50–100 μm.[6] The polymer was coated on the surface of the etched glass of the optical fiber by dipping it in a BEH-PPV in xylene solution (20 g/ℓ). The thickness of the polymer layer was estimated to be less than 1 μm.

In the experiments conducted under ns pumping, the samples were excited by the second harmonic of a Q-switched Nd: yttrium aluminum garnet (YAG) laser focused by a cylindrical lens from the side of the fiber. The repetition rate was 2–10 Hz, the pulse width was ~10 ns, and pulse energies up to 10 μJ were used. For the fs experiments, a tunable amplified CPM laser system was used with an excitation wavelength of 555 nm and a pulse duration of 100 fs. This system provided pulses with up to 30 nJ energy at a repetition rate of 1 kHz. In both experiments, the excitation area was typically $\sim 100\times 100$ μm^2. The diameter of the fiber used for the ns experiment was 100 μm, and it was 56 μm for the fs experiment. Light output from the fiber was detected from the side at an angle of 90° relative to the direction of the pumping beam. The samples were held in a vacuum chamber (10 mTorr) to avoid degradation by irradiation and oxidation. Using a monochromator of 25 cm focal length, a spectral resolution of 0.2 nm could be achieved.

Low intensity fluoresce and lasing spectra under ns pumping are shown in Fig. 1. The intensity of the emission is normalized to the intensity of low intensity fluorescence. The fluorescence spectrum that was observed at an excitation of 0.03 μJ (0.3 mJ/cm^2) shows two bands with maxima around 590 and 625 nm. Laser emission from WGM oscillation occurred in the central part of the band at a longer wavelength for excitation of 0.5 μJ (5 mJ/cm^2). About 10 laser modes can be resolved with a separation of 1.5 nm. The width of each mode is about 0.5 nm, clearly larger than the resolution of our detection system. The inset of Fig. 1 shows an expanded region around the lasing peaks. The relatively large width of the modes is possibly due to the inhomogeneity of the coated polymer layer or to a reduction of Q caused by surface irregularities. The Q value is estimated to be 1200 from the spectra width of the laser modes. The highest intensity mode emerged at 627 nm at the threshold and shifted to

[a]Electronic mail: kawabe@ccit.arizona.edu
[b]Department of Chemistry, The University of Arizona, Tucson, AZ 85721.
[c]Donnelly Corporation, Advanced Technology Center, 4545 E. Ft Lowell Rd., Tucson, AZ 85712-1108.
[d]Department of Applied Physics, Faculty of Engineering, University of Tokyo, Bunkyo-ku, Tokyo 113 Japan; Also at Cooperative Excitation Project, ERATO, Japan Science and Technology Corporation, 3-2-1 Sakato, Takatsu-ku, Kawasaki 213-0012, Japan.
[e]Tsukuba Research Laboratory, Japan Synthetic Rubber Co. Ltd., Tsukuba, Ibaraki 305 Japan.

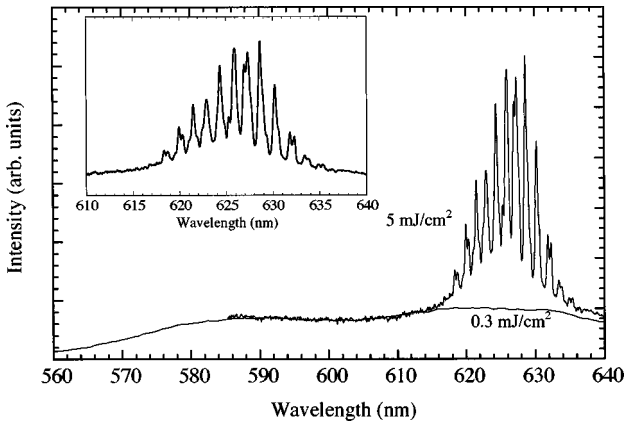

FIG. 1. Fluorescence and lasing spectra obtained by ns pumping. The fluoresce spectrum was observed that the excitation of 0.03 μJ (0.3 mJ/cm^2). Laser emission was obtained at 0.5 μJ (5 mJ/cm^2) excitation.

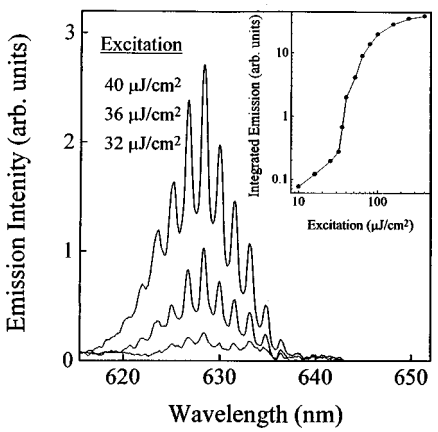

FIG. 3. Emission spectra obtained by fs pumping at three different excitation intensities. Inset: Emission intensity as a function of excitation.

623 nm for higher pump intensity. In gain measurements, a similar shift has been observed.[10,11]

The relation between pump fluence and output power showed superlinear dependence in the range where laser oscillation was observed as shown in Fig. 2. The exact lasing threshold was sensitive to the excitation configuration. The typical threshold pump fluence was 1–10 mJ/cm^2. These values are comparable to the reported values for dye-doped polymer WGM lasers,[6] and also correspond approximately to the threshold for line narrowing emission observed in PPV derivative films.[13,15,16] Saturation and decrease of the emission in the high excitation region were caused by the gain saturation and the degradation of the samples. In the fs experiment, very similar emission spectra were observed as shown in Fig. 3. In this case, the threshold of the lasing was about 30 μJ/cm^2. At the threshold, the emission intensity increased by more than one order of magnitude, and showed a much more pronounced threshold behavior than in the case of ns pumping.

The mode separation of WGMs can be estimated by the relation, $\Delta\lambda=\lambda^2/d\pi n_{\mathrm{eff}}$. Here, d is the diameter of the fiber, and n_{eff} is the effective refractive index of the oscillating modes. With $n_{\mathrm{eff}}=1.5$, the observed mode separations in the fs experiments agrees well with the calculated value of WGMs for a 56-μm-diam microring laser. Therefore, the observed modes are included in a series with the same mode order number in this case. Assuming a refractive index of 1.8

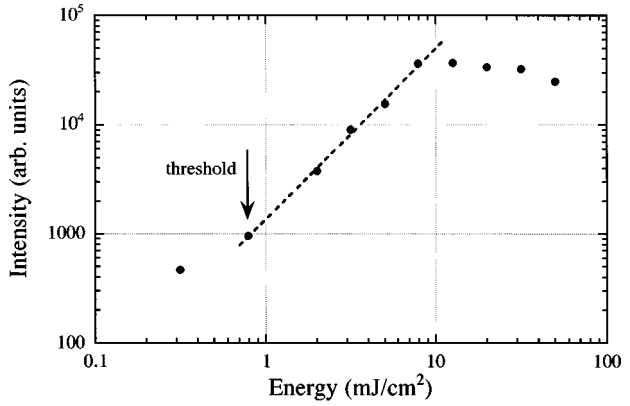

FIG. 2. Emission intensity as a function of excitation for ns pumping.

for the PPV derivative, coupling volume of the radiation field was mainly located in the glass fiber. It may be possible to reduce the lasing threshold by optimizing the coupling between the field and the polymer, i.e., by varying the thickness of the polymer layer. In the case of larger diameter microrings, the mode separation disagrees with the calculated value and substructures appear because of the oscillations due to higher order modes.[18]

The exciton lifetime of our BEH-PPV samples has been measured to be 50 ps.[10] The photon lifetime in the resonator estimated from the value of Q is of the order of 1 ps which is much smaller than the exciton lifetime. Thus, samples are under quasisteady-state pumping with ns pulses. On the other hand, transient behavior is expected for fs pumping. The threshold pump fluence for ns pumping was about two orders of magnitude higher than that for the fs pumping. However, the relevant parameter that has to be compared is the density of excited states. Under steady-state conditions, the density is proportional to the energy fluence times the ratio between lifetime and pulse duration, whereas in the transient regime the density is directly proportional to the energy fluence. Since lifetime to pulse length ratio for ns excitation is $\sim$0.005 which is comparable to the ratio of energy fluence between fs and ns, the threshold densities for laser oscillation are roughly equal within the experimental error for both regimes.

Considering the low carrier mobility of organics ($\sim 10^{-4}$ cm^2/Vs) and typical thickness of organic light emitting devices (100 nm) with operating voltages of 10 V, it takes approximately 100 ns to inject electronic carriers through the device. Therefore, it is important to demonstrate that lasing can be achieved with optical pulses in the ns regime. Thus, our results illustrate the possibility to develop polymer laser diodes. Another important parameter is the required current to obtain the exciton density equivalent to the threshold. If all photon are assumed to be absorbed in the polymer, the upper limit of the current is estimated to be $\sim$50 kA/cm^2 from the relation of $j=pe/\hbar\omega$. This estimate has some uncertainty due to the unknown film thickness of the polymer on the fibers. Here, j and p are current and light intensity, respectively, $\hbar\omega$ is a photon energy, and e is the charge of an electron. Our preliminary results showed that 0.3 kA/cm^2 is achievable by pulsed operation of a thin film

light emitting diode (LED) using the same polymer.[19] The gap between these two current densities for lasing may be reduced by optimizing the cavity quality, material processing, and layer structures.

In conclusion, we observed laser emission in whispering gallery modes from a semiconducting polymer PPV under transient and quasisteady-state pumping conditions. The threshold for laser oscillations is 1 mJ/cm^2 (0.1 MW/cm^2) and 30 μJ/cm^2 (300 MW/cm^2) for ns and fs pumping, respectively. The threshold exciton density is approximately the same in the two regimes. The lasing intensity showed superlinear dependence on the excitation energy above the threshold.

This work was supported by the US Office of Naval Research (ONR) through the MURI Center for Advanced Multifunctional Nonlinear Optical Polymers and Molecular Assemblies (CAMP).

[1] A. J. Campillo, J. D. Eversole, and H-B. Lin, Phys. Rev. Lett. **67**, 437 (1991).
[2] S. L. McCall, A. F. J. Levi, R. E. Slusher, S. J. Peartson, and R. A. Logan, Appl. Phys. Lett. **60**, 289 (1992).
[3] M. Kuwata-Gonokami, K. Takeda, H. Yasuda, and K. Ema, Jpn. J. Appl. Phys., Part 1 **31**, L99 (1992).
[4] J. C. Knight, H. S. T. Driver, R. J. Hutcheon, and G. N. Robertson, Opt. Lett. **17**, 1280 (1992).
[5] M. Osuge and K. Ujihara, J. Appl. Phys. **76**, 2588 (1994).
[6] M. Kuwata-Gonokami, R. H. Jordan, A. Dodabalapur, H. K. Katz, M. L. Schilling, R. E. Slusher, and S. Ozawa, Opt. Lett. **20**, 2093 (1995).
[7] J. P. Zhang, D. Y. Chu, S. L. Wu, S. T. Ho, W. G. Bi, C. W. Tu, and R. C. Tiberio, Phys. Rev. Lett. **75**, 2678 (1995).
[8] J. H. Burroughes, D. D. C. Bradley, A. R. Brown, R. N. Marks, K. Mackay, R. H. Friend, P. L. Burns, and A. B. Holms, Nature (London) **347**, 539 (1990).
[9] D. Braun and A. J. Heeger, Appl. Phys. Lett. **58**, 1982 (1991).
[10] A. Schülzgen, Ch. Speigelberg, M. M. Morrell, S. B. Mendes, P. M. Allemand, Y. Kawabe, M. Kuwata-Gonokami, S. Honkanen, M. Fallahi, B. Kippelen, and N. Peyghambarian (unpublished).
[11] Ch. Spiegelberg, A. Schülzgen, P. M. Allemand, B. Kippelen, and N. Peyghambarian (unpublished).
[12] N. Tessler, G. J. Denton, and R. H. Friend, Nature (London) **382**, 695 (1996).
[13] F. Hide, M. A. Diaz-Garcia, B. J. Schwartz, M. R. Anderson, Q. Pei, and A. J. Heeger, Science **237**, 1833 (1996).
[14] S. V. Frolov, W. Gellermann, M. Ozaki, K. Yoshino, and Z. V. Vardeny, Phys. Rev. Lett. **78**, 729 (1997).
[15] M. A. Diaz-Garcia, F. Hide, B. J. Schwartz, M. D. McGehee, M. R. Anderson, and A. J. Heeger, Appl. Phys. Lett. **70**, 3191 (1997).
[16] G. J. Denton, N. Tessler, M. A. Stevens, and R. H. Friend, Adv. Magn. Reson. **9**, 547 (1997).
[17] E. Harlev and F. Wudl (private communication).
[18] J. C. Knight, H. S. T. Driver, and G. N. Robertson, J. Opt. Soc. Am. B **11**, 2046 (1994).
[19] M. M. Morrell, B. Kippelen, and N. Peyghambarian (unpublished).